Petrological, mineralogical and geochemical peculiarities of Archaean cratons

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Abstract
The most outstanding features of Archaean cratons are their extraordinary thickness and enduring longevity. Seismically, Archaean cratonic fragments are sharply-bounded deep roots of mechanically strong, cold lithospheric mantle, clearly distinguishable from non-cratonic lithosphere. Rhenium-depletion of deep cratonic xenolith whole rocks and sulphide inclusions in diamond indicate that melting was broadly coeval with formation of the overlying proto-cratonic crust, which was of limited mechanical strength.

A very important process of proto-cratonic development was vertical crustal reorganisation that eventually yielded a thermally stable, cratonised crust with a highly K-U-Th-rich uppermost crust and much more depleted deeper crust. Clastic sedimentary rocks available for geochemical study are predominantly found in the youngest parts of supracrustal stratigraphies and over-represent the highly evolved rocks that appeared during cratonisation. Vertical crustal reorganisation was driven by crustal radiogenic heat and emplacement of proto-craton-wide, incubating and dense supracrustal mafic and ultramafic volcanic rocks. Statistical analysis of these cover sequences shows a preponderance of basalt and a high abundance of ultramafic lavas with a dearth of picrite. The ultramafic lavas can be grouped into Ti-enriched and Ti-depleted types and high pressure and temperature experimental data indicate that the latter formed from previously depleted mantle at temperatures in excess of 1700°C.

Most mantle harzburgite xenoliths from cratonic roots are highly refractory, containing very magnesian olivine and many have a high modal abundance of orthopyroxene.
High orthopyroxene mode is commonly attributed to metasomatic silica-enrichment or a non-
pyrolitic mantle source but much of the excess silica requirement disappears if melting
occurred at high pressures of 4-6 GPa. Analysis of experimental data demonstrates that
melting of previously depleted harzburgite can yield liquids with highly variable Si/Mg ratios
and low Al₂O₃ and FeO contents, as found in komatiites, and complementary high Cr/Al
residues. In many harzburgites, there is an intimate spatial association of garnet and spinel
with orthopyroxene, which indicates formation of the Al-phase by exsolution upon cooling
and decompression. New and published rare earth element (REE) data for garnet and
orthopyroxene show that garnet has inherited its sinusoidal REE pattern from the
orthopyroxene. The lack of middle-REE depletion in these refractory residues is consistent
with the lack of middle- over heavy-REE fractionation in most komatiites. This suggests that
such pyroxene or garnet (or precursor phases) were present during komatiite melting. In the
Kaapvaal craton, garnet exsolution upon significant cooling occurred as early as 3.2 Ga and
geoobarometry of diamond inclusions from ancient kimberlites also supports cool Archaean
cratonic geotherms. This requires that some mantle roots have extended to 300 to possibly
400 km and that early cratons must have been much larger than 500 km in diameter.

We maintain that the Archaean-Proterozoic boundary continues to be of geological
significance, despite the recognition that upper crustal chemistry, as sampled by sedimentary
rocks, became more evolved from ca. 3 Ga onwards. The boundary coincides with the
disappearance of widespread komatiite and marks the end of formation of typical refractory
cratonic lithosphere. This may signify a fundamental change in the thermal structure of the
mantle after which upwellings no longer resulted in very high temperature perturbations. One
school of thought is that the thermal re-ordering occurred at the core-mantle boundary
whereas others envisage Archaean plumes to have originated at the base of the upper mantle.
Here we speculate that Archaean cratonic roots may contain remnants of older domains of
non-convecting mantle. These domains are potential carriers of isotope anomalies and their
base could have constituted a mechanical and thermal boundary layer. Above laterally
extensive barriers, emerging proto-cratons were protected from the main mantle heat loss.
The eventual collapse of these mechanical barriers terminated very high temperature
upwellings and dismembered portions of the barrier were incorporated into the cratonic
mantle during the final Neoarchaean 'superplume' event. The surviving cratons may therefore
preserve biased evidence of geological processes that operated during the Archaean.
1. Introduction

The word *kratogen* was first used in the German geological literature to distinguish stable continental crust from orogenic crust that had been involved in mountain-forming processes. The term derives from the Greek word *krátos*, meaning strength or power, and was later shortened to *Kraton* before being accepted into English usage as *craton*. There is wide agreement that a craton represents a strong, thick and coherent piece of continental lithosphere that has maintained long-term tectonic stability and traditionally, cratons have been viewed as nuclei of continents. Before the availability of vast quantities of seismic data and their computational analysis, cratons were geologically and topographically recognised as forming flat shield areas comprised of exposed crystalline basement or stable platform sediment successions, typically of early Precambrian age.

Global seismic tomography is one powerful method for differentiating the roots of cratons because the mantle portion of cratons is characterised by very fast travel speeds of seismic waves. The seismic architecture of the lithosphere and surrounding asthenosphere can be defined by comparing the SH body-wave velocity to a standard model (e.g. Grand, 1994). Cratonic lithosphere roots below most exposed Archaean shields have an average SH body-wave velocity 2-3% faster than reference models (e.g. Begg et al., 2009) whereas Proterozoic lithospheric mantle is characterised by SH body-wave velocity only 1% faster (e.g. Lebedev et al., 2009). Faster travel speeds are primarily the result of substantially lower temperature than adjacent convecting asthenosphere and only secondarily the result of compositional difference (e.g. Griffin, 1999), the maximum likely contribution of which being 1/4 of the actual observed velocity difference (Schutt and Lesher, 2006).

In their analysis of the lithospheric structure of the African continent, Begg et al. (2009) demonstrated that it is possible to clearly differentiate and visualise the Archaean cratonic mantle roots from the rest of the African continent. Figure 1 (annotated and reproduced from Begg et al.’s (2009) figure 10) shows a view from the deep mantle transition zone in the southern Atlantic, looking upward in a NE direction towards Africa. The volumes of red material (showing SH body-wave velocity >1.9%) clearly delineate the sharply bounded roots of the three broad Archaean cratons within Africa: West Africa, Congo (including Tanzania), and southern Africa (mostly the Kalahari craton).

On the global shear speed structure maps of the upper mantle of Schaeffer and Lebedev (2013), most cratonic mantle roots are seen extending to at least 150 km depth (their figure 11) with deviations of SH body-wave velocity as high as +6%, requiring ambient
temperature of less than 1,000°C. Because density of mantle peridotite is an inverse function
derelative to oceanic asthenosphere due to their extremely refractory chemistry, essentially
exhausted of spinel and clinopyroxene, depleted in garnet and rich in low density Mg-rich
olivine (e.g. Schutt and Lesher, 2006). Thus, the buoyancy of cratons and the fast SH body-
wave velocity in cratonic mantle roots together are very strong evidence for extreme (40% or
more) melt depletion in Archaean cratonic roots.

A very pleasing completely independent confirmation of this geophysical picture of
Archaean cratonic mantle roots has emerged from petrological and geochemical study of
mantle xenoliths. Multiple lines of evidence show that deep cratonic xenoliths, representing
samples from between ca. 80 and 250 km root depth (e.g. Kopylova and Caro, 2004), mostly
experienced very high degrees of melt extraction. Perhaps the best line of evidence is the
composition of the predominant peridotite phase olivine, whose Mg content is most strongly
influenced by the extent of melt depletion (e.g. Baker and Stolper, 1994). Compared to
olivines from oceanic, abyssal peridotites with average forsterite contents of 90.5 to 91.0%,
those in ancient cratonic harzburgites are much more magnesian, averaging between 92.5 and
93.0% forsterite component (e.g. Pearson and Wittig, 2008). Available experimental data
indicate that these compositions require that the host peridotites experienced extraction of 35-
45% liquid (see Bernstein et al, 2007 and Pearson and Wittig, 2008) through repeated melting
events.

After such extensive extraction of melt, the residue ends up vastly depleted in
incompatible elements. Because Re is much more incompatible than Os, ancient harzburgitic
peridotites develop retarded, very unradiogenic Os-isotope compositions (e.g. Walker et al.,
1989) that can be used to calculate a Re-depletion age. When initially discovered, this
demonstrated that at least for the Kaapvaal craton, melt extraction must have occurred prior
to 2 Ga. In more recent studies, peridotitic sulphide inclusions in diamonds were analysed
and these have yielded even older Re-depletion ages. For example: 3.1 to 3.5 Ga for the
Siberian craton (Pearson et al., 1999); 3.4 Ga for the southern Zimbabwe craton (Smith et al.,
2009); and ca. 3.3 Ga for the Kaapvaal craton (e.g. Shirey and Richardson, 2011). In the
Slave craton, Aulbach et al. (2004) retrieved monosulphide solution inclusions with
Mesoarchaean Re-depletion ages from peridotitic olivine xenocrysts. A subset of the
analysed inclusions form a 3.4 Ga isochron, attesting to the extreme antiquity of the deep
residual lithosphere there. Finally, Archaean mafic to ultramafic intrusions from the
Zimbabwe craton contain extremely unradiogenic Os that Naegler et al. (1997) interpreted to
indicate the beginning of cratonic root depletion from 3.8 Ga onwards, coincident with the oldest known zircon crystals known from this craton (Bolhar et al., 2017).

In summary, the overwhelming petrological evidence from harzburgite xenoliths combined with Re-Os isotope systematics indicates ancient melt extraction events consistent with the presently cool and refractory nature of Archaean cratonic roots inferred from seismology. The lithospheric mantle root of some cratons was subsequently lost or eroded (e.g. Miller et al., 2012) and has in many places been modified (e.g. Griffin et al., 2003) and cannot be directly compared with the root that was in place during the Archaean.

Complementary observations are made on the proto-cratonic surface where Archaean mafic rocks are clearly much more magnesian than those erupted after 2.5 Ga (e.g. Keller and Schoene, 2012). Albeit not the volumetrically dominant component of greenstone belts (e.g. Sproule et al., 2002), komatiite with between ca. 20-35 wt% MgO is known from most cratons and where greenstone belt stratigraphy is well understood (e.g. from komatiite-related Ni-ore exploration; Lesher and Barnes, 2009), and multiple episodes of komatiite emplacement over periods lasting ca. 50 Ma are evident (e.g. Thurston et al., 2008). There is an on-going debate regarding the type of melting that caused komatiite formation, ranging from dry, very hot and deep ‘plume’ type melting (Berry et al., 2008) to aqueous fluid fluxing in (Parman et al., 1997) and away from subduction zones (Sobolev et al., 2016) with intermediate models envisaging more complex fluid fluxing (Herzberg, 2016). Regardless of the melting mechanism, komatiite requires advanced stages of peridotite melting that consumed olivine and orthopyroxene (Nesbitt et al., 1979), an observation that at least qualitatively complements the highly refractory nature of cratonic harzburgite. The strong agreement about the general (albeit not total) disappearance of komatiites at the end of the Archaean is also consistent with the fact that, on average, Archaean cratonic roots have faster SH body-wave velocities (e.g. Schaeffer and Lebedev, 2013) than Proterozoic mantle lithosphere and that Archaean harzburgites are much more strongly depleted than Proterozoic equivalents (e.g. Griffin et al., 2003).

The relatively sudden (in the context of the vast Precambrian time span) demise of komatiite production at the Archaean-Proterozoic (A-P) boundary was accompanied by other notable disruptions in secular trends, which collectively may have relevance for understanding how Archaean continental lithosphere differed from younger continents. These include: the evolution of the maximum MgO content of highly magnesian magmas, which remained almost constant throughout the Archaean but then dropped (Campbell and Griffiths, 1992); several general changes in the nature of mafic and felsic magmas emplaced towards
the end of the Archaean (Keller and Schoene, 2012); pronounced changes in the composition
of clastic (Condie, 1993) and hydrogenous sediments (Konhauser et al., 2009) across the A-P
boundary; a jump in the O-isotope composition of zircon (Spencer et al., 2017); and a sharp
shift in triple-O-isotope composition of clastic sedimentary rocks (Bindeman et al., 2018).
Condie (1993) demonstrated that the Archaean land surface had a much higher abundance of
komatiite and basalt than would be estimated from outcrop areas at the present-day exposure
level. Archaean clastic sedimentary rocks have very high relative abundance of Co, Ni and Cr
whereas Proterozoic sedimentary rocks do not significantly differ from Phanerozoic
equivalents (Figs. 2a,b). Most recently, Tang et al. (2016) used the Ni/Co ratio in elastic
sedimentary rocks as a proxy for MgO content with which it was possible to also show the
dramatic transition from mafic to felsic upper continental crust (UCC) during the
Neoarchaean (see Fig. 2c).

Notwithstanding the harmonious overall picture of Archaean continental build-up and
mantle melting, it turns out that the reconciliation of the seismic information from cratonic
mantle roots with petrology of Archaean harzburgite and komatiite fails in terms of persistent
deviation of mineralogical and geochemical detail from model prediction (e.g. Walter, 1998;
Bédard, 2006; Pearson and Wittig, 2008). Furthermore, current models for Archaean craton
formation struggle to achieve mass balance and have limited success explaining the co-
ocurrence of komatiite with the more dominant tholeiitic basalts. There is the additional
issue that Archaean cratonic mantle roots were apparently already sufficiently cold to permit
diamond formation (Boyd et al., 1985; Ballard and Pollack, 1988; Kaminsky et al., 2002;
Miller et al., 2012) yet for the most part, the Archaean crust lacked mechanical strength and
experienced repeated re-melting events. Thus, the Archaean continental lithosphere
apparently existed as a proto-cratonic composite of a strong deep mantle root with a weak
crustal lid before complete lithospheric stabilisation (i.e. cratonisation) that typically, but not
exclusively, happened towards the end of the Archaean.

Unless these discrepancies and apparent mutual inconsistencies can be resolved, there
is limited hope that the full geological and tectonic significance of Archaean cratons can be
reconstructed. The aims of this paper are to review discrepancies that have long been known
within the specialist disciplines for a wider geochemical readership, to synthesise mutual
inconsistencies and to provide perspectives on possible common solutions.
2. Brief review of some pertinent problematic aspects of Archaean mantle melting and lithospheric mantle mineralogy

2.1 Incompatible element mass balance

Jordan (1975, 1978) developed pioneering models for cratonic stabilisation, in which he first clearly demonstrated the need for depleting the mantle portion of stable shield lithosphere through basalt extraction into the crust. The resulting root possesses the required physical characteristics to make it more refractory, less dense and rheologically distinct from the surrounding asthenosphere. Since these early models, many lines of evidence have emerged supporting the view that the continental crust and the depleted mantle portion of cratonic lithosphere share a very long common history, including melt depletion events coeval with crust formation pulses (e.g. Pearson et al., 1995; Naegler et al., 1997). However, the elegance of the basalt extraction hypothesis is compromised by the problem of mass balance.

The issue is that the preserved cratonic mantle root alone cannot nearly have supplied all the incompatible trace elements present in the Archaean crust (e.g. Arndt et al., 2002; Carlson et al., 2005) as can be illustrated with the example of the highly incompatible radioactive heat producers U, Th and K. In average continental crust these are 62±2 times enriched over primitive mantle. This ratio is quite precisely known from heat flow data and continental composition estimates (Rudnick and Gao, 2003), primitive mantle U and Th concentrations and terrestrial Pb-isotope composition (Sun and McDonough, 1989), and the canonical Earth K/U ratio (Jochum et al., 1983). The enrichment ratio is somewhat lower (45±2) for average continental crust of Archaean age as the crustal column generates a lower average modern heat flow of 41 mW m⁻² on cratonic provinces (Michaut et al., 2009).

Nevertheless, a 165 km deep cratonic mantle root (yielding a 200 km deep craton) only contains 5.8 times the mass of a 35 km thick crust, not the required 45±2 times. This means that unless the precursor mantle was much more enriched than primitive mantle, the undifferentiated rock mass contained in a craton could only have provided ca. 15-20% of the inventory of the very highly incompatible elements.

Therefore, Archaean crust formation must also have tapped into magmas that originated from the convecting asthenospheric mantle (Arndt et al., 2002). Such magmas could have travelled through the growing Archaean continental lithosphere (e.g. Carlson et al.,
Regardless, the discussion of Archean mantle source regions must distinguish between the ultra-depleted cratonic roots and the more modestly depleted convecting asthenosphere.

2.2 Mismatch between predicted and observed mineral modes in cratonic harzburgites

At relatively low pressures (1-3 GPa), melting experiments of fertile mantle compositions predict that the residue after 25-30% melt extraction should be devoid of clinopyroxene and Al-phase (spinel and/or garnet) and consist of >80% olivine (Baker and Stolper, 1994; Walter, 1998) and that at 40% degrees of melting, the residue should be nearly mono-mineralic dunite. At higher pressures, garnet can remain in the residue at up to 40-50% melt extraction. Because the Fo-content of olivine increases approximately linearly with the extent of melt extraction (Pearson and Wittig, 2008), a strong correlation between modal olivine content and Fo-component is expected. This relationship was prominently described for oceanic peridotites by Boyd (1989) and is shown in figure 3a. Boyd (1989) next compared this oceanic trend with data from post-Archean subcontinental lithosphere brought to the surface by off-craton kimberlites. The comparison confirmed that, to a first order (Fig. 3b), the data followed the oceanic trend. However, harzburgites from the Archean Kaapvaal craton (Fig. 3c) were found to deviate markedly from the trend, plotting at higher Fo values (92-93%) but containing only 55 to 80% modal olivine instead of the expected dunitic abundance (>90%). Essentially, the harzburgites are much richer in orthopyroxene than expected and the resulting rock has an unusually high Si/Mg ratio (as enstatite is the most Si-rich of the common mantle minerals).

This mismatch between observed and predicted mineralogy is very strongly expressed in Kaapvaal harzburgites but it is also found in many other Archean cratonic harzburgites with <60% modal olivine, for example from the Slave, Siberian and Wyoming cratons (e.g. Boyd et al., 1997; Eggler et al., 1987; Ionov et al., 2010; Kopylova et al., 1999; Kopylova and Caro, 2004; Newton et al., 2015; Regier et al, 2018; Solovjeva et al., 1994). Xenolith populations of only few kimberlites, particularly from the North Atlantic craton (Greenland; e.g. Bizzarro and Stevenson, 2003; Bernstein et al., 2006), are dominated by dunite as expected from Boyd's (1989) trend, but with generally high Fo-component. On average, high orthopyroxene abundance is more common and has been very widely discussed in the
literature (e.g. Bernstein et al., 2007) without finding wide petrological consensus. Proposed explanations include: the high Si/Mg ratio was inherited from the precursor mantle that may have differed from pyrolite due to crystal fractionation in a magma ocean (Herzberg et al., 1988); the high modal abundance of orthopyroxene reflects melt extraction at much greater depth (>8 GPa) than experienced by oceanic harzburgites (Canil, 1992; Herzberg, 1993); density sorting in plume melt residues (Arndt et al., 2002); later addition of Si, possibly from subduction zone metasomatism (Kesson and Ringwood, 1989) or serpentinisation (Canil and Lee, 2009); late Proterozoic within plate melt infiltration of hydrous melt (Wasch et al., 2009); reaction between depleted harzburgite either with foundering eclogite restite (Bédard, 2006) or eclogite-derived melts (Aulbach et al., 2011); and originally oceanic origin at a shallow very hot ridge (Rollinson, 2010). As can be appreciated, these various scenarios have substantially different ramifications for the early Earth.

A second unexpected feature of Archaean cratonic harzburgite is that it commonly contains garnet. High pressure experiments (e.g. Takahashi, 1986; Canil, 1991; Walter, 1998) show that at pressures exceeding 6 GPa, minor residual garnet can persist in harzburgite even after 40% melt extraction. However, it appears that garnet is not a rare accessory but a common phase in the Archaean cratonic mantle cross-section (Griffin et al., 1999; 2002) and this is at odds with the widely-held model that the refractory lithosphere grew by orogenic stacking of residues from low pressure melting (e.g. Bernstein et al., 2007; Canil, 2004; Rollinson, 2010; Lee and Chin, 2014). Indeed, Nixon et al. (1987) demonstrated that the common occurrence of garnets (with a characteristic sub-calcic chemistry) was restricted to xenocrysts and garnet-bearing xenoliths from kimberlites that intruded through Archaean cratonic lithosphere (Kaapvaal and Zimbabwe) whereas off-craton kimberlites erupted through Proterozoic geology were devoid of such garnets (see Fig. 4). It is possible that the overabundance of orthopyroxene and the dominance of garnet-bearing harzburgite are related through exsolution of garnet from orthopyroxene (e.g. Cox et al., 1987) and/or that garnet grew in response to metasomatism (e.g. Stachel, et al., 1998; Pearson and Wittig, 2008). Regardless, Sm/Nd and Lu/Hf isotope data show that garnet growth in many cratons occurred during the Archaean (e.g. Shu et al. 2013; Koorneef et al., 2017).

2.3 Seismic evidence for garnet in Archaean lithosphere and lithospheric buoyancy constraints
In the early long-range seismic refraction profiles, such as those conducted across continental North America (e.g. Green and Hales, 1968), it was noticed that there is an increase in impedance at depths between 60-80 km. Because impedance is the product of seismic velocity and density and since temperature is unlikely to decrease with depth, Hales (1969) proposed that this observation represented the spinel to garnet transformation. Many seismic studies have since found similar increases in impedance (e.g. Revenaugh and Jordan, 1991, Woods et al., 1991) but the observation is not generally accepted to reflect the spinel to garnet transition globally, although the observed extent of velocity change fits well with experimental constraints on the phase transition (Webb and Wood, 1986).

Whereas the detection of the impedance increase can reasonably be expected in the more fertile oceanic mantle, where it occurs near the modelled depth of 50-60 km, Revenaugh and Jordan (1991) commented that in continental lithosphere, basalt depletion should strongly diminish the strength of the reflector by reducing modal abundances of spinel and garnet. This should be particularly true for cratonic lithosphere where multiple lines of evidence suggest extensive melt extraction. However, it appears that the impedance increase is just as prominent in the lithosphere of several Archaean cratons, including the Baltic Shield, Western Australia and the Congo craton (Lebedev et al., 2009). Indeed, in the original work by Hales (1969) the strongest velocity increase was found in the Lake Superior region, underlain by Archaean lithosphere. In comparison with the oceanic realm, the increase in seismic velocity in cratonic regions occurs from the Moho down to depths of 100-150 km and is less sharp. Lebedev et al. (2009) explained this with the survival of spinel to greater depth due to increase in the Cr$_2$O$_3$ activity in the refractory residual harzburgite. Experimental data support stability of spinel to greater depth in Cr-rich bulk compositions, but these studies (e.g. Klemme, 2004; Ziberna et al., 2013; Ziberna and Klemme, 2016) also predict very low overall modal spinel abundances. Thus, an additional explanation for the increase of the shear wave velocity below cratons could be the appearance of ultra-depleted, sub-calcic and calcic garnet, which is often found to have equilibrium pressures in the region of 3-5 GPa (e.g. Gibson et al., 2013; Gibson, 2017), corresponding to depths of ca. 90-150 km. The 80-90 km range is also the depth for which Griffin et al. (2002) report the shallowest appearance of Cr-rich garnet macrocrysts in a number of Archaean cratons. Regardless of the importance of spinel, the seismic evidence apparently supports the empirical petrological evidence for garnet being an important constituent phase of many Archaean cratonic roots.

Although cratonic lithosphere is commonly thought of as being buoyant, this is only true in terms of composition but due to its much lower temperature than asthenosphere, the
cratonic root is, on average, only barely positively buoyant (e.g. Rudnick, 1994). In fact, Schutt and Lesher (2006) calculated that residual cratonic peridotite formed above 110 km may be negatively buoyant by the time it has cooled to the cratonic geotherm. These authors suggested that this could imply an inherent tendency for relatively shallow depleted residues to undergo vertical downward motion. A final poorly understood aspect of cratonic buoyancy is that there is a lack of evidence for proto-cratons having ever stood substantially elevated above sea level during their formation (Galer and Mezger, 1998; Bindeman et al., 2018). This is surprising particularly for cratonic fragments that formed rapidly (e.g. many on the subprovinces of the Superior craton; e.g. Thurston et al., 2008). At the solidus, the cratonic root density becomes substantially lower than that of fertile mantle (e.g. Zibrona and Klemme, 2015) and this would buoy the craton, yet entire stratigraphies of basalt and komatiite were apparently emplaced in water (e.g. Houle et al., 2009) into environments with limited topography. This is also true for many of those demonstrably deposited ensialically through evolved continental crust (e.g. Chauvel et al., 1985, 1993; Blenkinsop et al., 1993). Many of these apparent inconsistencies may stem from the fact that in the Archaean, proto-cratons had not yet reached their mechanical rigidity, with much reduced strength in the middle and lower crust (Ashwal et al., 1987; Kramers et al., 2001; Benn and Kamber, 2009).

2.4 Experimental vs. empirical constraints on komatiite major element chemistry

At least since the experiments by Takahashi and Scarfe (1985) it has been clear that high degree, high temperature melting of fertile mantle at 150-200 km depth can produce liquids that are as magnesian as komatiite. Because komatiite is defined as an ultramafic volcanic rock with MgO >18 wt% and depleted in other oxides (e.g. Arndt et al. 2008), Takahashi and Scarfe (1985) were technically correct in relating their experimental melts to komatiite. However, there remain very significant issues with the widely held opinion that komatiites are very high degree melts. Arndt (1977a) pointed out that since mantle melts are mostly buoyant relative to refractory olivine, it would be physically impossible for early melts not to escape. Schmeling and Arndt (2017) showed that mantle melts are less dense than the main harzburgite phase olivine to a depth of 230 km, making it implausible that liquids would have pooled at 50-230 km source depths until 30-40% of melting. By contrast, it is possible to form liquids with >30 wt% MgO at modest degrees of melting from more refractory
peridotite that had previously been depleted or melting at greater depth (e.g. 14 GPa; Takahashi, 1986). At mantle transition zone depths, relative densities permit large degree melts to accumulate before eventual escape (Schmeling and Arndt, 2017).

There are also issues regarding the finer details of major element composition of experimental versus empirical komatiite compositions (e.g. Walter, 1998). Many of these problems may relate to the inability of small-scale experiments to simulate the natural process of melting in large-scale thermal upwellings (e.g. Robin-Popieul et al., 2012). Herzberg (2016) provided a recent discussion on Al-undepleted komatiite liquids. It was found that at a given parental MgO content the observed SiO$_2$ contents for classic Neoarchaean Abitibi greenstone belt komatiites are too low by comparison with experimental liquids, certainly outside analytical error. The diagrams from Herzberg (2016) most relevant to this mismatch are reproduced in figure 5. Relative to an accumulated fractional melting liquid with 30 wt% MgO, the observed liquid line of descent (LLD) has ca. 0.8 wt% too little SiO$_2$ (Fig. 5a). More significantly, in terms of FeO vs. MgO systematics, the empirical data favour a batch melt (Fig. 5b), which is ca. 1.7 wt% higher in SiO$_2$ at MgO 30 wt% than the empirical LLD (Fig. 5a). If these komatiites are representative, then either the melting models are in error, the source of komatiite was not pyrolitic, melting did not occur at dry conditions (Herzberg's (2016) preferred option), or a combination of several factors. Herzberg (2016) also explored to what extent the experimental liquids of Walter's (1998) experiments could match the empirical Neoarchaean Abitibi greenstone belt komatiites in MgO vs. SiO$_2$ space (his figure 3a). This comparison revealed that certain liquids from the highest pressure range runs (6 and 7 GPa) coincided with the empirical LLD but at MgO contents of 27-28 wt%, i.e. lower than the presumed 30 wt% MgO parental melt deduced from melt inclusions (Sobolev et al., 2016).

In addition to the MgO/SiO$_2$ mismatch, Walter (1998) commented on a paradox concerning the depth of melting. When plotting empirical data and experimental melting contours in CaO/Al$_2$O$_3$/ vs. Al$_2$O$_3$ space (in which olivine crystallisation causes predictable vertical LLD), Walter (1998) found (his Fig. 7a) that early Archaean komatiites could be generated at 8-10 GPa with up to 30% melting and that Neoarchaean komatiites at 7 GPa with 30 to >50% melting. However, these samples have flat chondrite-normalised rare earth element (REE) patterns, which apparently precluded garnet as a stable phase in the source. Walter (1998) commented (p. 49): "This creates a paradox"; in that major elements (CaO/Al$_2$O$_3$) require garnet on the solidus yet REE do not show middle (M) and light (L) enrichment over heavy (H) REE expected from residual garnet. In terms of Sm/Yb or Gd/Yb,
komatiites show a surprising lack of strong fractionation. Walter (1998) speculated that the mantle of at least some of the cratons (e.g. Kaapvaal) could have had a major and trace element composition different from pyrolite. The melting depth conundrum was further developed by Canil (2004), who pointed out that the high Cr/Al ratio of cratonic harzburgites was difficult to reconcile with melt extraction deeper than 3 GPa. One proposed solution was that the cratonic lithosphere grew by stacking and thickening.

Studying FeO/MgO systematics of cratonic peridotites, Lee and Chin (2014) proposed that most Archaean mafic magmas could be sourced by melting mantle between 2 and 4 GPa at temperatures of 1500 to 1650°C. Because nearly all their studied peridotites yielded higher equilibration pressures and lower equilibration temperatures, these authors also advocated vertical foundering and orogenic thickening as the key process for craton growth. However, Lee and Chin (2014) explicitly filtered against orthopyroxene-rich peridotite samples by (p. 277) "excluding those samples with anomalously high SiO$_2$ for a given Mg#$", on the basis that these were considered to implicitly reflect metasomatic Si-addition.

In summary, there are clearly recognisable patterns in most proposals that attempt to reconcile experimental with empirical petrological and seismic observations regarding Archaean subcontinental lithosphere formation. Many of these relate to the minerals orthopyroxene and garnet both of which are apparently more abundant in Archaean cratonic lithosphere than available experimental constraints permit. Whereas many ancient cratonic harzburgites are too rich in SiO$_2$ for their MgO content or Mg#, many komatiites seem to suffer from the opposite problem. Finally, while major element systematics require garnet in the source of many komatiites, their REE patterns do not show the expected deficit in HREE.

The main aim of this paper is to explore whether there could be a common solution to the issues of Archaean craton formation.

3. Datasets, analysis and samples

3.1 Datasets and data analysis

We revisited four datasets relevant to Archaean craton formation. Firstly, we reanalysed major and compatible trace elements systematics of Archaean igneous rocks previously studied by Keller and Schoene (2012) and Kamber (2015). Specifically, we used the pre-compiled files from the GEOROC database for the following cratons: Baltic shield; Dharwar craton; Kaapvaal craton; North Atlantic craton; North China craton; Slave craton; Superior craton; Tanzania craton, Western Australian cratons; and Zimbabwe craton. These were
selected to include cratons for which geochemical and petrological datasets also exist for peridotites, because they host classic komatiite exposures and for which statistically significant datasets exist. Each dataset was filtered to only include extrusive igneous rocks for which whole rock analyses are reported. The analysis of the distribution of MgO and Ni contents of Archaean volcanic rocks was conducted with kernel density estimation (KDE), which is a robust method to make statistical inferences on population distributions of a finite data sample. The method lends itself to estimate the number of modes in a population (Silverman, 1981) and is commonly used to analyse detrital U/Pb zircon and other age populations (e.g. Petrus et al., 2016). The programme used in this study was DensityPlotter (Vermeesch, 2012).

A second dataset that was revisited separately is that for komatiites and komatiitic basalts from the Neoarchaean Abitibi and Pontiac Subprovinces (Superior Province) reported by Sproule et al. (2002). This is a very comprehensive dataset, including analyses for the classic Pyke Hill and Alexo samples. Representative analyses were tabulated in Sproule et al. (2002) and only these are found in GEOROC. Here we used the full dataset (Mike Lesher, pers. comm. 2017) to test whether, within one greenstone belt, temporal trends existed in terms of Mg/Si systematics (e.g. Herzberg, 2016).

Global deep-time geochemistry datasets for shale are a rich source of information regarding the evolution of the uppermost crust (e.g. Smit and Mezger, 2017; Bindemann et al., 2018). Here we revisited the shale geochemistry compilation reported by Tang et al. (2016) in their supplemental materials S1 and added data for Neoarchaean argillite (Barrie, 2005), which is a very fine-grained sedimentary rock containing reduced C. This rock type is extensively studied by exploration geochemists due to its utility in vectoring volcanogenic massive sulphide (VMS) deposits but these data rarely find their way into peer-reviewed publications.

The final datasets revisited are the experimental ultramafic liquid compositions and modal abundances reported in Walter (1998) and Takahashi et al. (1993). Additional highly valuable experiments have been conducted (e.g. Takahashi and Scarfe, 1985; Takahashi, 1986; Canil, 1991; 1992; Takahashi et al., 1993; Baker and Stolper, 1994; Longhi, 2002; Schutt and Lesher; 2006) but those conducted at high pressure did not report full modal abundances of reaction products. We compared their phase relationships to those of Walter (1998) and Takahashi et al. (1993) to test for consistency and representativeness.
3.2 Samples

As explained in the introduction and preceding section, the minerals orthopyroxene and garnet hold a critical role in Archaean craton formation (e.g. Gibson, 2017). In 2015 and 2017, we collected cratonic peridotites from the Bultfontein pans, Kimberley, Republic of South Africa (28.739155°S, 24.818094°E) with the aim of better documenting the mutual phase relations (e.g. Cox et al., 1987; Saltzer et al., 2001) between garnet and orthopyroxene and to better determine their interesting trace element geochemistry (Wasch et al., 2009). Here we report data for two representative samples. Sample BP002 is dominated by a spectacular orthopyroxene megacryst of which a single crystal fragment measuring 15 x 12 x 7 cm is preserved within a granular garnet harzburgite. Tomlinson et al. (2018) reported a crystallographic and geochemical study for this sample. The other sample is a coarse-grained garnet harzburgite (17BSK051) with a high modal abundance of orthopyroxene typical of harzburgites from the Kimberley pipes (e.g. Boyd, 1989; Grégoire et al., 2003; Simon et al., 2007; Wasch et al., 2009). Analytical details can be found in the Electronic Supplement.

4. Results

4.1 The composition of the uppermost Archaean continental crust

Three prominent studies have investigated the temporal evolution of the composition of the continental crust with large lithogeochemistry compilation datasets (Keller and Schoene, 2012; Tang et al., 2016; Smit and Mezger, 2017). They have in common that they analysed time series of averaged geochemical parameters (e.g. MgO content, Ni/Co, Cr/Zn and Cr/U) of lithological groups such as mafic and felsic igneous and sedimentary rocks. By contrast, the purpose of the analysis performed here was to study the modal distribution of volcanic rocks that were erupted onto the various cratons in the Archaean.

Figure 6 shows histograms for MgO wt% distribution in volcanic rocks on 6 cratons for which >500 datapoints are available in GEOROC (panels A-F) and a combined histogram for the Zimbabwe, Tanzania, Slave and North China cratons (panel G) for which there are smaller datasets. For each dataset, MgO contents were re-calculated to a volatile-free basis for those samples (>90%) for which full major element data and loss on ignition are reported. The metadata are available in Table_S1. Data for the few (<1%) rocks with >50 wt% MgO were excluded from further analysis on account of clearly being cumulates. Superimposed onto the histograms are KDE curves (blue solid lines). Sizes of sample populations,
histogram bin widths and fitting bandwidths (which determine KDE sensitivity) are listed in Table 1.

A qualitative look at the histograms and the KDEs reveals a common pattern. There is a prominent population of evolved felsic volcanic rocks with MgO between 0.7 and 2 wt% and then a very prominent population with MgO between 5 and 10 wt%. These are separated by a strong gap. A compositional gap at similar MgO is well known in ocean island rocks (Chayes, 1963) and arc systems (e.g. Reubi and Blundy, 2009) where it has been attributed to differentiation and re-melting within the crust.

In all cases, the 5 to 10 wt% MgO basaltic population contains the largest number of samples. Moving towards higher MgO contents, the next common pattern is the paucity of samples with MgO in the 15 to 20 wt% range. Beyond 18 wt% MgO, all cratons show one or two broad populations of ultramafic volcanic rocks. With the exception of the Baltic shield data, where a population around 25 wt% MgO is quite prominent, clear ultramafic populations are not easily obvious. One reason it is difficult to test whether there is a dominant population in a linear histogram of MgO is that the ultramafic samples are naturally distributed over a large number of bins. This is because in an ultramafic parent rich in MgO, relatively limited olivine and pyroxene removal or accumulation will cause rapid change in MgO of several wt%, i.e. several histogram bins. By contrast, the same extent of crystal fractionation from a much lower initial MgO content in a felsic or basaltic magma will only spread the data across one or two bins. This is better appreciated when the data are subjected to a logarithmic translation of the x-axis values. The translation results in unevenly sized bin widths and separates data with low MgO content into more bins. By contrast, the ultramafic rocks are binned into fewer categories. The resulting histogram and KDE for the combined data from all cratons considered are shown on figure 6H. This clearly exposes the gap in erupted volcanic rocks with MgO between 15 and 20 wt% and emphasises the ultramafic population centred around ca. 28 wt% MgO. To our knowledge, this has not explicitly been reported before but has been hypothesised to exist based on MgO vs. siderophile element distributions by Arndt (1991).

The KDE method can also be used to statistically estimate the number of modes in a population (Silverman, 1981) but the method depends on the errors of each measurement. Because GEOROC does not report precision and accuracy of individual data, reasonable errors were assigned to data as follows: from 0.1-1.0±1; >1.0-5.0±0.2; >5.0±0.5 wt% MgO. When applied to the combined dataset from all cratons (see Fig. 7a), this method finds four prominent populations: one felsic with 1.0 wt% MgO accounting for 18.1% of the data; one
dominant basaltic with 6.0 wt% MgO containing 50.6% of the data; and two ultramafic at 21.2 and 35.7 wt% MgO, respectively, collectively accounting for 31.3% of the data. Similar calculations were performed for all individual datasets displayed on figure 6 (panels A-G) and results are listed in table 1. The preferred number of populations varied between three and five but in all cases, there is a felsic population at 1.2±0.59 wt% accounting for 18.6±7.0% of the data and a dominant basaltic population at 6.3±1.4 wt% MgO containing roughly half of the data (47.9±7.6). The next important population is ultramafic with 28.3±2.4 wt% MgO making up 21.9±4.1% of the data. It is found in all datasets but the North Atlantic craton. A high-Mg basalt population with 14.7±1.8 wt% is found on four cratons and on these it collectively contains 18.8±4.6% of the data. Finally, a highly ultramafic population with 39.0±3.8 MgO wt% (likely dominated by olivine cumulate rocks) is also found on four cratons, there containing 9.5±2.9% of the data.

It is not the intention to ascribe specific petrologic significance to these statistical analyses but the study of the histograms shows that the dominant Archaean volcanic rock was basalt and the second-most important volcanic rock type was ultramafic. The broadly komatiitic rocks are almost certainly over-represented in global datasets due to the understandable scientific interest in these rocks. However, the important finding of the paucity of picritic compositions (reflected in the absence of an identified population in the 15-20 wt% MgO range) cannot be an artefact of overly keen interest in komatiite, which would be difficult to distinguish from altered picrite or komatiitic basalt in the field or the core shed. Finally, the analysis also shows that Archaean basalt had nearly indistinguishable average MgO content and distribution as during the Phanerozoic (e.g. Kamber, 2015).

Because Ni content increases strongly with degree of melting and hence Mg content (Arndt, 1977b), the Ni concentrations of volcanic rocks of the same cratons was also compiled. Many published Ni analyses are performed by XRF on ignited powder and therefore concentrations were not corrected for loss on ignition. A total of 9,288 analyses were compiled (for metadata see Table_S1) and of these 37 had Ni concentration greater than 5,000 ppm and were not further considered. Due to the huge range in Ni concentrations (i.e. 0.1 to 5,000 ppm), the linear histograms show very little detail and instead a logarithmic translation was performed (Fig. 7b). This very clearly shows two dominant populations representing basalt with ca. 200 ppm Ni and komatiite with ca. 2,000 ppm Ni. The felsic population is spread over many bins with a minor peak at ca. 20 ppm. Thus, the Ni distribution shows a similar pattern as the MgO distribution, highlighting the dearth of picritic compositions.
The final analysis was a comparison between rocks 2.5 to 2.9 Ga in age and those emplaced earlier. This was motivated by the proposal that there was no sharp A-P boundary but that geology instead changed in a transitional fashion from ca. 3 Ga onwards (e.g. Shirey and Richardson, 2011). More recently, Tang et al. (2016) and Smit and Mezger (2017) suggested that the UCC had started to become more felsic from 2.9 Ga onwards (see Fig. 2c) prompting an interest to study the chemistry of volcanic rocks emplaced either side of 2.9 Ga. For this comparison, data from the Kaapvaal, Zimbabwe, Dharwar, Superior and Western Australian cratons were separated according to preferred age stated in the original publications. These cratons expose a higher proportion of Meso- and Palaeoarchaean rocks than most others.

The comparison shows, both in terms of MgO (Fig. 7c, d) and Ni (Fig. 7e, f), that there is no obvious distinction between the two age groups, suggesting that the basaltic mode always dominated and that an equally ultramafic komatiite mode persisted right through the Archaean Eon. This is consistent with the observation of time-series of maximum inferred MgO contents of komatiites, showing no clear trend towards lower MgO with decreasing age (e.g. Campbell and Griffiths, 1992; Kamber, 2010). There is a small indication that picrite has become somewhat more common since 2.9 Ga but we note that this could be an artefact of the lower density of data for the >2.9 Ga age group.

4.2 Comparison of compositions of komatiites with predictions of melting models

4.2.1 A comparison of komatiite parental melt composition with experimental liquids and calculated melts of previously depleted mantle

It is widely argued that many observed komatiite liquid compositions are not easily reconciled with experimental liquids, calling for a variety of processes, such as fluid-induced melting (e.g. Herzberg, 2016) or ultra-deep melt segregation (e.g. Robin-Popieul et al., 2012) to explain the discrepancies. Komatiites are traditionally classified into Al-depleted and Al-undepleted based on CaO/Al$_2$O$_3$ and Al$_2$O$_3$/TiO$_2$ systematics (Nesbitt et al., 1979). In partly serpentinised samples, Al$_2$O$_3$/TiO$_2$ is more robust and samples with values <15 are classed as Al-depleted, whereas those with 15> Al$_2$O$_3$/TiO$_2$ <25 are termed Al-undepleted. Rarer samples with Al$_2$O$_3$/TiO$_2$ >25 are termed Ti-depleted. In view of the importance of the
Al₂O₃/TiO₂ ratio, we revisited the data from the Takahashi et al. (1993) and Walter (1998) experiments relating to the Al₂O₃ and TiO₂ systematics of experimental liquids as a function of melt fraction. These datasets provided chemical analyses for all experimental phases as well as modal abundance estimates. Importantly, their phase relations are compatible with more experiments that do not report modal abundance.

The first thing to note is that whereas Al-uptake into the melt is complex (Fig. 8a), Ti essentially behaves as an incompatible element with strong initial enrichment in low degree melts (Fig. 8b), regardless of pressure (see also Longhi, 2002). As expected for an incompatible element, at all degrees of partial melting, the Ti content in the liquid is higher than in the starting lherzolite. The two highest melt fraction liquids of Takahashi et al. (1993) are an apparent exception to this trend (Fig. 8b) but this is likely an artefact of rounding low concentration values to too few significant digits in the published data table. Regardless, when the Al₂O₃/TiO₂ ratio is plotted against Al₂O₃ (Fig. 8c) and TiO₂ (Fig. 8d) it is evident that TiO₂ exerts dominant control over the ratio (see also Robin-Popieul et al., 2012). The important inference arising from the experimental data is that an originally lherzolitic mantle source that became partially depleted (e.g. via extraction of 10-20% melt) will have a substantially higher Al₂O₃/TiO₂ ratio than primitive mantle (ca. 22) or the N-MORB source (ca. 25). If such a source was later remelted, it would yield liquids with very low Ti contents (<0.2 wt%) and very high Al₂O₃/TiO₂ ratios.

There is currently a lack of experimental komatiitic compositions with the very low TiO₂ concentrations found in some natural komatiites (e.g. Wilson, 2003). This very likely stems from a lack of successful experiments from refractory (harzburgitic) starting compositions. To gain a first-order understanding of what liquids from refractory mantle could be we calculated the differential melt compositions from isobaric series of the Walter (1998) and Takahashi et al. (1993) experiments assuming mass conservation (Table_S2). The validity of these calculations depends on the quality of the chemical data for the two batch melt liquids, the accuracy of the melt fraction estimate, and the stability of the phases at the liquidus. The results show (Fig. 8e) that all the hypothetical melts from variably refractory mantle have higher Al₂O₃/TiO₂ than the starting lherzolite. Hypothetical melts from very highly depleted mantle have very low TiO₂ concentrations, in many cases resulting in negative apparent TiO₂ values. These are artefacts of the cumulative uncertainties that could be removed if the Ti concentrations of high degree melts were analysed with dedicated long-acquisition time electron microprobe analyses (e.g. Donovan et al., 2011). Nevertheless, the low Ti concentrations (0.1-0.2 wt%) and the high Al₂O₃/TiO₂ ratios (25-200) compare well
with the ranges reported for Ti-depleted komatiites of the Abitibi greenstone belt (Sproule et al., 2002) and in so-called Al-enriched komatiites of the Barberton greenstone belt (e.g. Wilson, 2003; Robin-Popieul et al., 2012). Thus, at least some komatiites could have resulted from repeated 10-20% melting of a progressively more refractory source, overcoming the issue of requiring high-degree melts that would be too buoyant not to escape (Arndt 1977a; Schmeling and Arndt, 2017).

We adopted the same approach to explore whether melts from variably refractory peridotite could help to resolve the SiO₂ vs. MgO mismatch documented by Herzberg (2016). Figure 8f shows three examples for 3, 4.6 and 7 GPa. The 3 GPa example is the composition of the melt required to take the 24% melt (experiment 30.14 of Walter, 1998) to 53% (experiment 30.11). It has 33.6 wt% of MgO and 45.7 wt% of SiO₂. The 4.6 GPa example shows the composition of the melt required to take the 22% liquid (experiment 1800°C of Takahashi et al., 1993) to 55% (experiment 1800°C). The 7 GPa example is the composition of the melt required to take the 16% melt (experiment 70.07 of Walter, 1998) to 47% (experiment 70.09). As can be seen by comparing panels (G) and (H) of figure 8, the hypothetical melts extend to higher MgO and more variable SiO₂ than the experimental liquids from fertile mantle and qualitatively illustrate that melting refractory peridotite could be one potential explanation for the Si/Mg and Al/Ti systematics of Archaean komatiites and picrites. Takahashi (1986) suggested that the range in SiO₂ values could reflect the presence of absence of pyroxene at the liquidus but these findings require further experimental and petrological exploration.

4.2.2 An extended comparison of Abitibi and Pontiac Subprovince komatiites with melting experiments of fertile mantle

The Neoarchaean Abitibi greenstone belt in Ontario and Quebec is a classic Subprovince of the Superior craton in which komatiites and mafic meta-volcanic rocks have been studied extensively. Because the greenstone belt apparently did not form onto pre-existing evolved crust, most of its igneous rocks preserve least contaminated mantle radiogenic isotope signatures (e.g. Ketchum et al., 2008) and are considered ‘juvenile’. An outstanding feature of the Abitibi greenstone belt is that across the nearly 700 km width of exposure, it displays a coherent vertical stratigraphy (e.g. Thurston et al., 2002) constrained with a very large number of high-precision U/Pb zircon dates from felsic volcanic rocks (e.g. Corfu, 1993). The huge wealth of information pertaining to the stratigraphic relationships is not widely
appreciated by the academic community, probably because most of the data support mineral exploration and are published in reports of geological survey organisations. Berger et al. (2011) explained that high-precision U/Pb zircon dates allowed the 55 rock assemblages recognised in 1991 to be simplified into the 9 assemblages shown in figure 9. The new stratigraphy comprises 7 assemblages dominated by volcanic rocks and an unconformably overlying set of two sedimentary rock assemblages. The 7 assemblages dominated by volcanic rocks are separated by condensed horizons (Thurston et al., 2008) composed of iron formations, chert and minor fine-grained siliciclastic sedimentary rocks (argillites).

The quality of stratigraphic control allows for an accurate assessment of petrological and potential temporal variety of komatiite within a greenstone belt. Komatiite occurs in four of the 7 lower assemblages (Fig. 9). The most widely known komatiites (Pyke Hill and Alexo) belong to the Kidd-Munro assemblage and have been extensively studied. Analysing major element systematics of these two occurrences, Herzberg (2016) established that batch melting produces a closer fit to observed MgO vs. FeO systematics than accumulated fractional melting (Fig. 5b) and furthermore, that compared to model predictions, the empirical data show a deficit in SiO$_2$ relative to MgO.

Here we revisit the much larger dataset of Sproule et al. (2002) who interrogated major and trace element geochemistry of 2,466 chemical analyses of high-Mg basalt, picrite and komatiite. Their sample suite includes representatives from all four ultramafic occurrences within the Abitibi greenstone belt as well as two younger occurrences in adjacent greenstone belts to the south (Pontiac Subprovince). They are age equivalent to the Blake and Timiskaming assemblages of the Abitibi greenstone belt and their chronologic positions are shown in figure 9. To achieve their wide stratigraphic and geographic coverage, Sproule et al. (2002) also analysed variably altered samples. After using the excessive loss on ignition criterion of Sproule et al. (2002) to avoid samples with strongly modified major element chemistry, 2,048 analyses remained.

We first tested whether this dataset also showed the relative dearth of picritic samples with MgO between 15 and 20 wt% by plotting the anhydrous rock MgO content in a histogram (Fig. 10a). As can be seen, the dominant population of data occurs around an MgO content of ca. 28 wt% with another prominent mode at ca. 40 wt% MgO, probably dominated by olivine-rich cumulates. The basaltic mode is not within the range of compositions studied by Sproule et al. (2002) whose dataset excluded most samples with MgO <12 wt%. However, the paucity of samples with MgO of 15-20 wt% is nonetheless clearly visible, confirming the picture from the preceding global analysis (Fig. 7).
In the context of this treatment, the most interesting phenomenon of the Abitibi greenstone belt ultramafic rocks is their great chemical variability. If the conventional komatiite classification criteria are applied to all rocks in Sproule et al. (2002) dataset (not just komatiites), 76% classify as Al-undepleted, 17% as Al-depleted and 7% as Ti-depleted types. Panels (B) and (C) of figure 10 show similar FeO vs. MgO and SiO$_2$ vs. MgO diagrams as figure 5. In this much larger dataset, it is evident that for any given MgO value there is a wide range in FeO (4-5 wt%) and SiO$_2$ (5-7 wt%) content. Contamination with continental crust and alteration are unlikely to explain the full spread in FeO vs. MgO space. Some of the most Fe-rich samples may contain excess accumulated chromite with or without olivine removal (see also Fig. 4b of Sproule et al., 2002) but even allowing for this process, a wide range in Mg# remains. The FeO, SiO$_2$ and MgO systematics do not correlate in any way with the extent of Al- and Ti-depletion. In SiO$_2$ vs. MgO space (Fig. 10c), the more elevated SiO$_2$ values observed in some samples could be explained with continental contamination or post-emplacement chert infiltration (e.g. Thurston et al., 2012) but the sub-silicic samples remain unexplained, confirming the finding of Herzberg (2016) regardless of whether the melts are better approximated by batch or accumulated fractional melting models.

A smaller subset of samples also reports REE data for which CI-normalised Sm/Yb ratios were calculated (Sm was preferred over Gd as more samples had values for the former). When plotted versus the Al$_2$O$_3$/TiO$_2$ ratio (Fig. 10d), the paradox explained by Walter (1998) becomes apparent. Namely, only the most Al-depleted samples show somewhat elevated Sm/Yb ratios whereas for samples with Al$_2$O$_3$/TiO$_2$ from 10 to 50, there is no correlation, which is unexpected if garnet controlled the degree of Al-depletion or enrichment (e.g. Nesbitt et al., 1979).

The final four panels of figure 10 show the same data but now colour-coded for stratigraphic assemblage to test whether the large petrological variability could correlate with age. However, there is no age progression in FeO vs. MgO and SiO$_2$ vs. MgO systematics. Smaller datasets (e.g. Pacaud) show tighter systematics but in the three dominant subsets (Kidd-Munro, Stoughton-Roquemaure and Tisdale), the full range of major element chemistries is represented.

In summary, high-Mg basalts, picrites and komatiites from the Abitibi and adjacent Pontiac Subprovince represent very wide petrological variety, including melts from more fertile mantle and from refractory sources. These diverse komatiites occur at several stratigraphic levels but are volumetrically much inferior to the dominant basalts (Fig. 9).
Over the reasonably narrow ca. 60 Ma time window, multiple mantle sources melted, likely at quite different depths.

4.3 Garnet and orthopyroxene textural relationships and REE systematics in Archaean orthopyroxene-rich cratonic harzburgite

The early studies on the relationships between Archaean cratonic harzburgites and komatiites established a paradigm of very deep (5-8 PGa) melting (e.g. Takahasi and Scarfe, 1985; Cox et al., 1987; Boyd, 1989; Canil, 1992; Herzberg, 1993). This has since been replaced with the currently prevailing idea (e.g. Canil, 2004; Lee and Chin, 2014; Regier et al., 2018) that proto-cratonic lithosphere thickened by stacking of harzburgite, which represents shallow melt residues (1-3 GPa). The paradigm shift was precipitated with the realisation that the minor and REE systematics of komatiites, Archaean cratonic garnets (Stachel et al., 1998), and of harzburgites (Canil, 2004) favoured melt extraction in the spinel stability field, implying subsequent thickening of the cratonic root. The high Cr/Al ratio of many cratonic harzburgites is quoted as very strong evidence against melt extraction deeper than 3 GPa, so much so that deeper melting is no longer discussed as a viable alternative by some authors (e.g. Regier et al., 2018). However, Tomlinson et al. (2018) offered observations that warrant a new look at the old paradigm of deep melting. These authors used phase relationships in a very coarse-grained orthopyroxene megacryst to postulate that most garnet in Archaean harzburgite was not originally present but formed by exsolution from orthopyroxene. In other words, during proto-craton formation the harzburgite was much hotter and only consisted of olivine and orthopyroxene, even though it technically resided in the stability field of garnet defined for pyrolitic mantle. Harzburgitic compositions can become garnet-free at the deep solidus and the Cr/Al systematics of experimental olivine and orthopyroxene residues (e.g. Walter, 1998) are compatible with the observed high Cr/Al many cratonic harzburgites at pressures of 3-7 GPa, provided the residue is highly depleted. Below we provide more data on the spatial association of garnet and orthopyroxene that has long been argued to contain evidence about the cooling trajectory (e.g. Canil, 1991) of harzburgites and also document much-needed REE concentration data for harzburgitic orthopyroxene.
4.3.1 The spatial relationship between garnet and orthopyroxene in garnet harzburgites

Cox et al. (1987) first systematically studied the spatial relationships of olivine, clinopyroxene, orthopyroxene and garnet in coarse-grained cratonic lherzolites. Their observations revealed one consistent phase relationship, namely a close spatial association of garnet and orthopyroxene. Because garnet was only found in association with orthopyroxene, they reasoned that garnet represented the Tschermak component of an original, higher-temperature orthopyroxene. Thus, garnet was a phase that exsolved from a parental higher-Al orthopyroxene upon cooling. Considering the experimental finding of Kushiro (1973), Cox et al. (1987) speculated that the more aluminous orthopyroxene precursors could have originated at pressures ranging from 4 to 8 GPa.

Using SEM-BSE, Saltzer et al. (2001) confirmed that the close spatial association of garnet and orthopyroxene also exists in more refractory harzburgites. They further discovered that those peridotites that had equilibrated at the greatest depth along the cratonic geotherm contain the most magnesian olivine. This observation does not in itself constrain the cooling trajectory, which Saltzer et al. (2000) suggested to have involved compression, a conclusion derived from comparison with experiments conducted on fertile peridotite compositions.

Tighter constrains on the cooling trajectory can be derived from analysis of spectacular examples of very coarse-grained orthopyroxenes that exhibit textural evidence for garnet exsolution. Gibson (2017) documented a range of garnet exsolution styles from very coarse-grained orthopyroxenes from the Tanzania and Kaapvaal cratons. A similar exceptionally well-preserved specimen of a large orthopyroxene single crystal (size of preserved relic 15 x 12 x 7 cm) from the Kimberley fields was studied by Tomlinson et al. (2018). Thermobarometry inferred equilibration at 4.4 GPa. By reconstituting the exsolved garnet (and, in some of the samples studied by Gibson (2017) additional clinopyroxene) into the orthopyroxene host, the Tschermak component of the original precursor pyroxene can be calculated and plotted as an isopleth into the garnet-orthopyroxene-olivine stability field of a model pseudosection. Provided that orthopyroxene formed on the solidus and that it was the only phase formed on the solidus, the original depth of formation can be obtained as the intersection of the isopleth with the solidus. In the cases of very large euhedral macrocrysts this assumption seems warranted and using this approach, Tomlinson et al. (2018) calculated that the precursor pyroxene of their studied sample formed at 5.9 GPa and 1,750°C, implying decompression upon cooling onto the cratonic geotherm. Tomlinson et al. (2018) noted that orthopyroxene with this Tschermak component could theoretically also exist on the very low
pressure solidus (0.3 GPa) within the plagioclase stability field but this is not considered a likely formation depth of any peridotites from the Kaapvaal craton (see also Lee and Chin, 2014).

It could be argued that the spectacular coarse-grained orthopyroxenes and their exsolved garnets are exotic curiosities with limited significance for the overall formation of the cratonic mantle. However, an important aspect of the specimen studied by Tomlinson et al. (2018) is that the megacryst is seen to disaggregate along its edges and that fragments of it, with garnet inclusions, are found in the enclosing harzburgite. Garnet and orthopyroxene in the harzburgite have exactly the same major and trace element composition as in the megacryst. The orthopyroxene carries substantial internal strain, a feature also noted by Gibson (2017), who attributed this to the mechanism of garnet exsolution but that could additionally have been caused during transport within the cratonic root. Considering that the peridotites apparently experienced vertical movement over tens of km, orthopyroxene disaggregation may have been a common process. This raises the possibility that a substantial proportion of garnet found in cratonic garnet harzburgites could have originated via exsolution, even if in most cases of granular peridotites, the exsolution process cannot be directly proven (Tomlinson et al, 2018). For example, Wasch et al. (2009) described conspicuous clots of coarse-grained orthopyroxene and garnet with some garnet visibly exsolved as linear bands within the orthopyroxene.

In figure 11, a series of section scans illustrates the progression from an example of an orthopyroxene megacryst with clear garnet exsolution lamellae to a ‘normal’ granular garnet harzburgite, with no obvious exsolution indication of the garnet. The intermediate examples are progressively less obvious cases for exsolution highlighting examples of garnet grains fully encased in orthopyroxene and relic alignment trails of garnet. A common feature of all samples (and most harzburgites from Kimberley) is the very coarse-grained nature of the orthopyroxene, often exceeding 10-15 mm in diameter. To further test the possibility that garnet in ordinary harzburgite originated by exsolution from formerly more aluminous orthopyroxene (e.g. Canil, 1991), we used automated phase mapping with large format SEM-EDS detectors to test the spatial relationship between garnet and orthopyroxene. The studied sample, 17BSK051, is a typical Kimberley field cratonic granular harzburgite, consisting of olivine, orthopyroxene and minor garnet with late accessory phlogopite. Garnet-orthopyroxene-olivine thermobarometry (Harley, 1984; Brey and Köhler, 1990) indicates equilibration at 954±5°C and 3.8±0.3 GPa, on the typical cratonic geotherm.
The fully-quantitative phase maps were analysed with the EDS system software tool 'AutoPhaseMap', which provides area and mean compositional information on identified phases. Shown together with a transmitted light thin section scan (Fig. 12) is a composite phase map and two isolated phase maps highlighting garnet, one alongside olivine, the other alongside orthopyroxene. These maps qualitatively show the close spatial association between garnet and orthopyroxene and the general paucity of shared grain boundaries between olivine and garnet. In two cases, garnet is nearly completely enclosed in orthopyroxene. In terms of area fraction of the three main phases, olivine occupies 68%, orthopyroxene 29% and garnet 3.0%. The mean Mg# of olivine from the phase map is 93.1, comparing very well with the mean of five spot analyses (93.2; Table 2). In the olivine mode vs. olivine Mg# diagram (Fig. 3f), the sample plots close to the 7 GPa residue trajectory (Walter, 1998) and is similar in mode and olivine composition to many of the Kaapvaal garnet harzburgites originally described by Boyd (1989).

4.3.3 Rare earth element systematics of selected garnets and orthopyroxenes in garnet harzburgites

Notwithstanding the wide agreement that the key phase for deciphering the Si/Mg harzburgite issue is orthopyroxene, the trace element and isotopic arguments have largely been made from analyses of garnet (e.g. Griffin et al., 1999; Stachel et al., 2004; Gibson et al., 2013; Shu and Brey, 2015). One main reason for focusing geochemical studies on garnet is that it has much higher partition coefficients than orthopyroxene or olivine for most trace elements (e.g. Sun and Liang, 2013), meaning that even in highly depleted peridotites, in situ analytical methods can quantify trace element concentrations. As shown here and elsewhere (e.g. Canil, 1992; Canil, 2004; Gibson, 2017; Tomlinson et al., 2018), many garnets found in highly depleted harzburgite formed upon cooling by exsolution from originally more alumino higher temperature orthopyroxene that existed on the solidus. Therefore, there is major petrological interest in the trace element geochemistry of orthopyroxene.

Of all measured trace elements in harzburgitic garnet, the REE are most frequently cited as requiring metasomatism to explain the unusual systematics. The key observation is that most analysed garnets do not show the simple steep CI-normalised REE pattern expected from experiments (e.g. Sun and Liang, 2013). Instead, at least half the studied garnets, including many diamond inclusions (e.g. Stachel et al., 1998; 2004), show some form of sinusoidally-shaped CI-normalised REE pattern with a 'hump' centred over the heavier LREE.
or the lightest MREE but with exceedingly low La and Ce concentrations (e.g. Gibson et al., 2013). This pattern is widely considered to imply metasomatic addition of LREE.

Here we report full REE data (Table 2) for garnet and co-existing orthopyroxene for the BP002 megacryst studied by Tomlinson et al. (2018) and for the phase-mapped granular harzburgite 17BSK051. The CI-normalised patterns for both phases (garnet: panels a and b and orthopyroxene: panels c and d) for both samples show the MREE-enrichment. For the analyses least compromised by kimberlite contamination and alteration (see also supplemental information), the HREE concentrations are below 0.5 ppb, with those of Tm and Lu below 0.1 ppb. Despite the large scatter in individual orthopyroxene HREE analyses (Fig. 13c), the pooled data from least contaminated analyses result in a smooth average patterns (Fig. 13g and h) characterised by near-symmetric parabola with an apex over Sm in Onuma-style diagrams. Both garnet and orthopyroxene patterns also show a secondary upswing in the heaviest HREE. For BP002, garnet/orthopyroxene apparent distribution coefficients (not shown) rise monotonically from 10 to 800 from La to Lu, as predicted from experimental data. In 17BSK051, garnet/orthopyroxene apparent distribution coefficients (not shown) rise monotonically from 2 to 500 from La to Er and stay around 500 for the remaining HREE. This could be an artefact of the uncertainties on the HREE concentrations for the orthopyroxene but a similar observation was also made by Wasch et al. (2009). Regardless, in both studied samples, garnet and orthopyroxene have sinusoidal REE patterns, strongly supporting the argument that garnet exsolved from orthopyroxene. The curious MREE-enrichment was therefore inherited from the precursor phase, implying that if metasomatism had occurred it would have predated the exsolution of garnet (Tomlinson et al., 2018). Alternatively, the relative enrichment in the MREE is unrelated to metasomatism and inherent to high Si/Mg harzburgite formation.

5. Discussion

Two of the most distinguishing features of Archaean cratons are the existence of highly refractory, strongly ultramafic harzburgite in the mantle root and the widespread occurrence of ultramafic lavas at the proto-cratic surface. When combined, these mutually related rock types could be thought of as ultramafic 'sandwich' layers of Archaean cratons. In this final section, we discuss the importance of mafic-ultramafic resurfacing for proto-cratic crustal architecture and stabilisation, analyse the types of mantle source regions that could have a formed the erupted lavas and residues found as xenoliths.


5.1 The role of mafic-ultramafic lids as conductive incubators

Several prominent papers have suggested that the A-P boundary did not represent a relatively sudden change in style of geology but that a more familiar geology began to emerge much earlier and more gradually, from ca. 3 Ga (e.g. Shirey and Richardson, 2011; Keller and Schoene, 2012; Dhuime et al., 2012; Tang et al., 2016; Smit and Mezger, 2017). Nearly all the supporting lines of evidence for this proposal are based on interpretation of geochemical and/or radiogenic isotope data. Several authors have argued that from ca. 3 Ga onwards, plate tectonics (Tang et al., 2016) or Wilson cycles (Shirey and Richardson, 2011) started operating. This geochemical view of a gradual transition from the Mesaoarchaeian to the Palaeoproterozoic contrasts with the traditional geological evidence, which is in favour of a much sharper end of the Neoarchaean. For example, Bédard (2018) pointed out that there is no obvious difference in the geology of Neoarchaean and Mesoarchaean or even older terranes, with the only exception of the appearance of K-rich granitoids at ca. 3 Ga. Here we propose that the seemingly incompatible lines of evidence can be reconciled by considering stratigraphic context, which is typically lost in large time-series compilations.

This is illustrated here with the example of the Abitibi greenstone belt, which has the best defined laterally coherent stratigraphy. Similar mafic-rock dominated cover sequences have been documented across other cratons, including the Zimbabwe (Wilson et al., 1995), the Eastern Dharwar (Jayananda et al., 2013), the East Pilbara (Hickmann and van Kranendonk, 2012), the Northeastern Superior (e.g. Maurice et al., 2009) and the Kalgoorlie terrane of the Yilgarn (Hayman et al., 2015). As illustrated earlier (Fig. 8), the stratigraphy of the Abitibi greenstone belt is made up of 7 lower assemblages dominated by volcanic rocks and two upper assemblages of sediment-dominated rocks deposited into successor basins (e.g. Ayer et al., 2002). Studies seeking to infer crustal evolution from the chemistry of sediment rely on faithful representativeness of the preserved sedimentary rocks, reflecting the full stratigraphy of the supracrustal strata.

However, in the Abitibi greenstone stratigraphy, sedimentary rocks of all types are only common in the uppermost two assemblages but exceedingly sparse within the 7 lower assemblages. There they occur between volcanic units as conformable condensed horizons (Thurston et al., 2008) composed of thin iron formation, chert, exhalite and so-called argillite, which is a very-fine grained sediment containing reduced C. Of these, only argillite has an appreciable siliciclastic component that could potentially be of use to the global geochemical
approach proposed by Keller and Schoene, (2012), Tang et al. (2016) and Smit and Mezger (2017). However, due to the precipitated authigenic component (Si, Fe) and the sequestration of transition metals from the water column into argillites, it is impossible to compare their chemistry with bona fide clastic metasediments. For example, the argillites of the Kidd-Munro and Kinojevis assemblages studied by Barrie (2005) in the Halloway area, have high average Co/Th and Cr/Th ratios, which would indicate a high mafic to ultramafic source component (Fig. 2b) but their average Ni/Co ratio is only 2.7±0.5, much lower than coeval siliciclastic sediments (Fig. 2d) compiled by Tang et al. (2016). This is not because the sediment source of the argillites is more felsic but because of the higher concentration of Co than Ni in VMS-type hydrothermal fluids. For many greenstone belts, the desired clastic sedimentary rocks sampling the main mafic-ultramafic magmatic units simply do not exist and are therefore not represented in global datasets.

Instead, the datasets are dominated by and skewed towards the sand-, silt- and mudstones deposited into the late successor basins. These formed in rifts (e.g. van Kranendonk et al., 2010), coevally with emplacement of potassic magmas (e.g. Mueller et al., 1994) post-dating major mafic and ultramafic volcanism. The chemistry of these sedimentary rocks is thus not representative of the bulk of the greenstone belt. In the Abitibi greenstone belt, Feng and Kerrich (1990) studied the chemistry of rift-hosted Porcupine and Timiskaming assemblage sedimentary rocks (the two youngest assemblages) and the Ni/Co of the various subunits from their study is shown in figure 2d. The mean of all units (4.9±0.5) confirms the trend identified by Tang et al. (2016) from global compilations (the mean of their 2.7 Ga data is 5.6±0.35). We propose that these sediment chemistries inform about the progress of intra-crustal differentiation and cratonic stabilisation rather than secular changes in crustal makeup or plate tectonics. Strong upward vertical movement of the radioactive heat producing incompatible elements K, U and Th in granitoid melts was accompanied by downward foundering of supracrustal lithologies, including of Timiskaming age (Lin et al., 2013). Because successor basins and their sediments only started forming as the crust was gaining mechanical stability (e.g. Bédard, 2018), there is an inherent bias towards preserving detritus from these more evolved sources. This limitation does not narrow the significance of the global analyses of sediment databases (e.g. Tang et al., 2016; Smit and Mezger, 2017) but complicates the tectonic interpretation of resulting temporal trends.

Our proposal is supported by studies that focus on the chemistry of the magmatic rather than on sedimentary rocks. Moyen and Laurent's (2017) analysis of the mafic igneous rock database concluded that (p.116-117): "the result is surprisingly disappointing, as rocks
throughout the Archaean show exactly the same geochemical pattern, from the oldest rocks known at Isua or Nuvvuagittuq to the latest Archaean e.g. in China or Zimbabwe – i.e. over about 1.5 Ga of geological history”. With respect to the ultramafic rocks, the present analysis of the MgO-distribution of Archaean volcanic rocks also lacks evidence for a change in the chemistries of lavas that erupted between the Meso- and Neoarchaean (Fig. 7). Furthermore, the maximum MgO of Archaean komatiites does not show a secular trend either (e.g. Kamber, 2010) and there is no evidence for a secular decrease of depletion extent within Archaean harzburgite suites but there is a strong change after the A-P boundary (e.g. Boyd, 1989; Griffin, 1999). We regard these as very important observations, because mafic and ultramafic magmas have potential to inform about changes in mantle melting regime.

By contrast, there is very strong evidence from many cratons (e.g. Smithies et al., 2009; Laurent et al., 2014) for a temporal and stratigraphic evolution of granitoid magmatism, expressed broadly as a trend towards more potassic compositions and shallower emplacement (for much more detail, see Moyen and Laurent, 2017). This trend is also observed in the Abitibi greenstone belt (Fig. 8c) where the oldest plutonic rocks are grey gneisses of diorite-tonalite composition, maturing with time to granodiorites and biotite-bearing granodiorites (e.g. Benn, 2004), before reaching granitic compositions that typify shallow late intrusions (e.g. Petrus et al., 2016). Laurent et al. (2014) interpreted the first appearance of sanukitoids at ca. 3 Ga to mark the onset of proper plate collision. However, the alternative explanation is that the evolution of felsic magmas is the reflection of progressive differentiation of the crust (Bédard, 2018).

There are two main factors that drove crustal differentiation in the Archaean. The first is the substantially higher radioactive heat production within the crust and the second is the repeated emplacement of thick supracrustal mafic-ultramafic lids (e.g. van Kranendonk et al., 2015, Wiemer et al. 2018). The resulting incubation accelerated intracrustal crustal reorganisation (Sandiford et al., 2004), with a general vertical rise of the incompatible heat producing elements in granitoids and granites. Accordingly, the late intrusion of high-K, shallow lopolithic intrusions (e.g. Frei et al., 1999) marks the mechanical stabilisation of the crust (e.g. Kamber, 2015; Bédard, 2018), completing the process of cratonisation.

The idea of vertical foundering or sagging of denser and colder greenstones between granitoid diapirs, also known as 'dome and basin' or 'balloon' tectonics (e.g. Ramsay, 1989), is not universally accepted (e.g. Kimura et al., 1993). However, thermal and viscoelastic mechanical modelling of inverted density profiles (Robin and Bailey, 2009; Thebaud and Rey, 2013) shows that it remains a plausible mechanism. Furthermore, field investigations (e.g.
Blenkinsop et al., 1993; Ayer et al., 2002; Johnson et al., 2016; Wiemer et al., 2016) on various cratons have questioned the scepticism towards ‘dome and basin’ tectonics that arose from the LITHOPROBE project (e.g. Clowes et al., 1992). It is perhaps unfortunate that ‘dome and basin’ tectonics has been termed 'sagduction' by some (e.g. Chardon et al., 1996) as this intracrustal process (e.g. Bouhallier et al., 1995) is not an alternative to subduction, which describes movements (that include a vertical component) of one plate relative to another. Crustal reorganisation through radioactive heat and conductive incubation is not unique to the Archaean (Sandiford and McLaren, 2002) but is much less common in the post-Archaean geological record. Because radioactive heat production decayed smoothly with time, it alone cannot explain the existence of the A-P boundary, which more likely represents the combination of several factors. Here we argue that after 2.5 Ga, the inability to commonly produce low viscosity high-Mg lavas as during the Archaean resulted in the reduction of extent of mafic-ultramafic resurfacing, i.e. the deposition of thinner ensialic greenstone blankets. It is difficult to reliably estimate the original thickness of supracrustal blankets because in most cases, the dense rocks have foundered and are now preserved as highly deformed remnants at mid-crustal or deeper levels (e.g. Bouhallier et al., 1995; Johnson et al., 2016) or could have been lost as foundered eclogite (e.g. Johnson et al., 2014). Furthermore, greenstone stratigraphies were not deposited with uniform thickness across the proto-cratons. Variations in thickness of volcanic units may reflect proximity to major volcanic centres, topography at the proto-cratonic surface and syn-depositional deformation (e.g. Berger et al., 2011). Observed stratigraphic thicknesses of rapidly emplaced volcanic units (‘groups’ or ‘assemblages’) range from 3-5 km in the Eastern Yilgarn (Hayman et al., 2015) and the Eastern Dharwar craton (Jayananda et al., 2013) to ca. 10 km in the East Pilbara terrane (Hickman and van Kranendonk, 2012) and Abitibi greenstone belt (Berger et al., 2011). Full greenstone belt thicknesses are much greater, reaching up to 20-25 km (Ayer et al., 2002; van Kranendonk et al., 2007). In numerical models, deposition of 5-15 km thick incubating lids of mafic-ultramafic cover sequences drives the observed crustal reorganisation. We argue that the markedly reduced post-Archaean conductive incubation together with reduced crustal heat production contributed to the observed change in geological style. In post-Archaean terranes that did experience strong basaltic to picritic volcanic resurfacing (e.g. the Palaeoproterozoic Flin Flon (Canada) and the Ashanti (West Africa) greenstone belts; Perrouyt et al., 2012), the crustal architecture has an Archaean flavour but komatiite is largely absent and the prevailing theory is that these Palaeoproterozoic terranes formed via arc-accretion (e.g. Lucas et al., 1996).
5.2 Melting of fertile, highly refractory or metasomatised mantle?

Turning from the ultramafic rocks at surface to those in the lithospheric root, the various possible petrological models that could have left many cratonic peridotites with high enstatite/olivine (i.e. high Si/Mg) ratio have been reviewed by Herzberg (1993). At the time, the preferred explanations ranged from melting residues of a non-pyrolitic mantle, metamorphic segregation (e.g. Boyd, 1989) to cumulate crystallisation from a high-SiO\textsubscript{2} magma ocean. One alternative model, metasomatic addition of SiO\textsubscript{2} in subduction zones (Kesson and Ringwood, 1989) was less popular on account of the high fluid/rock ratio required (e.g. Canil, 1992) and the mismatch of observed with predicted CaO, Al\textsubscript{2}O\textsubscript{3} and SiO\textsubscript{2} systematics (Herzberg, 1993). With the discovery of the sinusoidal REE patterns in cratonic harzburgite garnets (Stachel et al., 1998) the pendulum swung in favour of metasomatic addition of LREE and SiO\textsubscript{2} (e.g. Simon et al., 2003, 2007; Bernstein et al., 2007; Pearson and Wittig, 2008; Wasch et al., 2009). However, a comprehensive survey of the O-isotope composition of variably Si-enriched cratonic peridotites has documented a uniform mantle parentage (Regier et al., 2018), strongly arguing against the subduction zone metasomatic model.

An aspect common to most of the quoted studies is that they compare cratonic harzburgite olivine modes and forsterite contents with the empirical trend from depleted modern oceanic peridotites (Fig. 3c) originally proposed by Boyd (1989). However, Boyd's trend was never scrutinised with experimental data. Comparison with the experimental melting residues of Walter (1998) shows a strong pressure effect (Fig. 3d). None of the isobaric experimental residues reproduce the (hand-drawn?) concave trend of Boyd (1989). At pressures of 5 GPa or higher, the experimental olivines are highly magnesian and exist in a residue containing 25-35 % modal combined pyroxene and garnet (Walter, 1998). It is recognised that this comparison is limited to one experimental dataset because most other relevant high-pressure studies did not tabulate estimates for modal abundance. We note, however, that in terms of phase relationships and stability fields, Walter's (1998) experiments are compatible with those conducted at lower pressure and those that did not report full modal abundances (e.g. Takahashi, 1986; Canil, 1991; 1992; Takahashi et al., 1993; Baker and Stolper, 1994; Longhi, 2002; Schutt and Lesher, 2006). There is thus no indication that the modes from these experiments would be very different. We agree with Canil (2008) that the mismatch between observed and expected cratonic harzburgite mineralogy is strongest.
the Kaapvaal craton but harzburgites with <60% modal olivine also exist from the Slave, Siberian and Wyoming cratons (Eggler et al., 1987; Ionov et al., 2010; Kopylova et al., 1999; Kopylova and Caro, 2004; Newton et al., 2015; Solovjeva et al., 1994). The relatively low modal abundance of olivine could mainly indicate that melt extraction occurred at high pressure but further in-depth analysis will be needed for a firmer conclusion. In terms of the variability in olivine mode at a given forsterite content (Fig. 3f), we note again the extremely coarse-grained nature of cratonic harzburgites (e.g. Figs. 11 and 12) and propose that some of the modal variability could stem from minor mineral segregation and from unrepresentativeness of small thin sections.

One aspect of the cratonic Si/Mg issue that has not yet been explored experimentally is the possibility that high-Mg harzburgites could be residues of multiple melt extraction events. Arndt (1977a) commented (p. 205) that "ultrabasic magmas probably form by a sequential melting process and are derived from a residuum composed of refractory minerals and trapped liquid left by previous episodes of partial melting and magma extraction". Our analysis of the experimental liquids of Walter (1998) shows that re-melting of a previously depleted source can produce a much wider range of Si/Mg in the liquids than single batch melting (Fig. 10g and h) as well as more extreme Fe/Mg and Al/Mg variability as seen in some komatiites. More experimental data on refractory starting compositions and modelling of melt-residue interaction are certainly needed to better understand the Si/Mg systematics of komatiites and harzburgites (e.g. Herzberg, 2016), particularly in light of the new O-isotope data (Regier et al., 2018) that argue against subduction zone metasomatism.

There are strong indications from the geochemistry of komatiites and komatiitic basalts that variably fertile sources were being melted (e.g. Sproule et al., 2002). Although the source fertility of komatiites is conventionally determined via Al (Nesbitt et al., 1979), experimental data show that the Ti concentration may be a simpler measure to assess whether a source had been previously depleted (see also Sproule et al., 2002; Robin-Popieul et al., 2012). This is because Ti remains incompatible across the entire melting depth interval relevant to cratonic lithosphere (Longhi, 2002). The archetypes of melts from ultra-depleted sources are from Commondale (Wilson, 2003) but komatiites with Al₂O₃/TiO₂ >25 have also been reported from localities outside the Kaapvaal craton (e.g. Sproule et al., 2002). This supports the proposal of Arndt (1977a) and suggests that additional experimental constraints are required for harzburgitic starting compositions. The low-Ti nature of many komatiites is shared (to some extent) by modern boninites but this common trait is, in our view, insufficient to justify the subduction zone origin proposed by Parman et al. (1997).
In terms of REE systematics, one key question of relevance to proto-craton formation is whether the sinusoidal REE pattern arose from metasomatism post-dating the original melt-extraction (e.g., Stachel et al., 2004) or whether it is an inherent feature of cratonic mantle related to its formation by high degree melting (e.g., Tomlinson et al., 2018). There is no doubt that cratonic lithosphere has experienced multiple episodes of metasomatic infiltration, particularly in the base (e.g., Nixon et al., 1987; Foley, 2008) but there is very strong evidence that the sinusoidal REE pattern is ancient and not related to Neoarchaean metasomatism. The following are the strongest lines of evidence. Firstly, the Hf-isotope composition of garnets with sinusoidal REE patterns are extreme, reaching to spectacular present-day ε(Hf) of up to +1,000 (Shu and Brey, 2015) with correspondingly ancient Archaean model ages. Shu et al. (2013) discovered that garnet separates (with sinusoidal REE patterns) from Kaapvaal craton kimberlites yield Lu/Hf regression lines of 3.2 Ga, suggesting that the sinusoidal REE pattern has to be at least as old. This age is substantially older than the Neoarchaean cratonisation. Secondly, Koorneef et al. (2017) succeeded in dating diamond inclusion garnet populations with strongly sinusoidal REE patterns to 2.95 Ga with 147Sm/143Nd. The majority of these garnets have very low Sm/Nd ratios, consistent with the hump in the REE pattern over Nd and Sm. Thirdly, as shown by earlier studies (Zhang et al., 2001; Simon et al., 2007; Wasch et al., 2009) and the new data on the BP002 megacryst (Tomlinson et al., 2018; this study), the REE of orthopyroxene also show a humped pattern, implying that the entire protolith peridotite carried this signature. Finally, Stachel et al. (2006) reported on diamond inclusions hosted in 2.7 Ga volcaniclastic rocks and lamprophyre dykes in the Wawa greenstone belt. The inclusion suite contains garnets very similar to those in common cratonic harzburgite and the nitrogen aggregation of the diamonds suggests residence times of tens to hundreds of Ma prior to crustal emplacement. Collectively, this evidence suggests that harzburgite with sinusoidal REE patterns existed in the Meso- and Neoarchaean cratonic mantle root.

This finding is relevant to komatiite geochemistry in two respects. Firstly, garnet and orthopyroxene are the main carriers of the REE in advanced melt residues between 4 and ca. 8 GPa. Depending on the magnitude of the hump over the heavier LREE (Nd-Gd), the Sm/Yb or Gd/Yb ratio of a melt in equilibrium with these phases will either be slightly sub- or supra-chondritic. The important conclusion is that if the komatiite source had a sinusoidal REE pattern, there will only be limited variability in Sm/Yb or Gd/Yb ratios in liquids. This is one possible solution to the paradox identified by Walter (1998), namely that major elements of
many komatiites require a garnet-bearing source but REE do not show the expected large Sm/Yb fractionation. As the new examples of garnet from exsolved harzburgite orthopyroxene (Tomlinson et al., 2018; this study Figs. 11 and 12) show, melt extraction at >3 GPa is not necessarily in conflict with the high Cr/Al ratios of residues, provided they were originally bi-mineralic olivine-orthopyroxene rocks. Secondly, a source with sinusoidal REE patterns has the potential to evolve to diverging Nd and Hf isotope compositions. Because the apex of the hump in the REE pattern is frequently around Sm or Nd, the resulting Sm/Nd does no differ much from chondrite and Nd-isotope ratios will evolve to mildly supra- or even sub-chondritic values. By contrast, the Lu/Hf ratio is mostly strongly superchondritic and Hf-isotope ratios rapidly evolve to positive ε(Hf) values. We propose that this could explain the Nd-Hf isotope systematics of the Commondale komatiites (Hoffmann and Wilson, 2017) and the decoupled Nd-Hf arrays of other Kaapvaal craton komatiites (Blichert-Toft et al., 2015).

It is beyond the scope of this paper to fully discuss the various proposals for the origin of the sinusoidal REE pattern and its possible relation to Si enrichment (e.g. Burgess and Harte, 2004). If the sinusoidal REE pattern can be attributed to metasomatism with (e.g. Gibson et al., 2013) or without previous melt depletion it has to be Archaean in age and, in some cases, clearly predates craton stabilisation. Just as with the Si/Mg systematics, there remain significant gaps in experimental knowledge for REE behaviour in peridotite at the conditions relevant to cratonic melting. For example, pseudo-sections for unmetasomatised fertile peridotite predict that the phase assemblage at the 5 GPa solidus is orthopyroxene-free, consisting only of olivine, clinopyroxene and garnet (e.g. Fig. 1 of Jennings and Holland, 2015). Thus, due to the expansion of the stability field of pigeonitic clinopyroxene above 1,500°C and pressures greater than 4.2 GPa (Takahashi, 1986), the peridotite that is melting is technically called a garnet wherlite, not a lherzolite. To our knowledge, no relevant experimental data exist for the REE partitioning of this type of pyroxene. Until the necessary experiments have been conducted, it will remain unclear if a sinusoidal REE patterns could result in residues of multiply melted normal peridotite without metasomatic REE addition.

5.3 Possible causes for the demise of very high-temperature mantle upwellings and the relevance for the Archaean-Proterozoic boundary
There remains much uncertainty regarding the A-P boundary. In our view, the geological evidence in favour of a boundary outweighs the geochemical indications for earlier (e.g. 3 Ga) planetary changes in tectonic style. If we accept the geochronological confirmation of the original postulation of the A-P boundary in the 19th century (Logan, 1857) as more than a fortuitous coincidence, the boundary must have major geological significance.

Most ideas seeking to explain the A-P boundary advocate a reorganisation of the mantle temperature structure and/or heat loss mechanism. Two schools of thought are most prominent. Firstly, a change within or above the core-mantle thermal boundary layer may have occurred and this affected the type of thermal upwellings that travelled through the mantle. For example, Campbell and Griffith (2014) attributed the A-P boundary to piling subducted oceanic plates into D", forming an insulating layer from which modern-style plumes began emerging since the Palaeoproterozoic. Likewise, Bédard (2018) envisaged mantle 'overturn upwelling zones' that episodically transported heat and material from the lower mantle towards the surface.

The second group of ideas has identified the mantle transition zone as a possible location where major change may have occurred at the A-P boundary. In these models, the base of the transition zone was a much stronger thermal boundary layer in the Archaean than thereafter. The hot mantle upwellings that caused komatiite magmatism could have originated at this boundary layer. Davies (2008) proposed that throughout the Archaean, subducted slabs piled up in the transition zone, forming a mechanical barrier that impeded heat loss from the lower mantle. According to his model, episodic collapse of the barrier occurred every 100-150 Ma and manifested as pulses of high temperature melting events in the upper mantle. From the A-P boundary onwards, slabs began to penetrate the lower mantle and the transition zone barrier stopped building up. Rino et al. (2004) advocated a similar model and they regarded the greater thickness of the subducted Archaean oceanic crust as key to the neutral buoyancy of slabs in the transition zone. Breuer and Spohn (1995) argued that the A-P boundary marked the change from 2-layer to single layer mantle convection and envisaged a pronounced flush instability that caused a global Neoarchaean igneous 'super event'.

In our view, all these models contain attractive aspects to explain the A-P boundary and it is noted that the Archaean pile-up of slabs in the transition zone is not mutually exclusive with post-Archaean slab pile-up in D". However, none of the current models has addressed the wider evidence from the crustal and mantle portions of cratonic lithosphere. We regard the following four key observations as most relevant for such a model:
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i) All cratons either still have or have formerly had deep mantle roots with geotherms corresponding to an average present-day heat flow of 41 mW m$^{-2}$ and strong concentration of radioactive heat production within the uppermost crust (e.g. Michaut et al., 2009).

Importantly, relatively cool cratonic mantle geotherms apparently also existed in the Archaean (Boyd et al., 1985). In the Kaapvaal craton and elsewhere, there is evidence that the Neoarchaean and early Proterozoic geotherms were surprisingly cool, only mildly more elevated than today's (around 45-50 mW m$^{-2}$; e.g. Nimis, 2002). This requires that in the Archaean, conductive heat loss extended to greater depth than the base of the current cratonic roots and/or that the original cratons were larger in extent than the fragments preserved today (Ballard and Pollack, 1988). This requirement is illustrated on figure 14. It is widely agreed that most cratons show evidence for post-Archaean base erosion (e.g. Foley, 2008; Xu and Qiu, 2017) supporting the notion that they were originally deeper. Rare xenoliths from the Kaapvaal craton contain majoritic garnet relics from 300-400 km depth (Haggerty and Sautter, 1990), deeper than the present-day base of the lithosphere. Regardless of preferred proto-craton thickness, strongly elevated lower crustal temperatures are unavoidable (Fig. 14).

ii) Throughout the Archaean, there is no evidence for secular change in komatiite Mg-content (Kamber, 2010; Campbell and Griffiths, 2014) and mafic magmas in general have remained chemically unchanged (e.g. Moyen and Laurent, 2017). But there was a sharp drop in MgO content of ultramafic magmas in the Palaeoproterozoic. By contrast, basaltic magmas have persisted through geological time with very similar mean MgO content as far back as the Palaeoarchaean (Kamber, 2015 and this study).

iii) Archaean crustal rocks, and particularly komatiites and associated fractionated basalts, preserve evidence for very ancient element fractionation of Sm/Nd, Hf/W and Sm/Lu (e.g. Caro et al., 2003; Touboul et al., 2012). The resulting $^{142}$Nd and $^{182}$W anomalies and the $^{143}$Nd/$^{176}$Hf divergence are more pronounced in Palaeo- and Mesoarchaean rocks (e.g. Bennett et al., 2007; Rizo et al., 2011) but still found in Neoarchaean samples (e.g. Debaille et al., 2013; Puchtel et al., 2016). It follows that the mantle domains harbouring such anomalies must have escaped convective mixing until the Neoarchaean and earliest Palaeoproterozoic.

iv) Although in many cratons the crust and the mantle portion experienced coeval events (melting, depletion, reworking, re-enrichment, etc.) the two reservoirs are not complementary. The preserved cratonic mantle root alone cannot have supplied all the incompatible trace elements present in the crust (e.g. Carlson et al., 2005). Therefore, crust
formation must have depleted greater volumes of mantle that never were or no longer are spatially associated with the craton.

One possible solution to the last two observations could be that proto-cratonic mantle formed in multiple ways. This could include initial shallow melt extraction from asthenosphere that only produced a relatively thin refractory root, maybe due to effective dripping of crustal restite residues (e.g. Bédard, 2006; Rollinson, 2010; Johnson et al., 2014). There is no a priori necessity that these proto-cratons grew from unusually hot mantle. Indeed, the tholeiitic lavas on many cratons could have been sourced from relatively shallow mantle (e.g. Wyman et al., 2002; Smithies et al., 2009). By contrast, the rocks that now constitute the deep cratonic mantle root may have originated in an unrelated part of the mantle – here termed ancient non-convecting domains (ANCD).

One plausible way of producing ANCD early in Earth history is crystallisation and crystal segregation in magma seas (e.g. Arndt et al., 2002; Hansen, 2015). The geotherm after crystallisation of a global magma ocean is very different from the adiabat and almost certainly leads to instability (e.g. Elkins Tanton et al., 2003; Kramers, 2007) that could have yielded domains of ANCD. An additional possibility is that giant impacts caused more local, shallower magma seas and their crystallisation products (cumulates) may have persisted (e.g. Hofmeister, 1983) as ANCDs surrounded by asthenosphere. Alternatively, stratified crystallisation products of magma seas can experience gravitational instability and may founder (e.g. Elkins Tanton et al., 2002) and could survive as non-pyrolitic ANCDs at depth. We note that cumulate formation during fractional crystallisation from a magma ocean has been suggested as one possible cause for the short-lived radiogenic isotope anomalies recorded in Archaean rocks (Caro et al., 2005). Finally, very deep mantle melting from lower mantle upwellings could have formed refractory mantle zones not originally associated with a proto-craton.

In figure 15, we illustrate that if ANCDs occupied laterally extensive areas in the mantle transition zone, they could have impeded heat transfer from the lower mantle. Heat would eventually escape, either through partly eroded ANCDs or along their margins as upwellings that triggered high temperature melting in the asthenosphere. Nascent continental lithosphere that drifted over areas covered by ANCDs at depth would be mostly protected from the extensive resurfacing that affected most of the remaining Earth. The nascent continental plates could have been similar to the ribbon continents proposed by Bédard (2018). Alternatively, they could have formed as arc-like collages (e.g. Wyman et al., 2002)
that occasionally experienced emplacement of much hotter and deeply sourced magmas through the ANCD.

With time, the accumulated effects of thermal upwellings weakened the ANCDs to the point of collapse, marking the A-P boundary. Remnants of the ANCDs were mostly recycled back into the asthenosphere but locally, became incorporated into the cratonic roots. The accompanying decompression and cooling could explain mineral reaction textures (including exsolutions) in garnet harzburgites and pyroxenites (e.g. Haggerty and Sautter, 1990). The dismembered deep parts of ANCDs would have carried with them transition zone minerals that could survive as diamond inclusions. The majority of transition zone garnet-structured diamond inclusions are of mixed peridotitic/pyroxenitic chemistry (Kiseeva et al., 2013) and only a minority has an eclogitic parentage. If these inclusions are samples from the deep ANCDs, their chemistry favours the idea that the barriers were mainly composed of pyroxenite progenitors rather than subducted oceanic crust but it is possible that slab pile-up as envisaged by Rino et al. (2004) and Davies (2008) added to the mechanical and thermal barrier. The initial Hf-isotope composition of Kaapvaal craton harzburgite garnets presented by Shu et al. (2013) are radiogenic and require that the precursor material separated from chondritic mantle at least a few hundred million years before 3.2 Ga. This constraint on timing is consistent with the idea that ANCDs could have formed in giant impact basins (e.g. Hansen, 2015) during the late bombardment (Bottke et al., 2012).

A model in which cratonic lithosphere grew from above and was fused with deep remnants could also explain why one class of harzburgites (those without high modal abundance orthopyroxene) appears to have mainly foundered after melt extraction (Canil, 2004; Lee and Chin, 2014) whereas others, like BP002, appear to have moved upwards (Haggerty and Sautter, 1991; Tomlinson et al., 2018). We also note that some cratons may have more thoroughly depleted lower roots and more lherzolitic shallower portions (e.g. Griffin et al., 2003). This observation could be explained by re-enrichment of formerly more strongly depleted harzburgite with fluids from depth (e.g. Gibson et al., 2013). Alternatively, the working model advanced here would envisage the less depleted zone to represent the area of fusion of shallower with deeper residues. It corresponds in depth both with the impedance increase found by Green and Hales (1968) and a 50 km thick zone of pronounced anisotropy with horizontal azimuths that could represent frozen-in drift (Sodoudi et al., 2013) of the shallow nascent continental plates.
6. Concluding remarks

Regardless of the validity of competing models for craton formation, the general difficulties of sourcing sufficient highly incompatible elements for cratonic crust (e.g. Bédard, 2006; Michaut et al., 2009) and the requirement for cool temperatures in the Archaean roots (e.g. Nimis, 2002) remain. With respect to the former, Kramers and Tolstikhin (1997) pointed out that the formation of a global Hadean protocrust could have led to an initial overall depletion of the entire mantle. When the Hadean crust was later comprehensively destroyed, the Eoarchaean asthenosphere could have ended up with an elevated inventory of incompatible elements if crustal recycling was restricted to a depth of 670 km. Melting of such enriched asthenosphere to form Archaean cratons could go a long way towards alleviating the mass imbalance between cratonic root and crust for highly incompatible elements. Furthermore, depending on the mass and composition of the Hadean crust, the upper mantle may also have ended up with a slightly elevated Si/Mg ratio with higher modal abundance of orthopyroxene.

Concerning the requirement for surprisingly cool temperatures in the Archaean cratonic roots, it is worth noting that very limited knowledge exists about the original dimension of Archaean cratons (e.g. Bleeker, 2003). The model of Ballard and Pollack (1988) assumed relatively modest diameters of proto-cratons (400-600 km) informed by the then prevailing notion that cratons contained small ancient nuclei onto which additional lithosphere accreted over time (e.g. Kusky, 1998). However, even in the Zimbabwe craton, where the evidence for a small original nucleus was once thought strongest, newer zircon U/Pb dates (e.g. Horstwood et al., 1999; Bolhar et al., 2017) and Re-depletion constrains on the cratonic root (Smith et al., 2009) now suggest that the craton had attained its full preserved size from the beginning. Furthermore, detailed comparison of crustal stratigraphies, radiogenic isotope characteristics, dyke swarm analysis and palaeomagnetic data imply that the Zimbabwe craton could have been part of the same craton as the Yilgarn (Smirnov et al., 2013). There is the additional possibility that this Zimgarn craton could itself only be a fragment of an even larger original craton (e.g. Bleeker, 2003) composed of additional cratons (including the Slave, Wyoming and North China) that all share the so-called high \( \mu \) Pb-isotope signature (Kamber et al., 2003; Kamber 2015). In terms of Archaean geodynamics, this opens the possibility that craton formation and preservation may not have been a ubiquitous process but that only two or three large early cratons with thick roots may have formed in environments that favoured their preservation and that they bias our understanding of how early terrestrial geology operated.
Acknowledgments

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References


Konhauser, K.O., Pecois, E., Lalonde, S.V., Papineau, D., Nisbet, E.G., Barley, M.E., Arndt, N.T.,
Zahnle, K., Kamber, B.S., 2009. Oceanic nickel depletion and a methanogen famine before the

and Proterozoic diamond growth from contrasting styles of large-scale magmatism. Nature
Communications, 8(1): 648.

Kopylova, M., Caro, G., 2004. Mantle xenoliths from the southeastern Slave craton: evidence for

from the Jericho kimberlite: implications for the thermal state of the mantle beneath the Slave

17.

Kramers, J.D., Tolstikhin, I.N., 1997. Two terrestrial lead isotope paradoxes, forward transport
modelling, core formation and the history of the continental crust. Chem. Geol., 139: 75-110.

Kramers, J.D., Kreissig, K., Jones, M.Q.W., 2001. Crustal heat production and style of metamorphism:
a comparison between two Archaean high grade provinces in the Limpopo Belt, southern Africa.


Kusky, T.M., 1998. Tectonic setting and terrane accretion of the Archean Zimbabwe craton. Geology,
26(2): 163-166.

Laurent, O., Martin, H., Moyen, J.-F., Doucelance, R., 2014. The diversity and evolution of late-
Archean granitoids: Evidence for the onset of “modern-style” plate tectonics between 3.0 and 2.5


Lin, S., Parks, J., Heaman, L.M., Simonetti, A., Corkery, M.T., 2013. Diapirism and sagduction as a
mechanism for deposition and burial of “Timiskaming-type” sedimentary sequences, Superior
Province: Evidence from detrital zircon geochronology and implications for the Borden Lake
conglomerate in the exposed middle to lower crust in the Kapuskasing uplift. Precambrian


Sun, C.G., Liang, Y., 2013. The importance of crystal chemistry on REE partitioning between mantle minerals (garnet, clinopyroxene, orthopyroxene, and olivine) and basaltic melts. Chemical Geology, 358: 23-36.


Figure captions

Figure 1: Three-dimensional representation of tomography of the African lithospheric mantle (reproduced from Begg et al., 2009). The viewer is positioned in the mantle transition zone below the southern Atlantic ocean and is looking upwards in a NE direction towards Africa (continental outline as a white line). The up to 250-275 km deep roots of the three broad cratonic regions of Africa are labelled and defined as red coloured volumes in which the SH body-wave velocity is >1.9% than the standard model. Blue colours show volumes of unusually slow SH body-wave velocities, mainly portraying the upwelling of hot mantle below the East Africa rift system.

Figure 2: Panel (A) and (B) redrawn in colour from Figs. 18 and 19 of Condie (1993). In both panels, the coloured symbols show shale compositions. Phanerozoic and Proterozoic shales overlap in composition but Archaean shales are much richer in compatible elements Sc, Co, and Cr. The Phanerozoic and Proterozoic UCC average composition overlaps with shale data but Archaean shales are more compatible element-enriched than UCC as a whole. Panels (C) and (D) show secular evolution of UCC Ni/Co ratio reconstructed from clastic sedimentary rock compilation of Tang et al. (2016). Panel (C) plots average values with standard errors and panel (D) same as (C) with additional data from the Abitibi greenstone belt (Feng and Kerrich, 1990; Barrie, 2005).

Figure 3: Empirical and experimental data plotting modal abundance of olivine in residual peridotite vs. forsterite component in olivine. Panels (A) to (C) redrawn from Boyd (1989) showing the oceanic trend (A), off-craton harzburgites (B) and Kaapvaal craton harzburgites (C). Panel (D) shows residue mineralogy of the experiments conducted on fertile peridotite by Walter (1998) colour-coded according to experimental pressure. The cross symbols show the position of the oceanic trend of Boyd (1989). Note that the y-axis scale extends to much more magnesian olivine. Panel (E) shows the compilation of empirical peridotite compositions of Bernstein et al. (2007). Panel (F) shows the same experimental data as panel (D) but at the same scale as Boyd's (1989) figures and the position of sample 17BSK051 studied herein (orange square symbol). See text for explanations.
Figure 4: Map showing kimberlite occurrences with and without sub-calcic garnet macrocrysts and lithospheric cross-section with interpretative areas of garnet formation and metasomatic alteration. Simplified and re-drawn in colour from Nixon et al.'s (1987) figures 261 and 267.

Figure 5: Comparison of empirical (Alexo and Pyke Hill) komatiite major element chemistry with petrological predictions reproduced from Herzberg (2016). Panel (A) shows MgO vs. SiO$_2$ compositional space in which the observed LLD (open circles whole rocks and solid grey circles melt inclusions), caused by olivine crystallisation, plots at lower SiO$_2$/MgO than either batch (red cross) or accumulated fractional (green cross) melting of the melting model predictions. Panel (B) shows MgO vs. FeO compositional space in which the observed LLD of the classic Abitibi greenstone belt komatiites strongly favour batch over accumulated fractional melting of a fertile peridotite (KR4003).

Figure 6: Magnesium (MgO wt%) distribution in Archaean metavolcanic rocks from six cratons (panels A-F) for which there are more than 500 compiled analyses; a composite of four cratons with fewer data (panel G) and for the combined dataset (panel H, note logarithmic x-scale). Each panel shows the raw data as histograms of MgO wt% re-calculated to anhydrous total of 100 wt%. Superimposed on the histograms is a kernel density function (parameters listed in Table 1) as a dark blue curve with light blue fill. Linear scale diagrams highlight the dominant basaltic population at MgO 5-9 wt%, the prominent evolved population with ca. 1 wt% MgO, and the much broader ultramafic population (MgO 25-30 wt%). In all panels, the paucity of rocks with MgO of ca. 18 wt% is evident. The composite figure (panel H) with logarithmic x-scale accentuates the gap between the basaltic and ultramafic populations.

Figure 7: Magnesium (MgO wt%) and Ni (ppm) distribution in Archaean metavolcanic rocks. Panel (A) shows linear histogram of a MgO on all studied cratons with four vertical lines denoting the preferred populations from kernel density function statistics. Panels (B) to (F) all show data after logarithmic translation. Panel (B) displays Ni distribution in all compiled Archaean metavolcanic rocks, highlighting the dominant basaltic population at ca. 200 ppm and the ultramafic population centred over ca. 2,000 ppm. Panels (C) and (D) show a comparison in MgO distribution between 2.5-2.9 and >2.9 Ga metavolcanic samples. Panels (E) and (F) show the same kind of comparison for Ni distribution.
Figure 8: Stratigraphic column (panel A) of the Abitibi greenstone belt after Ayer et al. (2002). Seven volcanic dominated assemblages are unconformably overlain by two sediment-dominated assemblages at the top. Column (B) shows approximate position of mafic to ultramafic metavolcanic samples interrogated by Sproule et al. (2002) and Herzberg (2016) as well as sedimentary samples described by Feng and Kerrich (1990) and Barrie (2005). Column (C) shows evolutionary trend of plutonic samples with time recorded in the Kenogamissi complex (after Benn, 2004), starting with tonalite and grey gneisses, moving to granodiorite, biotite-bearing granodiorite and ending with proper granite.

Figure 9: Major and selected REE systematics in Abitibi and adjacent greenstone belt high-Mg basalts, picrites and komatiites (all data from Sproule et al., 2002). Panel (A) shows a histogram of MgO (wt%) content of all least-altered samples. Panels (B) and (C) show MgO (wt%) vs FeO (wt%) and SiO$_2$ (wt%) systematics, respectively. The samples are grouped into Al-undepleted, Al-depleted and Ti-depleted suites according to the Al$_2$O$_3$/TiO$_2$ criteria >25, 15-25, <15. Shown for reference are the position of bulk continental crust (from Rudnick and Gao, 2003) and the preferred modelled parental komatiite melts by Herzberg (2016) also shown on figure 5. Panel (D) plots Al$_2$O$_3$/TiO$_2$ versus CI-chondrite (after Anders and Grevesse, 1989) normalised Sm/Yb. Panels (E) and (F) show MgO (wt%) vs SiO$_2$ (wt%) vs FeO (wt%) systematics separated into 6 stratigraphic assemblages. Panels (G) and (H) show MgO (wt%) vs SiO$_2$ (wt%) systematics separated into 6 stratigraphic assemblages (same symbols as panels (E) and (F)).

Figure 10: Selected geochemical systematics of high-P experiments relevant to komatiite petrogenesis. Panels (A) and (B) show dependence of Al$_2$O$_3$ and TiO$_2$ concentration, respectively, on extent of melting for experiments performed by Takahashi et al. (1993) and Walter (1998) on slightly different lherzolite starting compositions (also shown). Panels (C) and (D) show dependence of the Al$_2$O$_3$/TiO$_2$ ratio of liquids on Al$_2$O$_3$ and TiO$_2$ content, respectively. Same symbols as panel (A). Panel (E) is a histogram of Al$_2$O$_3$/TiO$_2$ ratios in low (<20%) and higher (>20%) degree liquids from fertile lherzolites (same data sources). Also shown are reconstructed hypothetical partial melts from refractory, previously melted harzburgite listed in Table_S2. See text for explanation. Panel (F) shows SiO$_2$ vs MgO relationships for three examples of calculated melts from refractory mantle. Blue and green colours are from 3 and 7 GPa experiments of Walter, purple from 4.6 GPa experiment of
Takahashi et al. (1993). In each case a higher and lower degree batch melt from fertile mantle is shown with diamond and circle symbols. The hypothetical refractory melt is represented by the square symbol and corresponds to the composition of the liquid required to move from the lower degree to the higher degree melt along the tie line. The distances reflect the lever rule to preserve mass. Panel (G) shows the high-Mg basalts, picrites and komatiites from (Sproule et al., 2002) in SiO$_2$ vs MgO space with superimposed model compositions (batch and accumulated fractional melts from Herzberg (2016)) and liquids of experiments by Takahashi et al. (1993) and Walter (1998). Panel (H) shows the same data and model but with calculated hypothetical refractory melts (see examples in panel F) from previously depleted mantle. Symbols are colour coded for pressure. Note that refractory mantle melts can reach very high MgO values with highly variable SiO$_2$ content, reflecting exhaustion of pyroxene.

Figure 11: Transmitted light section scans of five garnet peridotites from the Bultfontein pans (Kimberley, Kaapvaal craton) arranged by decreasing petrographic evidence for garnet exsolution (note that these are over-thick thin sections). From the top, sample BP002 (Tomlinson et al. 2018, this study) with regular garnet lamellae in orthopyroxene; 17BSK069d with semi-regular garnet lamellae and lines of equant garnet in a single large (32 mm) orthopyroxene; CLA18 with large (~10 mm) orthopyroxene crystals, where garnet elongate and aligned is located within or adjacent to the orthopyroxene; granular peridotite CLA05, in which garnet occurs as inclusions within and adjacent to coarse (<5mm) orthopyroxene; and garnet harzburgite 17BSK051 (this study) in which garnet grains are spatially associated with coarse orthopyroxene.

Figure 12: Transmitted light scan of garnet harzburgite 17BSK051 (A) and three phase maps. Panel (B) shows a composite map with olivine in green, orthopyroxene in blue, garnet in red and phlogopite in purple. Panels (C) and (D) show isolated maps of garnet with olivine and orthopyroxene, respectively. See text for explanation.

Figure 13: CI-chondrite-normalised REE patterns (Andres and Grevesse, 1998) (panels A-F) and Onuma-type diagrams (panels G and H). Panels (A) and (C) show garnet and orthopyroxene data for megacryst BP002 and (B) and (D) equivalent data for granular harzburgite 17BSK051. Green coloured symbols denote analyses least affected by contamination and/or alteration. Red coloured symbols show variably compromised analyses.
In BP002, intergranular (black in transmitted light) contaminant (likely kimberlite) was also analysed and this was used to calculate mixing models with average most depleted garnet (panel E) and orthopyroxene (panel F). Very good coincidences with average contaminated analyses by admixing between 0.1% and 0.12% contaminant. Panel (G) shows precursor phase (blue) REE concentrations calculated from modal relative abundances of garnet (red) and orthopyroxene (green) in an Onuma-type diagram for BP002. Panel (G) shows mean most depleted garnet (red) and orthopyroxene (green) of 17BSK051 in an Onuma-type diagram.

Figure 14: Geotherms calculated by Ballard and Pollack (1988) for the Kaapvaal craton. Panels (A) and (B) depict central cratonic geotherms from models 'Present 2' (P2) and 'Archaean 2' (A2), respectively. (A) In model P2, the reduction in central cratonic heat flow to 40 mW m\(^{-2}\) was achieved with a lithospheric root of 220 km depth and a reduced internal lithospheric heat production. (B) Model A2 shows the same 400 km diameter craton but at 3.2 Ga with 2.5 x higher internal heat production and the mantle potential temperature 200ºC hotter than at present (stippled line is model P2 for reference). The higher internal heat production results in a Moho temperature 300ºC hotter than in the present-day model and the higher mantle potential temperature elevates the mantle root geothermal overall. To achieve a similar geotherm as P2 with the higher internal heat production would require dropping the 3.2 Ga mantle potential temperature 200ºC relative to present day. Panel (C) shows an alternative model for the Archaean (A2'), in which the craton is 1.5x larger in diameter than A2 and the root extents to 500 km instead of 220 km. This results in similar temperatures as present-day at depths of 120-180 km, the interval where most studied xenoliths derive from but maintains a high Moho temperature.

Figure 15: Schematic model illustrating growth and evolution of cratonic lithosphere from the Mesoarchaean (A) and the latest Neoarchaean (B) into the post-Archaean (C). In the Mesoarchaean, ancient non-convecting domains (ANCD) existed in the upper mantle as remnants of cumulates that crystallised from magma oceans and seas and from very deep melting in hot upwellings. The ANCDs are not envisaged to have been a global barrier but where they were extensive, heat flow from the lower mantle was impeded. A nascent craton with a modest mantle root is shown forming above an ANCD. Occasional upwelling of heat began to erode through the ANCD and lower-mantle derived melts may have metasomatised the cumulates. By the late Neoarchaean, the NACDs had been dislodged by repeated pulses
of upwellings form the lower mantle. The majority of the barrier was recycled back into the
asthenosphere but occasional fragments and melt residues from the NACD became entrained
into the Neoarchaean 'superplumes' which fused this material with the shallower proto-
cratonic root. In post-Archaean events, the base of the lithosphere began to erode and shallow.
Petrological, mineralogical and geochemical peculiarities of Archaean cratons

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Abstract

The most outstanding features of Archaean cratons are their extraordinary thickness and enduring longevity. Seismically, Archaean cratonic fragments are sharply-bounded deep roots of mechanically strong, cold lithospheric mantle, clearly distinguishable from non-cratonic lithosphere. Rhenium-depletion of deep cratonic xenolith whole rocks and sulphide inclusions in diamond indicate that melting was broadly coeval with formation of the overlying proto-cratonic crust, which was of limited mechanical strength.

A very important process of proto-cratonic development was vertical crustal reorganisation that eventually yielded a thermally stable, cratonised crust with a highly K-U-Th-rich uppermost crust and much more depleted deeper crust. Clastic sedimentary rocks available for geochemical study are predominantly found in the youngest parts of supracrustal stratigraphies and over-represent the highly evolved rocks that appeared during cratonisation. Vertical crustal reorganisation was driven by crustal radiogenic heat and emplacement of proto-craton-wide, incubating and dense supracrustal mafic and ultramafic volcanic rocks. Statistical analysis of these cover sequences shows a preponderance of basalt and a high abundance of ultramafic lavas with a dearth of picrite. The ultramafic lavas can be grouped into Ti-enriched and Ti-depleted types and high pressure and temperature experimental data indicate that the latter formed from previously depleted mantle at temperatures in excess of 1700°C.

Most mantle harzburgite xenoliths from cratonic roots are highly refractory, containing very magnesian olivine and many have a high modal abundance of orthopyroxene.
High orthopyroxene mode is commonly attributed to metasomatic silica-enrichment or a non-pyrolitic mantle source but much of the excess silica requirement disappears if melting occurred at high pressures of 4-6 GPa. Analysis of experimental data demonstrates that melting of previously depleted harzburgite can yield liquids with highly variable Si/Mg ratios and low Al₂O₃ and FeO contents, as found in komatiites, and complementary high Cr/Al residues. In many harzburgites, there is an intimate spatial association of garnet and spinel with orthopyroxene, which indicates formation of the Al-phase by exsolution upon cooling and decompression. New and published rare earth element (REE) data for garnet and orthopyroxene show that garnet has inherited its sinusoidal REE pattern from the orthopyroxene. The lack of middle-REE depletion in these refractory residues is consistent with the lack of middle- over heavy-REE fractionation in most komatiites. This suggests that such pyroxene or garnet (or precursor phases) were present during komatiite melting. In the Kaapvaal craton, garnet exsolution upon significant cooling occurred as early as 3.2 Ga and geobarometry of diamond inclusions from ancient kimberlites also supports cool Archaean cratonic geotherms. This requires that some mantle roots have extended to 300 to possibly 400 km and that early cratons must have been much larger than 500 km in diameter.

We maintain that the Archaean-Proterozoic boundary continues to be of geological significance, despite the recognition that upper crustal chemistry, as sampled by sedimentary rocks, became more evolved from ca. 3 Ga onwards. The boundary coincides with the disappearance of widespread komatiite and marks the end of formation of typical refractory cratonic lithosphere. This may signify a fundamental change in the thermal structure of the mantle after which upwellings no longer resulted in very high temperature perturbations. One school of thought is that the thermal re-ordering occurred at the core-mantle boundary whereas others envisage Archaean plumes to have originated at the base of the upper mantle.

Here we speculate that Archaean cratonic roots may contain remnants of older domains of non-convecting mantle. These domains are potential carriers of isotope anomalies and their base could have constituted a mechanical and thermal boundary layer. Above laterally extensive barriers, emerging proto-cratons were protected from the main mantle heat loss.

The eventual collapse of these mechanical barriers terminated very high temperature upwellings and dismembered portions of the barrier were incorporated into the cratonic mantle during the final Neoarchaean 'superplume' event. The surviving cratons may therefore preserve biased evidence of geological processes that operated during the Archaean.
1. Introduction

The word *kratogen* was first used in the German geological literature to distinguish stable continental crust from orogenic crust that had been involved in mountain-forming processes. The term derives from the Greek word *krátos*, meaning strength or power, and was later shortened to *Kraton* before being accepted into English usage as *craton*. There is wide agreement that a craton represents a strong, thick and coherent piece of continental lithosphere that has maintained long-term tectonic stability and traditionally, cratons have been viewed as nuclei of continents. Before the availability of vast quantities of seismic data and their computational analysis, cratons were geologically and topographically recognised as forming flat shield areas comprised of exposed crystalline basement or stable platform sediment successions, typically of early Precambrian age.

Global seismic tomography is one powerful method for differentiating the roots of cratons because the mantle portion of cratons is characterised by very fast travel speeds of seismic waves. The seismic architecture of the lithosphere and surrounding asthenosphere can be defined by comparing the SH body-wave velocity to a standard model (e.g. Grand, 1994). Cratonic lithosphere roots below most exposed Archaean shields have an average SH body-wave velocity 2-3% faster than reference models (e.g. Begg et al., 2009) whereas Proterozoic lithospheric mantle is characterised by SH body-wave velocity only 1% faster (e.g. Lebedev et al., 2009). Faster travel speeds are primarily the result of substantially lower temperature than adjacent convecting asthenosphere and only secondarily the result of compositional difference (e.g. Griffin, 1999), the maximum likely contribution of which being 1/4 of the actual observed velocity difference (Schutt and Lesher, 2006).

In their analysis of the lithospheric structure of the African continent, Begg et al. (2009) demonstrated that it is possible to clearly differentiate and visualise the Archaean cratonic mantle roots from the rest of the African continent. Figure 1 (annotated and reproduced from Begg et al.’s (2009) figure 10) shows a view from the deep mantle transition zone in the southern Atlantic, looking upward in a NE direction towards Africa. The volumes of red material (showing SH body-wave velocity >1.9%) clearly delineate the sharply bounded roots of the three broad Archaean cratons within Africa: West Africa, Congo (including Tanzania), and southern Africa (mostly the Kalahari craton).

On the global shear speed structure maps of the upper mantle of Schaeffer and Lebedev (2013), most cratonic mantle roots are seen extending to at least 150 km depth (their figure 11) with deviations of SH body-wave velocity as high as +6%, requiring ambient
temperature of less than 1,000°C. Because density of mantle peridotite is an inverse function
of temperature, cratonic roots at such low temperatures can only maintain neutral buoyancy
relative to oceanic asthenosphere due to their extremely refractory chemistry, essentially
exhausted of spinel and clinopyroxene, depleted in garnet and rich in low density Mg-rich
olivine (e.g. Schutt and Lesher, 2006). Thus, the buoyancy of cratons and the fast SH body-
wave velocity in cratonic mantle roots together are very strong evidence for extreme (40% or
more) melt depletion in Archaean cratonic roots.

A very pleasing completely independent confirmation of this geophysical picture of
Archaean cratonic mantle roots has emerged from petrological and geochemical study of
mantle xenoliths. Multiple lines of evidence show that deep cratonic xenoliths, representing
samples from between ca. 80 and 250 km root depth (e.g. Kopylova and Caro, 2004), mostly
experienced very high degrees of melt extraction. Perhaps the best line of evidence is the
composition of the predominant peridotite phase olivine, whose Mg content is most strongly
influenced by the extent of melt depletion (e.g. Baker and Stolper, 1994). Compared to
olivines from oceanic, abyssal peridotites with average forsterite contents of 90.5 to 91.0%,
those in ancient cratonic harzburgites are much more magnesian, averaging between 92.5 and
93.0% forsterite component (e.g. Pearson and Wittig, 2008). Available experimental data
indicate that these compositions require that the host peridotites experienced extraction of 35-
45% liquid (see Bernstein et al, 2007 and Pearson and Wittig, 2008) through repeated melting
events.

After such extensive extraction of melt, the residue ends up vastly depleted in
incompatible elements. Because Re is much more incompatible than Os, ancient harzburgitic
peridotites develop retarded, very unradiogenic Os-isotope compositions (e.g. Walker et al.,
1989) that can be used to calculate a Re-depletion age. When initially discovered, this
demonstrated that at least for the Kaapvaal craton, melt extraction must have occurred prior
to 2 Ga. In more recent studies, peridotitic sulphide inclusions in diamonds were analysed
and these have yielded even older Re-depletion ages. For example: 3.1 to 3.5 Ga for the
Siberian craton (Pearson et al., 1999); 3.4 Ga for the southern Zimbabwe craton (Smith et al.,
2009); and ca. 3.3 Ga for the Kaapvaal craton (e.g. Shirey and Richardson, 2011). In the
Slave craton, Aulbach et al. (2004) retrieved monosulphide solution inclusions with
Mesoarchaean Re-depletion ages from peridotitic olivine xenocrysts. A subset of the
analysed inclusions form a 3.4 Ga isochron, attesting to the extreme antiquity of the deep
residual lithosphere there. Finally, Archaean mafic to ultramafic intrusions from the
Zimbabwe craton contain extremely unradiogenic Os that Naegler et al. (1997) interpreted to
indicate the beginning of cratonic root depletion from 3.8 Ga onwards, coincident with the oldest known zircon crystals known from this craton (Bolhar et al., 2017).

In summary, the overwhelming petrological evidence from harzburgite xenoliths combined with Re-Os isotope systematics indicates ancient melt extraction events consistent with the presently cool and refractory nature of Archaean cratonic roots inferred from seismology. The lithospheric mantle root of some cratons was subsequently lost or eroded (e.g. Miller et al., 2012) and has in many places been modified (e.g. Griffin et al., 2003) and cannot be directly compared with the root that was in place during the Archaean.

Complementary observations are made on the proto-cratonic surface where Archaean mafic rocks are clearly much more magnesian than those erupted after 2.5 Ga (e.g. Keller and Schoene, 2012). Albeit not the volumetrically dominant component of greenstone belts (e.g. Sproule et al., 2002), komatiite with between ca. 20-35 wt% MgO is known from most cratons and where greenstone belt stratigraphy is well understood (e.g. from komatiite-related Ni-ore exploration; Lesher and Barnes, 2009), and multiple episodes of komatiite emplacement over periods lasting ca. 50 Ma are evident (e.g. Thurston et al., 2008). There is an on-going debate regarding the type of melting that caused komatiite formation, ranging from dry, very hot and deep 'plume' type melting (Berry et al., 2008) to aqueous fluid fluxing in (Parman et al., 1997) and away from subduction zones (Sobolev et al., 2016) with intermediate models envisaging more complex fluid fluxing (Herzberg, 2016). Regardless of the melting mechanism, komatiite requires advanced stages of peridotite melting that consumed olivine and orthopyroxene (Nesbitt et al., 1979), an observation that at least qualitatively complements the highly refractory nature of cratonic harzburgite. The strong agreement about the general (albeit not total) disappearance of komatiites at the end of the Archaean is also consistent with the fact that, on average, Archaean cratonic roots have faster SH body-wave velocities (e.g. Schaeffer and Lebedev, 2013) than Proterozoic mantle lithosphere and that Archaean harzburgites are much more strongly depleted than Proterozoic equivalents (e.g. Griffin et al., 2003).

The relatively sudden (in the context of the vast Precambrian time span) demise of komatiite production at the Archaean-Proterozoic (A-P) boundary was accompanied by other notable disruptions in secular trends, which collectively may have relevance for understanding how Archaean continental lithosphere differed from younger continents. These include: the evolution of the maximum MgO content of highly magnesian magmas, which remained almost constant throughout the Archaean but then dropped (Campbell and Griffiths, 1992); several general changes in the nature of mafic and felsic magmas emplaced towards
the end of the Archaean (Keller and Schoene, 2012); pronounced changes in the composition of clastic (Condie, 1993) and hydrogenous sediments (Konhauser et al., 2009) across the A-P boundary; a jump in the O-isotope composition of zircon (Spencer et al., 2017); and a sharp shift in triple-O-isotope composition of clastic sedimentary rocks (Bindeman et al., 2018).

Condie (1993) demonstrated that the Archaean land surface had a much higher abundance of komatiite and basalt than would be estimated from outcrop areas at the present-day exposure level. Archaean clastic sedimentary rocks have very high relative abundance of Co, Ni and Cr whereas Proterozoic sedimentary rocks do not significantly differ from Phanerozoic equivalents (Figs. 2a,b). Most recently, Tang et al. (2016) used the Ni/Co ratio in clastic sedimentary rocks as a proxy for MgO content with which it was possible to also show the dramatic transition from mafic to felsic upper continental crust (UCC) during the Neoarchaean (see Fig. 2c).

Notwithstanding the harmonious overall picture of Archaean continental build-up and mantle melting, it turns out that the reconciliation of the seismic information from cratonic mantle roots with petrology of Archaean harzburgite and komatiite fails in terms of persistent deviation of mineralogical and geochemical detail from model prediction (e.g. Walter, 1998; Bédard, 2006; Pearson and Wittig, 2008). Furthermore, current models for Archaean craton formation struggle to achieve mass balance and have limited success explaining the co-occurrence of komatiite with the more dominant tholeiitic basalts. There is the additional issue that Archaean cratonic mantle roots were apparently already sufficiently cold to permit diamond formation (Boyd et al., 1985; Ballard and Pollack, 1988; Kaminsky et al., 2002; Miller et al., 2012) yet for the most part, the Archaean crust lacked mechanical strength and experienced repeated re-melting events. Thus, the Archaean continental lithosphere apparently existed as a proto-cratic composite of a strong deep mantle root with a weak crustal lid before complete lithospheric stabilisation (i.e. cratonisation) that typically, but not exclusively, happened towards the end of the Archaean.

Unless these discrepancies and apparent mutual inconsistencies can be resolved, there is limited hope that the full geological and tectonic significance of Archaean cratons can be reconstructed. The aims of this paper are to review discrepancies that have long been known within the specialist disciplines for a wider geochemical readership, to synthesise mutual inconsistencies and to provide perspectives on possible common solutions.
2. Brief review of some pertinent problematic aspects of Archaean mantle melting and lithospheric mantle mineralogy

2.1 Incompatible element mass balance

Jordan (1975, 1978) developed pioneering models for cratonic stabilisation, in which he first clearly demonstrated the need for depleting the mantle portion of stable shield lithosphere through basalt extraction into the crust. The resulting root possesses the required physical characteristics to make it more refractory, less dense and rheologically distinct from the surrounding asthenosphere. Since these early models, many lines of evidence have emerged supporting the view that the continental crust and the depleted mantle portion of cratonic lithosphere share a very long common history, including melt depletion events coeval with crust formation pulses (e.g. Pearson et al., 1995; Naegler et al., 1997). However, the elegance of the basalt extraction hypothesis is compromised by the problem of mass balance.

The issue is that the preserved cratonic mantle root alone cannot nearly have supplied all the incompatible trace elements present in the Archaean crust (e.g. Arndt et al., 2002; Carlson et al., 2005) as can be illustrated with the example of the highly incompatible radioactive heat producers U, Th and K. In average continental crust these are $62\pm2$ times enriched over primitive mantle. This ratio is quite precisely known from heat flow data and continental composition estimates (Rudnick and Gao, 2003), primitive mantle U and Th concentrations and terrestrial Pb-isotope composition (Sun and McDonough, 1989), and the canonical Earth K/U ratio (Jochum et al., 1983). The enrichment ratio is somewhat lower ($45\pm2$) for average continental crust of Archaean age as the crustal column generates a lower average modern heat flow of 41 mW m$^{-2}$ on cratonic provinces (Michaut et al., 2009).

Nevertheless, a 165 km deep cratonic mantle root (yielding a 200 km deep craton) only contains 5.8 times the mass of a 35 km thick crust, not the required $45\pm2$ times. This means that unless the precursor mantle was much more enriched than primitive mantle, the undifferentiated rock mass contained in a craton could only have provided ca. 15-20% of the inventory of the very highly incompatible elements.

Therefore, Archaean crust formation must also have tapped into magmas that originated from the convecting asthenospheric mantle (Arndt et al., 2002). Such magmas could have travelled through the growing Archaean continental lithosphere (e.g. Carlson et al.,...
Regardless, the discussion of Archaean mantle source regions must distinguish between the ultra-depleted cratonic roots and the more modestly depleted convecting asthenosphere.

2.2 Mismatch between predicted and observed mineral modes in cratonic harzburgites

At relatively low pressures (1-3 GPa), melting experiments of fertile mantle compositions predict that the residue after 25-30% melt extraction should be devoid of clinopyroxene and Al-phase (spinel and/or garnet) and consist of >80% olivine (Baker and Stolper, 1994; Walter, 1998) and that at 40% degrees of melting, the residue should be nearly mono-mineralic dunite. At higher pressures, garnet can remain in the residue at up to 40-50% melt extraction. Because the Fo-content of olivine increases approximately linearly with the extent of melt extraction (Pearson and Wittig, 2008), a strong correlation between modal olivine content and Fo-component is expected. This relationship was prominently described for oceanic peridotites by Boyd (1989) and is shown in figure 3a. Boyd (1989) next compared this oceanic trend with data from post-Archaean subcontinental lithosphere brought to the surface by off-craton kimberlites. The comparison confirmed that, to a first order (Fig. 3b), the data followed the oceanic trend. However, harzburgites from the Archaean Kaapvaal craton (Fig. 3c) were found to deviate markedly from the trend, plotting at higher Fo values (92-93%) but containing only 55 to 80% modal olivine instead of the expected dunitic abundance (>90%). Essentially, the harzburgites are much richer in orthopyroxene than expected and the resulting rock has an unusually high Si/Mg ratio (as enstatite is the most Si-rich of the common mantle minerals).

This mismatch between observed and predicted mineralogy is very strongly expressed in Kaapvaal harzburgites but it is also found in many other Archaean cratonic harzburgites with <60% modal olivine, for example from the Slave, Siberian and Wyoming cratons (e.g. Boyd et al., 1997; Eggler et al., 1987; Ionov et al., 2010; Kopylova et al., 1999; Kopylova and Caro, 2004; Newton et al., 2015; Regier et al, 2018; Solovjeva et al.,1994). Xenolith populations of only few kimberlites, particularly from the North Atlantic craton (Greenland; e.g. Bizzarro and Stevenson, 2003; Bernstein et al., 2006), are dominated by dunite as expected from Boyd's (1989) trend, but with generally high Fo-component. On average, high orthopyroxene abundance is more common and has been very widely discussed in the
literature (e.g. Bernstein et al., 2007) without finding wide petrological consensus. Proposed explanations include: the high Si/Mg ratio was inherited from the precursor mantle that may have differed from pyrolite due to crystal fractionation in a magma ocean (Herzberg et al., 1988); the high modal abundance of orthopyroxene reflects melt extraction at much greater depth (>8 GPa) than experienced by oceanic harzburgites (Canil, 1992; Herzberg, 1993); density sorting in plume melt residues (Arndt et al., 2002); later addition of Si, possibly from subduction zone metasomatism (Kesson and Ringwood, 1989) or serpentinisation (Canil and Lee, 2009); late Proterozoic within plate melt infiltration of hydrous melt (Wasch et al., 2009); reaction between depleted harzburgite either with foundering eclogite restite (Bédard, 2006) or eclogite-derived melts (Aulbach et al., 2011); and originally oceanic origin at a shallow very hot ridge (Rollinson, 2010). As can be appreciated, these various scenarios have substantially different ramifications for the early Earth.

A second unexpected feature of Archaean cratonic harzburgite is that it commonly contains garnet. High pressure experiments (e.g. Takahashi, 1986; Canil, 1991; Walter, 1998) show that at pressures exceeding 6 GPa, minor residual garnet can persist in harzburgite even after 40% melt extraction. However, it appears that garnet is not a rare accessory but a common phase in the Archaean cratonic mantle cross-section (Griffin et al., 1999; 2002) and this is at odds with the widely-held model that the refractory lithosphere grew by orogenic stacking of residues from low pressure melting (e.g. Bernstein et al., 2007; Canil, 2004; Rollinson, 2010; Lee and Chin, 2014). Indeed, Nixon et al. (1987) demonstrated that the common occurrence of garnets (with a characteristic sub-calcic chemistry) was restricted to xenocrysts and garnet-bearing xenoliths from kimberlites that intruded through Archaean cratonic lithosphere (Kaapvaal and Zimbabwe) whereas off-craton kimberlites erupted through Proterozoic geology were devoid of such garnets (see Fig. 4). It is possible that the overabundance of orthopyroxene and the dominance of garnet-bearing harzburgite are related through exsolution of garnet from orthopyroxene (e.g. Cox et al., 1987) and/or that garnet grew in response to metasomatism (e.g. Stachel, et al., 1998; Pearson and Wittig, 2008). Regardless, Sm/Nd and Lu/Hf isotope data show that garnet growth in many cratons occurred during the Archaean (e.g. Shu et al. 2013; Koorneef et al., 2017).

2.3 Seismic evidence for garnet in Archaean lithosphere and lithospheric buoyancy constraints
In the early long-range seismic refraction profiles, such as those conducted across continental North America (e.g. Green and Hales, 1968), it was noticed that there is an increase in impedance at depths between 60-80 km. Because impedance is the product of seismic velocity and density and since temperature is unlikely to decrease with depth, Hales (1969) proposed that this observation represented the spinel to garnet transformation. Many seismic studies have since found similar increases in impedance (e.g. Revenaugh and Jordan, 1991, Woods et al., 1991) but the observation is not generally accepted to reflect the spinel to garnet transition globally, although the observed extent of velocity change fits well with experimental constraints on the phase transition (Webb and Wood, 1986).

Whereas the detection of the impedance increase can reasonably be expected in the more fertile oceanic mantle, where it occurs near the modelled depth of 50-60 km, Revenaugh and Jordan (1991) commented that in continental lithosphere, basalt depletion should strongly diminish the strength of the reflector by reducing modal abundances of spinel and garnet. This should be particularly true for cratonic lithosphere where multiple lines of evidence suggest extensive melt extraction. However, it appears that the impedance increase is just as prominent in the lithosphere of several Archaean cratons, including the Baltic Shield, Western Australia and the Congo craton (Lebedev et al., 2009). Indeed, in the original work by Hales (1969) the strongest velocity increase was found in the Lake Superior region, underlain by Archaean lithosphere. In comparison with the oceanic realm, the increase in seismic velocity in cratonic regions occurs from the Moho down to depths of 100-150 km and is less sharp. Lebedev et al. (2009) explained this with the survival of spinel to greater depth due to increase in the Cr$_2$O$_3$ activity in the refractory residual harzburgite. Experimental data support stability of spinel to greater depth in Cr-rich bulk compositions, but these studies (e.g. Klemme, 2004; Ziberna et al., 2013; Ziberna and Klemme, 2016) also predict very low overall modal spinel abundances. Thus, an additional explanation for the increase of the shear wave velocity below cratons could be the appearance of ultra-depleted, sub-calcic and calcic garnet, which is often found to have equilibrium pressures in the region of 3-5 GPa (e.g. Gibson et al., 2013; Gibson, 2017), corresponding to depths of ca. 90-150 km. The 80-90 km range is also the depth for which Griffin et al. (2002) report the shallowest appearance of Cr-rich garnet macrocrysts in a number of Archaean cratons. Regardless of the importance of spinel, the seismic evidence apparently supports the empirical petrological evidence for garnet being an important constituent phase of many Archaean cratonic roots.

Although cratonic lithosphere is commonly thought of as being buoyant, this is only true in terms of composition but due to its much lower temperature than asthenosphere, the
cratonic root is, on average, only barely positively buoyant (e.g. Rudnick, 1994). In fact, Schutt and Lesher (2006) calculated that residual cratonic peridotite formed above 110 km may be negatively buoyant by the time it has cooled to the cratonic geotherm. These authors suggested that this could imply an inherent tendency for relatively shallow depleted residues to undergo vertical downward motion. A final poorly understood aspect of cratonic buoyancy is that there is a lack of evidence for proto-cratons having ever stood substantially elevated above sea level during their formation (Galer and Mezger, 1998; Bindeman et al., 2018). This is surprising particularly for cratonic fragments that formed rapidly (e.g. many on the subprovinces of the Superior craton; e.g. Thurston et al., 2008). At the solidus, the cratonic root density becomes substantially lower than that of fertile mantle (e.g. Ziberman and Klemme, 2015) and this would buoy the craton, yet entire stratigraphies of basalt and komatiite were apparently emplaced in water (e.g. Houlé et al., 2009) into environments with limited topography. This is also true for many of those demonstrably deposited ensialically through evolved continental crust (e.g. Chauvel et al., 1985, 1993; Blenkinsop et al., 1993). Many of these apparent inconsistencies may stem from the fact that in the Archaean, proto-cratons had not yet reached their mechanical rigidity, with much reduced strength in the middle and lower crust (Ashwal et al., 1987; Kramers et al., 2001; Benn and Kamber, 2009).

2.4 Experimental vs. empirical constraints on komatiite major element chemistry

At least since the experiments by Takahashi and Scarfe (1985) it has been clear that high degree, high temperature melting of fertile mantle at 150-200 km depth can produce liquids that are as magnesian as komatiite. Because komatiite is defined as an ultramafic volcanic rock with MgO >18 wt% and depleted in other oxides (e.g. Arndt et al. 2008), Takahashi and Scarfe (1985) were technically correct in relating their experimental melts to komatiite. However, there remain very significant issues with the widely held opinion that komatiites are very high degree melts. Arndt (1977a) pointed out that since mantle melts are mostly buoyant relative to refractory olivine, it would be physically impossible for early melts not to escape. Schmeling and Arndt (2017) showed that mantle melts are less dense than the main harzburgite phase olivine to a depth of 230 km, making it implausible that liquids would have pooled at 50-230 km source depths until 30-40% of melting. By contrast, it is possible to form liquids with >30 wt% MgO at modest degrees of melting from more refractory
peridotite that had previously been depleted or melting at greater depth (e.g. 14 GPa; Takahashi, 1986). At mantle transition zone depths, relative densities permit large degree melts to accumulate before eventual escape (Schmeling and Arndt (2017).

There are also issues regarding the finer details of major element composition of experimental versus empirical komatiite compositions (e.g. Walter, 1998). Many of these problems may relate to the inability of small-scale experiments to simulate the natural process of melting in large-scale thermal upwellings (e.g. Robin-Popieul et al., 2012).

Herzberg (2016) provided a recent discussion on Al-undepleted komatiite liquids. It was found that that at a given parental MgO content the observed SiO$_2$ contents for classic Neoarchaean Abitibi greenstone belt komatiites are too low by comparison with experimental liquids, certainly outside analytical error. The diagrams from Herzberg (2016) most relevant to this mismatch are reproduced in figure 5. Relative to an accumulated fractional melting liquid with 30 wt% MgO, the observed liquid line of descent (LLD) has ca. 0.8 wt% too little SiO$_2$ (Fig. 5a). More significantly, in terms of FeO vs. MgO systematics, the empirical data favour a batch melt (Fig. 5b), which is ca. 1.7 wt% higher in SiO$_2$ at MgO 30 wt% than the empirical LLD (Fig. 5a). If these komatiites are representative, then either the melting models are in error, the source of komatiite was not pyrolitic, melting did not occur at dry conditions (Herzberg's (2016) preferred option), or a combination of several factors. Herzberg (2016) also explored to what extent the experimental liquids of Walter's (1998) experiments could match the empirical Neoarchaean Abitibi greenstone belt komatiites in MgO vs. SiO$_2$ space (his figure 3a). This comparison revealed that certain liquids from the highest pressure range runs (6 and 7 GPa) coincided with the empirical LLD but at MgO contents of 27-28 wt%, i.e. lower than the presumed 30 wt% MgO parental melt deduced from melt inclusions (Sobolev et al., 2016).

In addition to the MgO/SiO$_2$ mismatch, Walter (1998) commented on a paradox concerning the depth of melting. When plotting empirical data and experimental melting contours in CaO/Al$_2$O$_3$ vs. Al$_2$O$_3$ space (in which olivine crystallisation causes predictable vertical LLD), Walter (1998) found (his Fig. 7a) that early Archaean komatiites could be generated at 8-10 GPa with up to 30% melting and that Neoarchaean komatiites at 7 GPa with 30 to >50% melting. However, these samples have flat chondrite-normalised rare earth element (REE) patterns, which apparently precluded garnet as a stable phase in the source. Walter (1998) commented (p. 49): "This creates a paradox"; in that major elements (CaO/Al$_2$O$_3$) require garnet on the solidus yet REE do not show middle (M) and light (L) enrichment over heavy (H) REE expected from residual garnet. In terms of Sm/Yb or Gd/Yb,
komatiites show a surprising lack of strong fractionation. Walter (1998) speculated that the mantle of at least some of the cratons (e.g. Kaapvaal) could have had a major and trace element composition different from pyrolite. The melting depth conundrum was further developed by Canil (2004), who pointed out that the high Cr/Al ratio of cratonic harzburgites was difficult to reconcile with melt extraction deeper than 3 GPa. One proposed solution was that the cratonic lithosphere grew by stacking and thickening.

Studying FeO/MgO systematics of cratonic peridotites, Lee and Chin (2014) proposed that most Archaean mafic magmas could be sourced by melting mantle between 2 and 4 GPa at temperatures of 1500 to 1650°C. Because nearly all their studied peridotites yielded higher equilibration pressures and lower equilibration temperatures, these authors also advocated vertical foundering and orogenic thickening as the key process for craton growth. However, Lee and Chin (2014) explicitly filtered against orthopyroxene-rich peridotite samples by (p. 277) "excluding those samples with anomalously high SiO₂ for a given Mg#", on the basis that these were considered to implicitly reflect metasomatic Si-addition.

In summary, there are clearly recognisable patterns in most proposals that attempt to reconcile experimental with empirical petrological and seismic observations regarding Archaean subcontinental lithosphere formation. Many of these relate to the minerals orthopyroxene and garnet both of which are apparently more abundant in Archaean cratonic lithosphere than available experimental constraints permit. Whereas many ancient cratonic harzburgites are too rich in SiO₂ for their MgO content or Mg#, many komatiites seem to suffer from the opposite problem. Finally, while major element systematics require garnet in the source of many komatiites, their REE patterns do not show the expected deficit in HREE. The main aim of this paper is to explore whether there could be a common solution to the issues of Archaean craton formation.

3. Datasets, analysis and samples

3.1 Datasets and data analysis

We revisited four datasets relevant to Archaean craton formation. Firstly, we reanalysed major and compatible trace elements systematics of Archaean igneous rocks previously studied by Keller and Schoene (2012) and Kamber (2015). Specifically, we used the pre-compiled files from the GEOROC database for the following cratons: Baltic shield; Dharwar craton; Kaapvaal craton; North Atlantic craton; North China craton; Slave craton; Superior craton; Tanzania craton, Western Australian cratons; and Zimbabwe craton. These were
selected to include cratons for which geochemical and petrological datasets also exist for
peridotites, because they host classic komatiite exposures and for which statistically
significant datasets exist. Each dataset was filtered to only include extrusive igneous rocks
for which whole rock analyses are reported. The analysis of the distribution of MgO and Ni
contents of Archaean volcanic rocks was conducted with kernel density estimation (KDE),
which is a robust method to make statistical inferences on population distributions of a finite
data sample. The method lends itself to estimate the number of modes in a population
(Silverman, 1981) and is commonly used to analyse detrital U/Pb zircon and other age
populations (e.g. Petrus et al., 2016). The programme used in this study was DensityPlotter
(Vermeesch, 2012).

A second dataset that was revisited separately is that for komatiites and komatiitic
basalts from the Neoarchaean Abitibi and Pontiac Subprovinces (Superior Province) reported
by Sproule et al. (2002). This is a very comprehensive dataset, including analyses for the
classic Pyke Hill and Alexo samples. Representative analyses were tabulated in Sproule et al.
(2002) and only these are found in GEOROC. Here we used the full dataset (Mike Lesher,
pers. comm. 2017) to test whether, within one greenstone belt, temporal trends existed in
terms of Mg/Si systematics (e.g. Herzberg, 2016).

Global deep-time geochemistry datasets for shale are a rich source of information
regarding the evolution of the uppermost crust (e.g. Smit and Mezger, 2017; Bindemann et al.,
2018). Here we revisited the shale geochemistry compilation reported by Tang et al. (2016)
in their supplemental materials S1 and added data for Neoarchaean argillite (Barrie, 2005),
which is a very fine-grained sedimentary rock containing reduced C. This rock type is
extensively studied by exploration geochemists due to its utility in vectoring volcanogenic
massive sulphide (VMS) deposits but these data rarely find their way into peer-reviewed
publications.

The final datasets revisited are the experimental ultramafic liquid compositions and
modal abundances reported in Walter (1998) and Takahashi et al. (1993). Additional highly
valuable experiments have been conducted (e.g. Takahashi and Scarfe, 1985; Takahashi,
1986; Canil, 1991; 1992; Takahashi et al., 1993; Baker and Stolper, 1994; Longhi, 2002;
Schutt and Lesher; 2006) but those conducted at high pressure did not report full modal
abundances of reaction products. We compared their phase relationships to those of Walter
(1998) and Takahashi et al. (1993) to test for consistency and representativeness.
3.2 Samples

As explained in the introduction and preceding section, the minerals orthopyroxene and garnet hold a critical role in Archaean craton formation (e.g. Gibson, 2017). In 2015 and 2017, we collected cratonic peridotites from the Bultfontein pans, Kimberley, Republic of South Africa (28.739155°S, 24.818094°E) with the aim of better documenting the mutual phase relations (e.g. Cox et al., 1987; Saltzer et al., 2001) between garnet and orthopyroxene and to better determine their interesting trace element geochemistry (Wasch et al., 2009).

Here we report data for two representative samples. Sample BP002 is dominated by a spectacular orthopyroxene megacryst of which a single crystal fragment measuring 15 x 12 x 7 cm is preserved within a granular garnet harzburgite. Tomlinson et al. (2018) reported a crystallographic and geochemical study for this sample. The other sample is a coarse-grained garnet harzburgite (17BSK051) with a high modal abundance of orthopyroxene typical of harzburgites from the Kimberley pipes (e.g. Boyd, 1989; Grégoire et al., 2003; Simon et al., 2007; Wasch et al., 2009). Analytical details can be found in the Electronic Supplement.

4. Results

4.1 The composition of the uppermost Archaean continental crust

Three prominent studies have investigated the temporal evolution of the composition of the continental crust with large lithogeochemistry compilation datasets (Keller and Schoene, 2012; Tang et al., 2016; Smit and Mezger, 2017). They have in common that they analysed time series of averaged geochemical parameters (e.g. MgO content, Ni/Co, Cr/Zn and Cr/U) of lithological groups such as mafic and felsic igneous and sedimentary rocks. By contrast, the purpose of the analysis performed here was to study the modal distribution of volcanic rocks that were erupted onto the various cratons in the Archaean.

Figure 6 shows histograms for MgO wt% distribution in volcanic rocks on 6 cratons for which >500 datapoints are available in GEOROC (panels A-F) and a combined histogram for the Zimbabwe, Tanzania, Slave and North China cratons (panel G) for which there are smaller datasets. For each dataset, MgO contents were re-calculated to a volatile-free basis for those samples (>90%) for which full major element data and loss on ignition are reported. The metadata are available in Table_S1. Data for the few (<<1%) rocks with >50 wt% MgO were excluded from further analysis on account of clearly being cumulates. Superimposed onto the histograms are KDE curves (blue solid lines). Sizes of sample populations,
A qualitative look at the histograms and the KDEs reveals a common pattern. There is a prominent population of evolved felsic volcanic rocks with MgO between 0.7 and 2 wt% and then a very prominent population with MgO between 5 and 10 wt%. These are separated by a strong gap. A compositional gap at similar MgO is well known in ocean island rocks (Chayes, 1963) and arc systems (e.g. Reubi and Blundy, 2009) where it has been attributed to differentiation and re-melting within the crust.

In all cases, the 5 to 10 wt% MgO basaltic population contains the largest number of samples. Moving towards higher MgO contents, the next common pattern is the paucity of samples with MgO in the 15 to 20 wt% range. Beyond 18 wt% MgO, all cratons show one or two broad populations of ultramafic volcanic rocks. With the exception of the Baltic shield data, where a population around 25 wt% MgO is quite prominent, clear ultramafic populations are not easily obvious. One reason it is difficult to test whether there is a dominant population in a linear histogram of MgO is that the ultramafic samples are naturally distributed over a large number of bins. This is because in an ultramafic parent rich in MgO, relatively limited olivine and pyroxene removal or accumulation will cause rapid change in MgO of several wt%, i.e. several histogram bins. By contrast, the same extent of crystal fractionation from a much lower initial MgO content in a felsic or basaltic magma will only spread the data across one or two bins. This is better appreciated when the data are subjected to a logarithmic translation of the x-axis values. The translation results in unevenly sized bin widths and separates data with low MgO content into more bins. By contrast, the ultramafic rocks are binned into fewer categories. The resulting histogram and KDE for the combined data from all cratons considered are shown on figure 6H. This clearly exposes the gap in erupted volcanic rocks with MgO between 15 and 20 wt% and emphasises the ultramafic population centred around ca. 28 wt% MgO. To our knowledge, this has not explicitly been reported before but has been hypothesised to exist based on MgO vs. siderophile element distributions by Arndt (1991).

The KDE method can also be used to statistically estimate the number of modes in a population (Silverman, 1981) but the method depends on the errors of each measurement. Because GEOROC does not report precision and accuracy of individual data, reasonable errors were assigned to data as follows: from 0.1-1.0±1; >1.0-5.0±0.2; >5.0±0.5 wt% MgO. When applied to the combined dataset from all cratons (see Fig. 7a), this method finds four prominent populations: one felsic with 1.0 wt% MgO accounting for 18.1% of the data; one
dominant basaltic with 6.0 wt% MgO containing 50.6% of the data; and two ultramafic at 21.2 and 35.7 wt% MgO, respectively, collectively accounting for 31.3% of the data. Similar calculations were performed for all individual datasets displayed on figure 6 (panels A-G) and results are listed in table 1. The preferred number of populations varied between three and five but in all cases, there is a felsic population at 1.2±0.59 wt% accounting for 18.6±7.0% of the data and a dominant basaltic population at 6.3±1.4 wt% MgO containing roughly half of the data (47.9±7.6). The next important population is ultramafic with 28.3±2.4 wt% MgO making up 21.9±4.1% of the data. It is found in all datasets but the North Atlantic craton. A high-Mg basalt population with 14.7±1.8 wt% is found on four cratons and on these it collectively contains 18.8±4.6% of the data. Finally, a highly ultramafic population with 39.0±3.8 MgO wt% (likely dominated by olivine cumulate rocks) is also found on four cratons, there containing 9.5±2.9% of the data.

It is not the intention to ascribe specific petrologic significance to these statistical analyses but the study of the histograms shows that the dominant Archaean volcanic rock was basalt and the second-most important volcanic rock type was ultra-mafic. The broadly komatiitic rocks are almost certainly over-represented in global datasets due to the understandable scientific interest in these rocks. However, the important finding of the paucity of picritic compositions (reflected in the absence of an identified population in the 15-20 wt% MgO range) cannot be an artefact of overly keen interest in komatiite, which would be difficult to distinguish from altered picrite or komatiitic basalt in the field or the core shed. Finally, the analysis also shows that Archaean basalt had nearly indistinguishable average MgO content and distribution as during the Phanerozoic (e.g. Kamber, 2015).

Because Ni content increases strongly with degree of melting and hence Mg content (Arndt, 1977b), the Ni concentrations of volcanic rocks of the same cratons was also compiled. Many published Ni analyses are performed by XRF on ignited powder and therefore concentrations were not corrected for loss on ignition. A total of 9,288 analyses were compiled (for metadata see Table S1) and of these 37 had Ni concentration greater than 5,000 ppm and were not further considered. Due to the huge range in Ni concentrations (i.e. 0.1 to 5,000 ppm), the linear histograms show very little detail and instead a logarithmic translation was performed (Fig. 7b). This very clearly shows two dominant populations representing basalt with ca. 200 ppm Ni and komatiite with ca. 2,000 ppm Ni. The felsic population is spread over many bins with a minor peak at ca. 20 ppm. Thus, the Ni distribution shows a similar pattern as the MgO distribution, highlighting the dearth of picritic compositions.
The final analysis was a comparison between rocks 2.5 to 2.9 Ga in age and those emplaced earlier. This was motivated by the proposal that there was no sharp A-P boundary but that geology instead changed in a transitional fashion from ca. 3 Ga onwards (e.g. Shirey and Richardson, 2011). More recently, Tang et al. (2016) and Smit and Mezger (2017) suggested that the UCC had started to become more felsic from 2.9 Ga onwards (see Fig. 2c) prompting an interest to study the chemistry of volcanic rocks emplaced either side of 2.9 Ga. For this comparison, data from the Kaapvaal, Zimbabwe, Dharwar, Superior and Western Australian cratons were separated according to preferred age stated in the original publications. These cratons expose a higher proportion of Meso- and Palaeoarchaean rocks than most others.

The comparison shows, both in terms of MgO (Fig. 7c, d) and Ni (Fig. 7e, f), that there is no obvious distinction between the two age groups, suggesting that the basaltic mode always dominated and that an equally ultramafic komatiite mode persisted right through the Archaean Eon. This is consistent with the observation of time-series of maximum inferred MgO contents of komatiites, showing no clear trend towards lower MgO with decreasing age (e.g. Campbell and Griffiths, 1992; Kamber, 2010). There is a small indication that picrite has become somewhat more common since 2.9 Ga but we note that this could be an artefact of the lower density of data for the >2.9 Ga age group.

4.2 Comparison of compositions of komatiites with predictions of melting models

4.2.1 A comparison of komatiite parental melt composition with experimental liquids and calculated melts of previously depleted mantle

It is widely argued that many observed komatiite liquid compositions are not easily reconciled with experimental liquids, calling for a variety of processes, such as fluid-induced melting (e.g. Herzberg, 2016) or ultra-deep melt segregation (e.g. Robin-Popieul et al., 2012) to explain the discrepancies. Komatiites are traditionally classified into Al-depleted and Al-undepleted based on CaO/Al$_2$O$_3$ and Al$_2$O$_3$/TiO$_2$ systematics (Nesbitt et al., 1979). In partly serpentinised samples, Al$_2$O$_3$/TiO$_2$ is more robust and samples with values <15 are classed as Al-depleted, whereas those with 15> Al$_2$O$_3$/TiO$_2$ <25 are termed Al-undepleted. Rarer samples with Al$_2$O$_3$/TiO$_2$ >25 are termed Ti-depleted. In view of the importance of the
Al₂O₃/TiO₂ ratio, we revisited the data from the Takahashi et al. (1993) and Walter (1998) experiments relating to the Al₂O₃ and TiO₂ systematics of experimental liquids as a function of melt fraction. These datasets provided chemical analyses for all experimental phases as well as modal abundance estimates. Importantly, their phase relations are compatible with more experiments that do not report modal abundance.

The first thing to note is that whereas Al-uptake into the melt is complex (Fig. 8a), Ti essentially behaves as an incompatible element with strong initial enrichment in low degree melts (Fig. 8b), regardless of pressure (see also Longhi, 2002). As expected for an incompatible element, at all degrees of partial melting, the Ti content in the liquid is higher than in the starting lherzolite. The two highest melt fraction liquids of Takahashi et al. (1993) are an apparent exception to this trend (Fig. 8b) but this is likely an artefact of rounding low concentration values to too few significant digits in the published data table. Regardless, when the Al₂O₃/TiO₂ ratio is plotted against Al₂O₃ (Fig. 8c) and TiO₂ (Fig. 8d) it is evident that TiO₂ exerts dominant control over the ratio (see also Robin-Popieul et al., 2012). The important inference arising from the experimental data is that an originally lherzolitic mantle source that became partially depleted (e.g. via extraction of 10-20% melt) will have a substantially higher Al₂O₃/TiO₂ ratio than primitive mantle (ca. 22) or the N-MORB source (ca. 25). If such a source was later remelted, it would yield liquids with very low Ti contents (<0.2 wt%) and very high Al₂O₃/TiO₂ ratios.

There is currently a lack of experimental komatiitic compositions with the very low TiO₂ concentrations found in some natural komatiites (e.g. Wilson, 2003). This very likely stems from a lack of successful experiments from refractory (harzburgitic) starting compositions. To gain a first-order understanding of what liquids from refractory mantle could be we calculated the differential melt compositions from isobaric series of the Walter (1998) and Takahashi et al. (1993) experiments assuming mass conservation (Table_S2). The validity of these calculations depends on the quality of the chemical data for the two batch melt liquids, the accuracy of the melt fraction estimate, and the stability of the phases at the liquidus. The results show (Fig. 8e) that all the hypothetical melts from variably refractory mantle have higher Al₂O₃/TiO₂ than the starting lherzolite. Hypothetical melts from very highly depleted mantle have very low TiO₂ concentrations, in many cases resulting in negative apparent TiO₂ values. These are artefacts of the cumulative uncertainties that could be removed if the Ti concentrations of high degree melts were analysed with dedicated long-acquisition time electron microprobe analyses (e.g. Donovan et al., 2011). Nevertheless, the low Ti concentrations (0.1-0.2 wt%) and the high Al₂O₃/TiO₂ ratios (25-200) compare well
with the ranges reported for Ti-depleted komatiites of the Abitibi greenstone belt (Sproule et al., 2002) and in so-called Al-enriched komatiites of the Barberton greenstone belt (e.g. Wilson, 2003; Robin-Popieul et al., 2012). Thus, at least some komatiites could have resulted from repeated 10-20% melting of a progressively more refractory source, overcoming the issue of requiring high-degree melts that would be too buoyant not to escape (Arndt 1977a; Schmeling and Arndt, 2017).

We adopted the same approach to explore whether melts from variably refractory peridotite could help to resolve the SiO$_2$ vs. MgO mismatch documented by Herzberg (2016).

Figure 8f shows three examples for 3, 4.6 and 7 GPa. The 3 GPa example is the composition of the melt required to take the 24% melt (experiment 30.14 of Walter, 1998) to 53% (experiment 30.11). It has 33.6 wt% of MgO and 45.7 wt% of SiO$_2$. The 4.6 GPa example shows the composition of the melt required to take the 22% liquid (experiment 1750°C of Takahashi et al., 1993) to 55% (experiment 1800°C). The 7 GPa example is the composition of the melt required to take the 16% melt (experiment 70.07 of Walter, 1998) to 47% (experiment 70.09). As can be seen by comparing panels (G) and (H) of figure 8, the hypothetical melts extend to higher MgO and more variable SiO$_2$ than the experimental liquids from fertile mantle and qualitatively illustrate that melting refractory peridotite could be one potential explanation for the Si/Mg and Al/Ti systematics of Archaean komatiites and picrites. Takahashi (1986) suggested that the range in SiO$_2$ values could reflect the presence of absence of pyroxene at the liquidus but these findings require further experimental and petrological exploration.

4.2.2 An extended comparison of Abitibi and Pontiac Subprovince komatiites with melting experiments of fertile mantle

The Neoarchaean Abitibi greenstone belt in Ontario and Quebec is a classic Subprovince of the Superior craton in which komatiites and mafic meta-volcanic rocks have been studied extensively. Because the greenstone belt apparently did not form onto pre-existing evolved crust, most of its igneous rocks preserve least contaminated mantle radiogenic isotope signatures (e.g. Ketchum et al., 2008) and are considered ‘juvenile’. An outstanding feature of the Abitibi greenstone belt is that across the nearly 700 km width of exposure, it displays a coherent vertical stratigraphy (e.g. Thurston et al., 2002) constrained with a very large number of high-precision U/Pb zircon dates from felsic volcanic rocks (e.g. Corfu, 1993). The huge wealth of information pertaining to the stratigraphic relationships is not widely
appreciated by the academic community, probably because most of the data support mineral exploration and are published in reports of geological survey organisations. Berger et al. (2011) explained that high-precision U/Pb zircon dates allowed the 55 rock assemblages recognised in 1991 to be simplified into the 9 assemblages shown in figure 9. The new stratigraphy comprises 7 assemblages dominated by volcanic rocks and an unconformably overlying set of two sedimentary rock assemblages. The 7 assemblages dominated by volcanic rocks are separated by condensed horizons (Thurston et al., 2008) composed of iron formations, chert and minor fine-grained siliciclastic sedimentary rocks (argillites).

The quality of stratigraphic control allows for an accurate assessment of petrological and potential temporal variety of komatiite within a greenstone belt. Komatiite occurs in four of the 7 lower assemblages (Fig. 9). The most widely known komatiites (Pyke Hill and Alexo) belong to the Kidd-Munro assemblage and have been extensively studied. Analysing major element systematics of these two occurrences, Herzberg (2016) established that batch melting produces a closer fit to observed MgO vs. FeO systematics than accumulated fractional melting (Fig. 5b) and furthermore, that compared to model predictions, the empirical data show a deficit in SiO₂ relative to MgO.

Here we revisit the much larger dataset of Sproule et al. (2002) who interrogated major and trace element geochemistry of 2,466 chemical analyses of high-Mg basalt, picrite and komatiite. Their sample suite includes representatives from all four ultramafic occurrences within the Abitibi greenstone belt as well as two younger occurrences in adjacent greenstone belts to the south (Pontiac Subprovince). They are age equivalent to the Blake and Timiskaming assemblages of the Abitibi greenstone belt and their chronologic positions are shown in figure 9. To achieve their wide stratigraphic and geographic coverage, Sproule et al. (2002) also analysed variably altered samples. After using the excessive loss on ignition criterion of Sproule et al. (2002) to avoid samples with strongly modified major element chemistry, 2,048 analyses remained.

We first tested whether this dataset also showed the relative dearth of picritic samples with MgO between 15 and 20 wt% by plotting the anhydrous rock MgO content in a histogram (Fig. 10a). As can be seen, the dominant population of data occurs around an MgO content of ca. 28 wt% with another prominent mode at ca. 40 wt% MgO, probably dominated by olivine-rich cumulates. The basaltic mode is not within the range of compositions studied by Sproule et al. (2002) whose dataset excluded most samples with MgO <12 wt%. However, the paucity of samples with MgO of 15-20 wt% is nonetheless clearly visible, confirming the picture from the preceding global analysis (Fig. 7).
In the context of this treatment, the most interesting phenomenon of the Abitibi greenstone belt ultramafic rocks is their great chemical variability. If the conventional komatiite classification criteria are applied to all rocks in Sproule et al. (2002) dataset (not just komatiites), 76% classify as Al-undepleted, 17% as Al-depleted and 7% as Ti-depleted types. Panels (B) and (C) of figure 10 show similar FeO vs. MgO and SiO\textsubscript{2} vs. MgO diagrams as figure 5. In this much larger dataset, it is evident that for any given MgO value there is a wide range in FeO (4-5 wt%) and SiO\textsubscript{2} (5-7 wt%) content. Contamination with continental crust and alteration are unlikely to explain the full spread in FeO vs. MgO space. Some of the most Fe-rich samples may contain excess accumulated chromite with or without olivine removal (see also Fig. 4b of Sproule et al., 2002) but even allowing for this process, a wide range in Mg\# remains. The FeO, SiO\textsubscript{2} and MgO systematics do not correlate in any way with the extent of Al- and Ti-depletion. In SiO\textsubscript{2} vs. MgO space (Fig. 10c), the more elevated SiO\textsubscript{2} values observed in some samples could be explained with continental contamination or post-emplacement chert infiltration (e.g. Thurston et al., 2012) but the sub-silicic samples remain unexplained, confirming the finding of Herzberg (2016) regardless of whether the melts are better approximated by batch or accumulated fractional melting models.

A smaller subset of samples also reports REE data for which CI-normalised Sm/Yb ratios were calculated (Sm was preferred over Gd as more samples had values for the former). When plotted versus the Al\textsubscript{2}O\textsubscript{3}/TiO\textsubscript{2} ratio (Fig. 10d), the paradox explained by Walter (1998) becomes apparent. Namely, only the most Al-depleted samples show somewhat elevated Sm/Yb ratios whereas for samples with Al\textsubscript{2}O\textsubscript{3}/TiO\textsubscript{2} from 10 to 50, there is no correlation, which is unexpected if garnet controlled the degree of Al-depletion or enrichment (e.g. Nesbitt et al., 1979).

The final four panels of figure 10 show the same data but now colour-coded for stratigraphic assemblage to test whether the large petrological variability could correlate with age. However, there is no age progression in FeO vs. MgO and SiO\textsubscript{2} vs. MgO systematics. Smaller datasets (e.g. Pacaud) show tighter systematics but in the three dominant subsets (Kidd-Munro, Stoughton-Roquemaure and Tisdale), the full range of major element chemistries is represented.

In summary, high-Mg basalts, picrites and komatiites from the Abitibi and adjacent Pontiac Subprovince represent very wide petrological variety, including melts from more fertile mantle and from refractory sources. These diverse komatiites occur at several stratigraphic levels but are volumetrically much inferior to the dominant basalts (Fig. 9).
Over the reasonably narrow ca. 60 Ma time window, multiple mantle sources melted, likely
at quite different depths.

4.3 Garnet and orthopyroxene textural relationships and REE systematics in
Archaean orthopyroxene-rich cratonic harzburgite

The early studies on the relationships between Archaean cratonic harzburgites and komatiites
established a paradigm of very deep (5-8 PGa) melting (e.g. Takahasi and Scarfe, 1985; Cox
et al., 1987; Boyd, 1989; Canil, 1992; Herzberg, 1993). This has since been replaced with the
currently prevailing idea (e.g. Canil, 2004; Lee and Chin, 2014; Regier et al., 2018) that
proto-cratonic lithosphere thickened by stacking of harzburgite, which represents shallow
melt residues (1-3 GPa). The paradigm shift was precipitated with the realisation that the
minor and REE systematics of komatiites, Archaean cratonic garnets (Stachel et al., 1998),
and of harzburgites (Canil, 2004) favoured melt extraction in the spinel stability field,
implying subsequent thickening of the cratonic root. The high Cr/Al ratio of many cratonic
harzburgites is quoted as very strong evidence against melt extraction deeper than 3 GPa, so
much so that deeper melting is no longer discussed as a viable alternative by some authors
(e.g. Regier et al., 2018). However, Tomlinson et al. (2018) offered observations that warrant
a new look at the old paradigm of deep melting. These authors used phase relationships in a
very coarse-grained orthopyroxene megacryst to postulate that most garnet in Archaean
harzburgite was not originally present but formed by exsolution from orthopyroxene. In other
words, during proto-craton formation the harzburgite was much hotter and only consisted of
olivine and orthopyroxene, even though it technically resided in the stability field of garnet
defined for pyrolitic mantle. Harzburgitic compositions can become garnet-free at the deep
solidus and the Cr/Al systematics of experimental olivine and orthopyroxene residues (e.g.
Walter, 1998) are compatible with the observed high Cr/Al many cratonic harzburgites at
pressures of 3-7 GPa, provided the residue is highly depleted. Below we provide more data
on the spatial association of garnet and orthopyroxene that has long been argued to contain
evidence about the cooling trajectory (e.g. Canil, 1991) of harzburgites and also document
much-needed REE concentration data for harzburgitic orthopyroxene.
4.3.1 The spatial relationship between garnet and orthopyroxene in garnet harzburgites

Cox et al. (1987) first systematically studied the spatial relationships of olivine, clinopyroxene, orthopyroxene and garnet in coarse-grained cratonic lherzolites. Their observations revealed one consistent phase relationship, namely a close spatial association of garnet and orthopyroxene. Because garnet was only found in association with orthopyroxene, they reasoned that garnet represented the Tschermak component of an original, higher-temperature orthopyroxene. Thus, garnet was a phase that exsolved from a parental higher-Al orthopyroxene upon cooling. Considering the experimental finding of Kushiro (1973), Cox et al. (1987) speculated that the more aluminous orthopyroxene precursors could have originated at pressures ranging from 4 to 8 GPa.

Using SEM-BSE, Saltzer et al. (2001) confirmed that the close spatial association of garnet and orthopyroxene also exists in more refractory harzburgites. They further discovered that those peridotites that had equilibrated at the greatest depth along the cratonic geotherm contain the most magnesian olivine. This observation does not in itself constrain the cooling trajectory, which Saltzer et al. (2000) suggested to have involved compression, a conclusion derived from comparison with experiments conducted on fertile peridotite compositions.

Tighter constrains on the cooling trajectory can be derived from analysis of spectacular examples of very coarse-grained orthopyroxenes that exhibit textural evidence for garnet exsolution. Gibson (2017) documented a range of garnet exsolution styles from very coarse-grained orthopyroxenes from the Tanzania and Kaapvaal cratons. A similar exceptionally well-preserved specimen of a large orthopyroxene single crystal (size of preserved relic 15 x 12 x 7 cm) from the Kimberley fields was studied by Tomlinson et al. (2018). Thermobarometry inferred equilibration at 4.4 GPa. By reconstituting the exsolved garnet (and, in some of the samples studied by Gibson (2017) additional clinopyroxene) into the orthopyroxene host, the Tschermak component of the original precursor pyroxene can be calculated and plotted as an isopleth into the garnet-orthopyroxene-olivine stability field of a model pseudosection. Provided that orthopyroxene formed on the solidus and that it was the only phase formed on the solidus, the original depth of formation can be obtained as the intersection of the isopleth with the solidus. In the cases of very large euhedral macrocrysts this assumption seems warranted and using this approach, Tomlinson et al. (2018) calculated that the precursor pyroxene of their studied sample formed at 5.9 GPa and 1,750°C, implying decompression upon cooling onto the cratonic geotherm. Tomlinson et al. (2018) noted that orthopyroxene with this Tschermak component could theoretically also exist on the very low
pressure solidus (0.3 GPa) within the plagioclase stability field but this is not considered a likely formation depth of any peridotites from the Kaapvaal craton (see also Lee and Chin, 2014).

It could be argued that the spectacular coarse-grained orthopyroxenes and their exsolved garnets are exotic curiosities with limited significance for the overall formation of the cratonic mantle. However, an important aspect of the specimen studied by Tomlinson et al. (2018) is that the megacryst is seen to disaggregate along its edges and that fragments of it, with garnet inclusions, are found in the enclosing harzburgite. Garnet and orthopyroxene in the harzburgite have exactly the same major and trace element composition as in the megacryst. The orthopyroxene carries substantial internal strain, a feature also noted by Gibson (2017), who attributed this to the mechanism of garnet exsolution but that could additionally have been caused during transport within the cratonic root. Considering that the peridotites apparently experienced vertical movement over tens of km, orthopyroxene disaggregation may have been a common process. This raises the possibility that a substantial proportion of garnet found in cratonic garnet harzburgites could have originated via exsolution, even if in most cases of granular peridotites, the exsolution process cannot be directly proven (Tomlinson et al, 2018). For example, Wasch et al. (2009) described conspicuous clots of coarse-grained orthopyroxene and garnet with some garnet visibly exsolved as linear bands within the orthopyroxene.

In figure 11, a series of section scans illustrates the progression from an example of an orthopyroxene megacryst with clear garnet exsolution lamellae to a 'normal' granular garnet harzburgite, with no obvious exsolution indication of the garnet. The intermediate examples are progressively less obvious cases for exsolution highlighting examples of garnet grains fully encased in orthopyroxene and relic alignment trails of garnet. A common feature of all samples (and most harzburgites from Kimberley) is the very coarse-grained nature of the orthopyroxene, often exceeding 10-15 mm in diameter. To further test the possibility that garnet in ordinary harzburgite originated by exsolution from formerly more aluminous orthopyroxene (e.g. Canil, 1991), we used automated phase mapping with large format SEM-EDS detectors to test the spatial relationship between garnet and orthopyroxene. The studied sample, 17BSK051, is a typical Kimberley field cratonic granular harzburgite, consisting of olivine, orthopyroxene and minor garnet with late accessory phlogopite. Garnet-orthopyroxene-olivine thermobarometry (Harley, 1984; Brey and Köhler, 1990) indicates equilibration at 954±5°C and 3.8±0.3 GPa, on the typical cratonic geotherm.
The fully-quantitative phase maps were analysed with the EDS system software tool 'AutoPhaseMap', which provides area and mean compositional information on identified phases. Shown together with a transmitted light thin section scan (Fig. 12) is a composite phase map and two isolated phase maps highlighting garnet, one alongside olivine, the other alongside orthopyroxene. These maps qualitatively show the close spatial association between garnet and orthopyroxene and the general paucity of shared grain boundaries between olivine and garnet. In two cases, garnet is nearly completely enclosed in orthopyroxene. In terms of area fraction of the three main phases, olivine occupies 68%, orthopyroxene 29% and garnet 3.0%. The mean Mg# of olivine from the phase map is 93.1, comparing very well with the mean of five spot analyses (93.2; Table 2). In the olivine mode vs. olivine Mg# diagram (Fig. 3f), the sample plots close to the 7 GPa residue trajectory (Walter, 1998) and is similar in mode and olivine composition to many of the Kaapvaal garnet harzburgites originally described by Boyd (1989).

4.3.3 Rare earth element systematics of selected garnets and orthopyroxenes in garnet harzburgites

Notwithstanding the wide agreement that the key phase for deciphering the Si/Mg harzburgite issue is orthopyroxene, the trace element and isotopic arguments have largely been made from analyses of garnet (e.g. Griffin et al., 1999; Stachel et al., 2004; Gibson et al., 2013; Shu and Brey, 2015). One main reason for focusing geochemical studies on garnet is that it has much higher partition coefficients than orthopyroxene or olivine for most trace elements (e.g. Sun and Liang, 2013), meaning that even in highly depleted peridotites, in situ analytical methods can quantify trace element concentrations. As shown here and elsewhere (e.g. Canil, 1992; Canil, 2004; Gibson, 2017; Tomlinson et al., 2018), many garnets found in highly depleted harzburgite formed upon cooling by exsolution from originally more aluminous higher temperature orthopyroxene that existed on the solidus. Therefore, there is major petrological interest in the trace element geochemistry of orthopyroxene.

Of all measured trace elements in harzburgitic garnet, the REE are most frequently cited as requiring metasomatism to explain the unusual systematics. The key observation is that most analysed garnets do not show the simple steep CI-normalised REE pattern expected from experiments (e.g. Sun and Liang, 2013). Instead, at least half the studied garnets, including many diamond inclusions (e.g. Stachel et al., 1998; 2004), show some form of sinusoidally-shaped CI-normalised REE pattern with a 'hump' centred over the heavier LREE.
or the lightest MREE but with exceedingly low La and Ce concentrations (e.g. Gibson et al.,
2013). This pattern is widely considered to imply metasomatic addition of LREE.

Here we report full REE data (Table 2) for garnet and co-existing orthopyroxene for
the BP002 megacryst studied by Tomlinson et al. (2018) and for the phase-mapped granular
harzburgite 17BSK051. The CI-normalised patterns for both phases (garnet: panels a and b
and orthopyroxene: panels c and d) for both samples show the MREE-enrichment. For the
analyses least compromised by kimberlite contamination and alteration (see also
supplemental information), the HREE concentrations are below 0.5 ppb, with those of Tm
and Lu below 0.1 ppb. Despite the large scatter in individual orthopyroxene HREE analyses
(Fig. 13c), the pooled data from least contaminated analyses result in a smooth average
patterns (Fig. 13g and h) characterised by near-symmetric parabola with an apex over Sm in
Onuma-style diagrams. Both garnet and orthopyroxene patterns also show a secondary
upswing in the heaviest HREE. For BP002, garnet/orthopyroxene apparent distribution
coefficients (not shown) rise monotonically from 10 to 800 from La to Lu, as predicted from
experimental data. In 17BSK051, garnet/orthopyroxene apparent distribution coefficients
(not shown) rise monotonically from 2 to 500 from La to Er and stay around 500 for the
remaining HREE. This could be an artefact of the uncertainties on the HREE concentrations
for the orthopyroxene but a similar observation was also made by Wasch et al. (2009).
Regardless, in both studied samples, garnet and orthopyroxene have sinusoidal REE patterns,
strongly supporting the argument that garnet exsolved from orthopyroxene. The curious
MREE-enrichment was therefore inherited from the precursor phase, implying that if
metasomatism had occurred it would have predated the exsolution of garnet (Tomlinson et al.,
2018). Alternatively, the relative enrichment in the MREE is unrelated to metasomatism and
inherent to high Si/Mg harzburgite formation.

5. Discussion
Two of the most distinguishing features of Archaean cratons are the existence of highly
refractory, strongly ultramafic harzburgite in the mantle root and the widespread occurrence
of ultramafic lavas at the proto-cratinic surface. When combined, these mutually related rock
types could be thought of as ultramafic 'sandwich' layers of Archaean cratons. In this final
section, we discuss the importance of mafic-ultramafic resurfacing for proto-cratinic crustal
architecture and stabilisation, analyse the types of mantle source regions that could have a
formed the erupted lavas and residues found as xenoliths.
5.1 The role of mafic-ultramafic lids as conductive incubators

Several prominent papers have suggested that the A-P boundary did not represent a relatively sudden change in style of geology but that a more familiar geology began to emerge much earlier and more gradually, from ca. 3 Ga (e.g. Shirey and Richardson, 2011; Keller and Schoene, 2012; Dhuime et al., 2012; Tang et al., 2016; Smit and Mezger, 2017). Nearly all the supporting lines of evidence for this proposal are based on interpretation of geochemical and/or radiogenic isotope data. Several authors have argued that from ca. 3 Ga onwards, plate tectonics (Tang et al., 2016) or Wilson cycles (Shirey and Richardson, 2011) started operating. This geochemical view of a gradual transition from the Mesoarchaean to the Palaeoproterozoic contrasts with the traditional geological evidence, which is in favour of a much sharper end of the Neoarchaean. For example, Bédard (2018) pointed out that there is no obvious difference in the geology of Neoarchaean and Mesoarchaean or even older terranes, with the only exception of the appearance of K-rich granitoids at ca. 3 Ga. Here we propose that the seemingly incompatible lines of evidence can be reconciled by considering stratigraphic context, which is typically lost in large time-series compilations.

This is illustrated here with the example of the Abitibi greenstone belt, which has the best defined laterally coherent stratigraphy. Similar mafic-rock dominated cover sequences have been documented across other cratons, including the Zimbabwe (Wilson et al., 1995), the Eastern Dharwar (Jayananda et al., 2013), the East Pilbara (Hickmann and van Kranendonk, 2012), the Northeastern Superior (e.g. Maurice et al., 2009) and the Kalgoorlie terrane of the Yilgarn (Hayman et al., 2015). As illustrated earlier (Fig. 8), the stratigraphy of the Abitibi greenstone belt is made up of 7 lower assemblages dominated by volcanic rocks and two upper assemblages of sediment-dominated rocks deposited into successor basins (e.g. Ayer et al., 2002). Studies seeking to infer crustal evolution from the chemistry of sediment rely on faithful representativeness of the preserved sedimentary rocks, reflecting the full stratigraphy of the supracrustal strata.

However, in the Abitibi greenstone stratigraphy, sedimentary rocks of all types are only common in the uppermost two assemblages but exceedingly sparse within the 7 lower assemblages. There they occur between volcanic units as conformable condensed horizons (Thurston et al., 2008) composed of thin iron formation, chert, exhalite and so-called argillite, which is a very-fine grained sediment containing reduced C. Of these, only argillite has an appreciable siliciclastic component that could potentially be of use to the global geochemical...
approach proposed by Keller and Schoene, (2012), Tang et al. (2016) and Smit and Mezger (2017). However, due to the precipitated authigenic component (Si, Fe) and the sequestration of transition metals from the water column into argillites, it is impossible to compare their chemistry with bona fide clastic metasediments. For example, the argillites of the Kidd-Munro and Kinojevis assemblages studied by Barrie (2005) in the Halloway area, have high average Co/Th and Cr/Th ratios, which would indicate a high mafic to ultramafic source component (Fig. 2b) but their average Ni/Co ratio is only 2.7±0.5, much lower than coeval siliciclastic sediments (Fig. 2d) compiled by Tang et al. (2016). This is not because the sediment source of the argillites is more felsic but because of the higher concentration of Co than Ni in VMS-type hydrothermal fluids. For many greenstone belts, the desired clastic sedimentary rocks sampling the main mafic-ultramafic magmatic units simply do not exist and are therefore not represented in global datasets.

Instead, the datasets are dominated by and skewed towards the sand-, silt- and mudstones deposited into the late successor basins. These formed in rifts (e.g. van Kranendonk et al., 2010), coevally with emplacement of potassic magmas (e.g. Mueller et al., 1994) post-dating major mafic and ultramafic volcanism. The chemistry of these sedimentary rocks is thus not representative of the bulk of the greenstone belt. In the Abitibi greenstone belt, Feng and Kerrich (1990) studied the chemistry of rift-hosted Porcupine and Timiskaming assemblage sedimentary rocks (the two youngest assemblages) and the Ni/Co of the various subunits from their study is shown in figure 2d. The mean of all units (4.9±0.5) confirms the trend identified by Tang et al. (2016) from global compilations (the mean of their 2.7 Ga data is 5.6±0.35). We propose that these sediment chemistries inform about the progress of intra-crustal differentiation and cratonic stabilisation rather than secular changes in crustal makeup or plate tectonics. Strong upward vertical movement of the radioactive heat producing incompatible elements K, U and Th in granitoid melts was accompanied by downward foundering of supracrustal lithologies, including of Timiskaming age (Lin et al., 2013). Because successor basins and their sediments only started forming as the crust was gaining mechanical stability (e.g. Bédard, 2018), there is an inherent bias towards preserving detritus from these more evolved sources. This limitation does not narrow the significance of the global analyses of sediment databases (e.g. Tang et al., 2016; Smit and Mezger, 2017) but complicates the tectonic interpretation of resulting temporal trends.

Our proposal is supported by studies that focus on the chemistry of the magmatic rather than on sedimentary rocks. Moyen and Laurent's (2017) analysis of the mafic igneous rock database concluded that (p.116-117): "the result is surprisingly disappointing, as rocks
throughout the Archaean show exactly the same geochemical pattern, from the oldest rocks
known at Isua or Nuvvuagittuq to the latest Archaean e.g. in China or Zimbabwe – i.e. over
about 1.5 Ga of geological history”. With respect to the ultramafic rocks, the present analysis
of the MgO-distribution of Archaean volcanic rocks also lacks evidence for a change in the
chemistries of lavas that erupted between the Meso- and Neoarchaean (Fig. 7). Furthermore,
the maximum MgO of Archaean komatiites does not show a secular trend either (e.g. Kamber,
2010) and there is no evidence for a secular decrease of depletion extent within Archaean
harzburgite suites but there is a strong change after the A-P boundary (e.g. Boyd, 1989;
Griffin, 1999). We regard these as very important observations, because mafic and ultramafic
magmas have potential to inform about changes in mantle melting regime.

By contrast, there is very strong evidence from many cratons (e.g. Smithies et al.,
2009; Laurent et al., 2014) for a temporal and stratigraphic evolution of granitoid magmatism,
expressed broadly as a trend towards more potassic compositions and shallower emplacement
(for much more detail, see Moyen and Laurent, 2017). This trend is also observed in the
Abitibi greenstone belt (Fig. 8c) where the oldest plutonic rocks are grey gneisses of diorite-
tonalite composition, maturing with time to granodiorites and biotite-bearing granodiorites
(e.g. Benn, 2004), before reaching granitic compositions that typify shallow late intrusions
(e.g. Petrus et al., 2016). Laurent et al. (2014) interpreted the first appearance of sanukitoids
at ca. 3 Ga to mark the onset of proper plate collision. However, the alternative explanation is
that the evolution of felsic magmas is the reflection of progressive differentiation of the crust
(Bédard, 2018).

There are two main factors that drove crustal differentiation in the Archaean. The first
is the substantially higher radioactive heat production within the crust and the second is the
repeated emplacement of thick supracrustal mafic-ultramafic lids (e.g. van Kranendonk et al.,
2015, Wiemer et al. 2018). The resulting incubation accelerated intracrustal crustal
reorganisation (Sandiford et al., 2004), with a general vertical rise of the incompatible heat
producing elements in granitoids and granites. Accordingly, the late intrusion of high-K,
shallow lopolithic intrusions (e.g. Frei et al., 1999) marks the mechanical stabilisation of the
crust (e.g. Kamber, 2015; Bédard, 2018), completing the process of cratonisation.

The idea of vertical foundering or sagging of denser and colder greenstones between
granitoid diapirs, also known as ’dome and basin’ or ’balloon’ tectonics (e.g. Ramsay, 1989),
is not universally accepted (e.g. Kimura et al., 1993). However, thermal and viscoelastic
mechanical modelling of inverted density profiles (Robin and Bailey, 2009; Thebaud and Rey,
2013) shows that it remains a plausible mechanism. Furthermore, field investigations (e.g.
Blenkinsop et al., 1993; Ayer et al., 2002; Johnson et al., 2016; Wiemer et al., 2016) on various cratons have questioned the scepticism towards ‘dome and basin’ tectonics that arose from the LITHOPROBE project (e.g. Clowes et al., 1992). It is perhaps unfortunate that ‘dome and basin’ tectonics has been termed 'sagduction' by some (e.g. Chardon et al., 1996) as this intracrustal process (e.g. Bouhallier et al., 1995) is not an alternative to subduction, which describes movements (that include a vertical component) of one plate relative to another. Crustal reorganisation through radioactive heat and conductive incubation is not unique to the Archaean (Sandiford and McLaren, 2002) but is much less common in the post-Archaean geological record. Because radioactive heat production decayed smoothly with time, it alone cannot explain the existence of the A-P boundary, which more likely represents the combination of several factors. Here we argue that after 2.5 Ga, the inability to commonly produce low viscosity high-Mg lavas as during the Archaean resulted in the reduction of extent of mafic-ultramafic resurfacing, i.e. the deposition of thinner ensialic greenstone blankets. It is difficult to reliably estimate the original thickness of supracrustal blankets because in most cases, the dense rocks have foundered and are now preserved as highly deformed remnants at mid-crustal or deeper levels (e.g. Bouhallier et al., 1995; Johnson et al., 2016) or could have been lost as foundered eclogite (e.g. Johnson et al., 2014). Furthermore, greenstone stratigraphies were not deposited with uniform thickness across the proto-cratons. Variations in thickness of volcanic units may reflect proximity to major volcanic centres, topography at the proto-cratonic surface and syn-depositional deformation (e.g. Berger et al., 2011). Observed stratigraphic thicknesses of rapidly emplaced volcanic units (‘groups’ or ‘assemblages’) range from 3-5 km in the Eastern Yilgarn (Hayman et al., 2015) and the Eastern Dharwar craton (Jayananda et al., 2013) to ca. 10 km in the East Pilbara terrane (Hickman and van Kranendonk, 2012) and Abitibi greenstone belt (Berger et al., 2011). Full greenstone belt thicknesses are much greater, reaching up to 20-25 km (Ayer et al., 2002; van Kranendonk et al., 2007). In numerical models, deposition of 5-15 km thick incubating lids of mafic-ultramafic cover sequences drives the observed crustal reorganisation.

We argue that the markedly reduced post-Archaean conductive incubation together with reduced crustal heat production contributed to the observed change in geological style. In post-Archaean terranes that did experience strong basaltic to picritic volcanic resurfacing (e.g. the Palaeoproterozoic Flin Flon (Canada) and the Ashanti (West Africa) greenstone belts; Perroux et al., 2012), the crustal architecture has an Archaean flavour but komatiite is largely absent and the prevailing theory is that these Palaeoproterozoic terranes formed via arc-accretion (e.g. Lucas et al., 1996).
5.2 Melting of fertile, highly refractory or metasomatised mantle?

Turning from the ultramafic rocks at surface to those in the lithospheric root, the various possible petrological models that could have left many cratonic peridotites with high enstatite/olivine (i.e. high Si/Mg) ratio have been reviewed by Herzberg (1993). At the time, the preferred explanations ranged from melting residues of a non-pyrolitic mantle, metamorphic segregation (e.g. Boyd, 1989) to cumulate crystallisation from a high-SiO$_2$ magma ocean. One alternative model, metasomatic addition of SiO$_2$ in subduction zones (Kesson and Ringwood, 1989) was less popular on account of the high fluid/rock ratio required (e.g. Canil, 1992) and the mismatch of observed with predicted CaO, Al$_2$O$_3$ and SiO$_2$ systematics (Herzberg, 1993). With the discovery of the sinusoidal REE patterns in cratonic harzburgite garnets (Stachel et al., 1998) the pendulum swung in favour of metasomatic addition of LREE and SiO$_2$ (e.g. Simon et al., 2003, 2007; Bernstein et al., 2007; Pearson and Wittig, 2008; Wasch et al., 2009). However, a comprehensive survey of the O-isotope composition of variably Si-enriched cratonic peridotites has documented a uniform mantle parentage (Regier et al., 2018), strongly arguing against the subduction zone metasomatic model.

An aspect common to most of the quoted studies is that they compare cratonic harzburgite olivine modes and forsterite contents with the empirical trend from depleted modern oceanic peridotites (Fig. 3c) originally proposed by Boyd (1989). However, Boyd's trend was never scrutinised with experimental data. Comparison with the experimental melting residues of Walter (1998) shows a strong pressure effect (Fig. 3d). None of the isobaric experimental residues reproduce the (hand-drawn?) concave trend of Boyd (1989). At pressures of 5 GPa or higher, the experimental olivines are highly magnesian and exist in a residue containing 25-35 % modal combined pyroxene and garnet (Walter, 1998). It is recognised that this comparison is limited to one experimental dataset because most other relevant high-pressure studies did not tabulate estimates for modal abundance. We note, however, that in terms of phase relationships and stability fields, Walter's (1998) experiments are compatible with those conducted at lower pressure and those that did not report full modal abundances (e.g. Takahashi, 1986; Canil, 1991; 1992; Takahashi et al., 1993; Baker and Stolper, 1994; Longhi, 2002; Schutt and Lesher; 2006). There is thus no indication that the modes from these experiments would be very different. We agree with Canil (2008) that the mismatch between observed and expected cratonic harzburgite mineralogy is strongest
the Kaapvaal craton but harzburgites with <60% modal olivine also exist from the Slave, Siberian and Wyoming cratons (Egglcer et al., 1987; Ionov et al., 2010; Kopylova et al., 1999; Kopylova and Caro, 2004; Newton et al., 2015; Solovjeva et al., 1994). The relatively low modal abundance of olivine could mainly indicate that melt extraction occurred at high pressure but further in-depth analysis will be needed for a firmer conclusion. In terms of the variability in olivine mode at a given forsterite content (Fig. 3f), we note again the extremely coarse-grained nature of cratonic harzburgites (e.g. Figs. 11 and 12) and propose that some of the modal variability could stem from minor mineral segregation and from unrepresentativeness of small thin sections.

One aspect of the cratonic Si/Mg issue that has not yet been explored experimentally is the possibility that high-Mg harzburgites could be residues of multiple melt extraction events. Arndt (1977a) commented (p. 205) that "ultrabasic magmas probably form by a sequential melting process and are derived from a residuum composed of refractory minerals and trapped liquid left by previous episodes of partial melting and magma extraction". Our analysis of the experimental liquids of Walter (1998) shows that re-melting of a previously depleted source can produce a much wider range of Si/Mg in the liquids than single batch melting (Fig. 10g and h) as well as more extreme Fe/Mg and Al/Mg variability as seen in some komatiites. More experimental data on refractory starting compositions and modelling of melt-residue interaction are certainly needed to better understand the Si/Mg systematics of komatiites and harzburgites (e.g. Herzberg, 2016), particularly in light of the new O-isotope data (Regier et al., 2018) that argue against subduction zone metasomatism.

There are strong indications from the geochemistry of komatiites and komatiitic basalts that variably fertile sources were being melted (e.g. Sproule et al., 2002). Although the source fertility of komatiites is conventionally determined via Al (Nesbitt et al., 1979), experimental data show that the Ti concentration may be a simpler measure to assess whether a source had been previously depleted (see also Sproule et al., 2002; Robin-Popieul et al., 2012). This is because Ti remains incompatible across the entire melting depth interval relevant to cratonic lithosphere (Longhi, 2002). The archetypes of melts from ultra-depleted sources are from Commondale (Wilson, 2003) but komatiites with $\text{Al}_2\text{O}_3/\text{TiO}_2 > 25$ have also been reported from localities outside the Kaapvaal craton (e.g. Sproule et al., 2002). This supports the proposal of Arndt (1977a) and suggests that additional experimental constraints are required for harzburgitic starting compositions. The low-Ti nature of many komatiites is shared (to some extent) by modern boninites but this common trait is, in our view, insufficient to justify the subduction zone origin proposed by Parman et al. (1997).
In terms of REE systematics, one key question of relevance to proto-craton formation is whether the sinusoidal REE pattern arose from metasomatism post-dating the original melt-extraction (e.g. Stachel et al., 2004) or whether it is an inherent feature of cratonic mantle related to its formation by high degree melting (e.g. Tomlinson et al., 2018). There is no doubt that cratonic lithosphere has experienced multiple episodes of metasomatic infiltration, particularly in the base (e.g. Nixon et al., 1987; Foley, 2008) but there is very strong evidence that the sinusoidal REE pattern is ancient and not related to Neoarchaean metasomatism. The following are the strongest lines of evidence. Firstly, the Hf-isotope composition of garnets with sinusoidal REE patterns are extreme, reaching to spectacular present-day ε(Hf) of up to +1,000 (Shu and Brey, 2015) with correspondingly ancient Archaean model ages. Shu et al. (2013) discovered that garnet separates (with sinusoidal REE patterns) from Kaapvaal craton kimberlites yield Lu/Hf regression lines of 3.2 Ga, suggesting that the sinusoidal REE pattern has to be at least as old. This age is substantially older than the Neoarchaean cratonisation. Secondly, Koorneef et al. (2017) succeeded in dating diamond inclusion garnet populations with strongly sinusoidal REE patterns to 2.95 Ga with 147Sm/143Nd. The majority of these garnets have very low Sm/Nd ratios, consistent with the hump in the REE pattern over Nd and Sm. Thirdly, as shown by earlier studies (Zhang et al., 2001; Simon et al., 2007; Wasch et al., 2009) and the new data on the BP002 megacryst (Tomlinson et al., 2018; this study), the REE of orthopyroxene also show a humped pattern, implying that the entire protolith peridotite carried this signature. Finally, Stachel et al. (2006) reported on diamond inclusions hosted in 2.7 Ga volcaniclastic rocks and lamprophyre dykes in the Wawa greenstone belt. The inclusion suite contains garnets very similar to those in common cratonic harzburgite and the nitrogen aggregation of the diamonds suggests residence times of tens to hundreds of Ma prior to crustal emplacement. Collectively, this evidence suggests that harzburgite with sinusoidal REE patterns existed in the Meso- and Neoarchaean cratonic mantle root.

This finding is relevant to komatiite geochemistry in two respects. Firstly, garnet and orthopyroxene are the main carriers of the REE in advanced melt residues between 4 and ca. 8 GPa. Depending on the magnitude of the hump over the heavier LREE (Nd-Gd), the Sm/Yb or Gd/Yb ratio of a melt in equilibrium with these phases will either be slightly sub- or supra-chondritic. The important conclusion is that if the komatiite source had a sinusoidal REE pattern, there will only be limited variability in Sm/Yb or Gd/Yb ratios in liquids. This is one possible solution to the paradox identified by Walter (1998), namely that major elements of
many komatiites require a garnet-bearing source but REE do not show the expected large
Sm/Yb fractionation. As the new examples of garnet from exsolved harzburgite
orthopyroxene (Tomlinson et al., 2018; this study Figs. 11 and 12) show, melt extraction
at >3 GPa is not necessarily in conflict with the high Cr/Al ratios of residues, provided they
were originally bi-mineralic olivine-orthopyroxene rocks. Secondly, a source with sinusoidal
REE patterns has the potential to evolve to diverging Nd and Hf isotope compositions.
Because the apex of the hump in the REE pattern is frequently around Sm or Nd, the
resulting Sm/Nd does no differ much from chondrite and Nd-isotope ratios will evolve to
mildly supra- or even sub-chondritic values. By contrast, the Lu/Hf ratio is mostly strongly
superchondritic and Hf-isotope ratios rapidly evolve to positive ε(Hf) values. We propose
that this could explain the Nd-Hf isotope systematics of the Commondale komatiites
(Hoffmann and Wilson, 2017) and the decoupled Nd-Hf arrays of other Kaapvaal craton
komatiites (Blichert-Toft et al., 2015).

It is beyond the scope of this paper to fully discuss the various proposals for the origin
of the sinusoidal REE pattern and its possible relation to Si enrichment (e.g. Burgess and
Harte, 2004). If the sinusoidal REE pattern can be attributed to metasomatism with (e.g.
Gibson et al., 2013) or without previous melt depletion it has to be Archaean in age and, in
some cases, clearly predates craton stabilisation. Just as with the Si/Mg systematics, there
remain significant gaps in experimental knowledge for REE behaviour in peridotite at the
conditions relevant to cratonic melting. For example, pseudo-sections for unmetasomatised
fertile peridotite predict that the phase assemblage at the 5 GPa solidus is orthopyroxene-free,
consisting only of olivine, clinopyroxene and garnet (e.g. Fig. 1 of Jennings and Holland,
2015). Thus, due to the expansion of the stability field of pigeonitic clinopyroxene above
1,500°C and pressures greater than 4.2 GPa (Takahashi, 1986), the peridotite that is melting
is technically called a garnet wherlite, not a lherzolite. To our knowledge, no relevant
experimental data exist for the REE partitioning of this type of pyroxene. Until the necessary
experiments have been conducted, it will remain unclear if a sinusoidal REE patterns could
result in residues of multiply melted normal peridotite without metasomatic REE addition.

5.3 Possible causes for the demise of very high-temperature mantle upwellings
and the relevance for the Archaean-Proterozoic boundary
There remains much uncertainty regarding the A-P boundary. In our view, the geological evidence in favour of a boundary outweighs the geochemical indications for earlier (e.g. 3 Ga) planetary changes in tectonic style. If we accept the geochronological confirmation of the original postulation of the A-P boundary in the 19th century (Logan, 1857) as more than a fortuitous coincidence, the boundary must have major geological significance.

Most ideas seeking to explain the A-P boundary advocate a reorganisation of the mantle temperature structure and/or heat loss mechanism. Two schools of thought are most prominent. Firstly, a change within or above the core-mantle thermal boundary layer may have occurred and this affected the type of thermal upwellings that travelled through the mantle. For example, Campbell and Griffith (2014) attributed the A-P boundary to piling subducted oceanic plates into D'', forming an insulating layer from which modern-style plumes began emerging since the Palaeoproterozoic. Likewise, Bédard (2018) envisaged mantle 'overturn upwelling zones' that episodically transported heat and material from the lower mantle towards the surface.

The second group of ideas has identified the mantle transition zone as a possible location where major change may have occurred at the A-P boundary. In these models, the base of the transition zone was a much stronger thermal boundary layer in the Archaean than thereafter. The hot mantle upwellings that caused komatiite magmatism could have originated at this boundary layer. Davies (2008) proposed that throughout the Archaean, subducted slabs piled up in the transition zone, forming a mechanical barrier that impeded heat loss from the lower mantle. According to his model, episodic collapse of the barrier occurred every 100-150 Ma and manifested as pulses of high temperature melting events in the upper mantle. From the A-P boundary onwards, slabs began to penetrate the lower mantle and the transition zone barrier stopped building up. Rino et al. (2004) advocated a similar model and they regarded the greater thickness of the subducted Archaean oceanic crust as key to the neutral buoyancy of slabs in the transition zone. Breuer and Spohn (1995) argued that the A-P boundary marked the change from 2-layer to single layer mantle convection and envisaged a pronounced flush instability that caused a global Neoarchaean igneous 'super event'.

In our view, all these models contain attractive aspects to explain the A-P boundary and it is noted that the Archaean pile-up of slabs in the transition zone is not mutually exclusive with post-Archaean slab pile-up in D''. However, none of the current models has addressed the wider evidence from the crustal and mantle portions of cratonic lithosphere. We regard the following four key observations as most relevant for such a model:
i) All cratons either still have or have formerly had deep mantle roots with geotherms corresponding to an average present-day heat flow of 41 mW m$^{-2}$ and strong concentration of radioactive heat production within the uppermost crust (e.g. Michaut et al., 2009). Importantly, relatively cool cratonic mantle geotherms apparently also existed in the Archaean (Boyd et al., 1985). In the Kaapvaal craton and elsewhere, there is evidence that the Neoarchaean and early Proterozoic geotherms were surprisingly cool, only mildly more elevated than today's (around 45-50 mW m$^{-2}$; e.g. Nimis, 2002). This requires that in the Archaean, conductive heat loss extended to greater depth than the base of the current cratonic roots and/or that the original cratons were larger in extent than the fragments preserved today (Ballard and Pollack, 1988). This requirement is illustrated on figure 14. It is widely agreed that most cratons show evidence for post-Archaean base erosion (e.g. Foley, 2008; Xu and Qiu, 2017) supporting the notion that they were originally deeper. Rare xenoliths from the Kaapvaal craton contain majoritic garnet relics from 300-400 km depth (Haggerty and Sautter, 1990), deeper than the present-day base of the lithosphere. Regardless of preferred proto-craton thickness, strongly elevated lower crustal temperatures are unavoidable (Fig. 14).

ii) Throughout the Archaean, there is no evidence for secular change in komatiite Mg-content (Kamber, 2010; Campbell and Griffiths, 2014) and mafic magmas in general have remained chemically unchanged (e.g. Moyen and Laurent, 2017). But there was a sharp drop in MgO content of ultramafic magmas in the Palaeoproterozoic. By contrast, basaltic magmas have persisted through geological time with very similar mean MgO content as far back as the Palaeoarchaean (Kamber, 2015 and this study).

iii) Archaean crustal rocks, and particularly komatiites and associated fractionated basalts, preserve evidence for very ancient element fractionation of Sm/Nd, Hf/W and Sm/Lu (e.g. Caro et al., 2003; Touboul et al., 2012). The resulting $^{142}$Nd and $^{182}$W anomalies and the $^{143}$Nd/$^{176}$Hf divergence are more pronounced in Palaeo- and Mesoarchaean rocks (e.g. Bennett et al., 2007; Rizo et al., 2011) but still found in Neoarchaean samples (e.g. Debaille et al., 2013; Puchtel et al., 2016). It follows that the mantle domains harbouring such anomalies must have escaped convective mixing until the Neoarchaean and earliest Palaeoproterozoic.

iv) Although in many cratons the crust and the mantle portion experienced coeval events (melting, depletion, reworking, re-enrichment, etc.) the two reservoirs are not complementary. The preserved cratonic mantle root alone cannot have supplied all the incompatible trace elements present in the crust (e.g. Carlson et al., 2005). Therefore, crust
formation must have depleted greater volumes of mantle that never were or no longer are
spatially associated with the craton.

One possible solution to the last two observations could be that proto-cratonic mantle
formed in multiple ways. This could include initial shallow melt extraction from
asthenosphere that only produced a relatively thin refractory root, maybe due to effective
dripping of crustal restite residues (e.g. Bédard, 2006; Rollinson, 2010; Johnson et al., 2014).
There is no a priori necessity that these proto-cratons grew from unusually hot mantle. Indeed,
the tholeiitic lavas on many cratons could have been sourced from relatively shallow mantle
(e.g. Wyman et al., 2002; Smithies et al., 2009). By contrast, the rocks that now constitute the
deep cratonic mantle root may have originated in an unrelated part of the mantle – here
termed ancient non-convecting domains (ANCD).

One plausible way of producing ANCD early in Earth history is crystallisation and
crystal segregation in magma seas (e.g. Arndt et al., 2002; Hansen, 2015). The geotherm after
crystallisation of a global magma ocean is very different from the adiabat and almost
certainly leads to instability (e.g. Elkins Tanton et al., 2003; Kramers, 2007) that could have
yielded domains of ANCD. An additional possibility is that giant impacts caused more local,
shallower magma seas and their crystallisation products (cumulates) may have persisted (e.g.
Hofmeister, 1983) as ANCDs surrounded by asthenosphere. Alternatively, stratified
crystallisation products of magma seas can experience gravitational instability and may
founder (e.g. Elkins Tanton et al., 2002) and could survive as non-pyrolitic ANCDs at depth.
We note that cumulate formation during fractional crystallisation from a magma ocean has
been suggested as one possible cause for the short-lived radiogenic isotope anomalies
recorded in Archaean rocks (Caro et al., 2005). Finally, very deep mantle melting from lower
mantle upwellings could have formed refractory mantle zones not originally associated with a
proto-craton.

In figure 15, we illustrate that if ANCDs occupied laterally extensive areas in the
mantle transition zone, they could have impeded heat transfer from the lower mantle. Heat
would eventually escape, either through partly eroded ANCDs or along their margins as
upwellings that triggered high temperature melting in the asthenosphere. Nascent continental
lithosphere that drifted over areas covered by ANCDs at depth would be mostly protected
from the extensive resurfacing that affected most of the remaining Earth. The nascent
continental plates could have been similar to the ribbon continents proposed by Bédard
(2018). Alternatively, they could have formed as arc-like collages (e.g. Wyman et al., 2002)
that occasionally experienced emplacement of much hotter and deeply sourced magmas through the ANCD.

With time, the accumulated effects of thermal upwellings weakened the ANCDs to the point of collapse, marking the A-P boundary. Remnants of the ANCDs were mostly recycled back into the asthenosphere but locally, became incorporated into the cratonic roots. The accompanying decompression and cooling could explain mineral reaction textures (including exsolutions) in garnet harzburgites and pyroxenites (e.g. Haggerty and Sautter, 1990). The dismembered deep parts of ANCDs would have carried with them transition zone minerals that could survive as diamond inclusions. The majority of transition zone garnet-structured diamond inclusions are of mixed peridotitic/pyroxenitic chemistry (Kiseeva et al., 2013) and only a minority has an eclogitic parentage. If these inclusions are samples from the deep ANCDs, their chemistry favours the idea that the barriers were mainly composed of pyroxenite progenitors rather than subducted oceanic crust but it is possible that slab pile-up as envisaged by Rino et al. (2004) and Davies (2008) added to the mechanical and thermal barrier. The initial Hf-isotope composition of Kaapvaal craton harzburgite garnets presented by Shu et al. (2013) are radiogenic and require that the precursor material separated from chondritic mantle at least a few hundred million years before 3.2 Ga. This constraint on timing is consistent with the idea that ANCDs could have formed in giant impact basins (e.g. Hansen, 2015) during the late bombardment (Bottke et al., 2012).

A model in which cratonic lithosphere grew from above and was fused with deep remnants could also explain why one class of harzburgites (those without high modal abundance orthopyroxene) appears to have mainly foundered after melt extraction (Canil, 2004; Lee and Chin, 2014) whereas others, like BP002, appear to have moved upwards (Haggerty and Sautter, 1991; Tomlinson et al., 2018). We also note that some cratons may have more thoroughly depleted lower roots and more lherzolitic shallower portions (e.g. Griffin et al., 2003). This observation could be explained by re-enrichment of formerly more strongly depleted harzburgite with fluids from depth (e.g. Gibson et al., 2013). Alternatively, the working model advanced here would envisage the less depleted zone to represent the area of fusion of shallower with deeper residues. It corresponds in depth both with the impedance increase found by Green and Hales (1968) and a 50 km thick zone of pronounced anisotropy with horizontal azimuths that could represent frozen-in drift (Soudoudi et al., 2013) of the shallow nascent continental plates.
6. Concluding remarks

Regardless of the validity of competing models for craton formation, the general difficulties of sourcing sufficient highly incompatible elements for cratonic crust (e.g. Bédard, 2006; Michaut et al., 2009) and the requirement for cool temperatures in the Archaean roots (e.g. Nimis, 2002) remain. With respect to the former, Kramers and Tolstikhin (1997) pointed out that the formation of a global Hadean protocrust could have led to an initial overall depletion of the entire mantle. When the Hadean crust was later comprehensively destroyed, the Eoarchaean asthenosphere could have ended up with an elevated inventory of incompatible elements if crustal recycling was restricted to a depth of 670 km. Melting of such enriched asthenosphere to form Archaean cratons could go a long way towards alleviating the mass imbalance between cratonic root and crust for highly incompatible elements. Furthermore, depending on the mass and composition of the Hadean crust, the upper mantle may also have ended up with a slightly elevated Si/Mg ratio with higher modal abundance of orthopyroxene.

Concerning the requirement for surprisingly cool temperatures in the Archaean cratonic roots, it is worth noting that very limited knowledge exists about the original dimension of Archaean cratons (e.g. Bleeker, 2003). The model of Ballard and Pollack (1988) assumed relatively modest diameters of proto-cratons (400-600 km) informed by the then prevailing notion that cratons contained small ancient nuclei onto which additional lithosphere accreted over time (e.g. Kusky, 1998). However, even in the Zimbabwe craton, where the evidence for a small original nucleus was once thought strongest, newer zircon U/Pb dates (e.g. Horstwood et al., 1999; Bolhar et al., 2017) and Re-depletion constrains on the cratonic root (Smith et al., 2009) now suggest that the craton had attained its full preserved size from the beginning. Furthermore, detailed comparison of crustal stratigraphies, radiogenic isotope characteristics, dyke swarm analysis and palaeomagnetic data imply that the Zimbabwe craton could have been part of the same craton as the Yilgarn (Smirnov et al., 2013). There is the additional possibility that this Zimgarn craton could itself only be a fragment of an even larger original craton (e.g. Bleker, 2003) composed of additional cratons (including the Slave, Wyoming and North China) that all share the so-called high $\mu$ Pb-isotope signature (Kamber et al., 2003; Kamber 2015). In terms of Archaean geodynamics, this opens the possibility that craton formation and preservation may not have been a ubiquitous process but that only two or three large early cratons with thick roots may have formed in environments that favoured their preservation and that they bias our understanding of how early terrestrial geology operated.
Acknowledgments

This contribution started as a keynote lecture at the 2017 Goldschmidt meeting in the session "Evolution of the Continental Crust and Mantle Lithosphere" and we would like to thank the convenors Roberta Rudnick, Nick Arndt and Sarah Brownlee for the invitation. We thank Review Papers Coordinator Tim Horscroft for the invitation to write this paper, Klaus Mezger for editorial handling and Nick Arndt and Dante Canil for their thoughtful reviews. Our sincere gratitude goes to the following individuals: Sergei Lebedev for interesting discussions on craton formation; Mike Lesher for giving access to the Abitibi Subprovince komatiite database and discussions on ultramafic magmatism; John Ayer and Phil Thurston for advice on greenstone belt stratigraphy; Jock Robey for facilitating collection of Kimberley peridotites; Paul Guyett for SEM-EDX analysis of garnet harzburgite; Mags Duncan for helping with data trawling and compilations; Elaine Cullen for re-drafting figures from cited publications; and Claire Kamber for drafting figures 8 and 15. The geochemistry facility at TCD is supported by grant SFI/RI/3227.
References


Djomani, Y.P., Swain, C.J., Deen, T., Bowden, P., 2009. The lithospheric architecture of Africa:
Benn, K., 2004. Late Archaean Kenogamissi complex, Abitibi Subprovince, Ontario, Canada: doming,
folding and deformation-assisted melt remobilisation during syntectonic batholith emplacement.
Zone, Ontario: A Late Archean Large Igneous Province (LIP) as a Substrate for Juvenile Crust.
Berger, B., Bleeker, W., van Breenem, O., Chapman, J., Peter, J., Layton-Matthews, D., Gemmell, J.,
2011. Results from the Targeted Geoscience Initiative III Kidd–Munro Project. Ontario Geological
Survey, Open File Report, 6258.
mantle beneath West Greenland: dunitic xenoliths from Ubekendt Ejland. Contributions to
Mineralogy and Petrology, 152(3): 335.
Bernstein, S., Kelemen, P.B., Hanghøj, K., 2007. Consistent olivine Mg# in cratonic mantle reflects
Bindeman, I., Zakharov, D., Palandri, J., Greber, N.D., Dauphas, N., Retallack, G., Hofmann, A.,
Lackey, J., Bekker, A., 2018. Rapid emergence of subaerial landmasses and onset of a modern
Bizzarro, M., Stevenson, R.K., 2003. Major element composition of the lithospheric mantle under the
North Atlantic craton: evidence from peridotite xenoliths of the Sarfartoq area, southwestern
Archean komatites from surface sampling and ICDP drilling in the Barberton Greenstone Belt,
South Africa. American Mineralogist, 100(11-12): 2396-2411.
formation in the Zimbabwe Craton deduced from the O-Hf isotopic record of 3.8–3.1 Ga detrital


1596 Konhauser, K.O., Pecoits, E., Lalonde, S.V., Papineau, D., Nisbet, E.G., Barley, M.E., Arndt, N.T.,
1597 Zahnle, K., Kamber, B.S., 2009. Oceanic nickel depletion and a methanogen famine before the
1600 and Proterozoic diamond growth from contrasting styles of large-scale magmatism. Nature
1601 Communications, 8(1): 648.
1602 Kopylova, M., Caro, G., 2004. Mantle xenoliths from the southeastern Slave craton: evidence for
1605 from the Jericho kimberlite: implications for the thermal state of the mantle beneath the Slave
1608 17.
1609 Kramers, J.D., Tolstikhin, I.N., 1997. Two terrestrial lead isotope paradoxes, forward transport
1610 modelling, core formation and the history of the continental crust. Chem. Geol., 139: 75-110.
1611 Kramers, J.D., Kreissig, K., Jones, M.Q.W., 2001. Crustal heat production and style of metamorphism:
1612 a comparison between two Archaean high grade provinces in the Limpopo Belt, southern Africa.
1616 Kusky, T.M., 1998. Tectonic setting and terrane accretion of the Archean Zimbabwe craton. Geology,
1617 26(2): 163-166.
1618 Laurent, O., Martin, H., Moyen, J.-F., Doucelance, R., 2014. The diversity and evolution of late-
1619 Archean granitoids: Evidence for the onset of “modern-style” plate tectonics between 3.0 and 2.5
1622 constraints from broad-band surface-wave dispersion. Lithos, 109(1): 96-111.
1625 Lesher, C., Barnes, S., 2009. Komatiite-associated Ni–Cu–(PGE) deposits, New developments in
1627 Lin, S., Parks, J., Heaman, L.M., Simonetti, A., Corkery, M.T., 2013. Diapirism and sagduction as a
1628 mechanism for deposition and burial of “Timiskaming-type” sedimentary sequences, Superior
1629 Province: Evidence from detrital zircon geochronology and implications for the Borden Lake
1630 conglomerate in the exposed middle to lower crust in the Kapuskasing uplift. Precambrian

49


Sun, C.G., Liang, Y., 2013. The importance of crystal chemistry on REE partitioning between mantle minerals (garnet, clinopyroxene, orthopyroxene, and olivine) and basaltic melts. Chemical Geology, 358: 23-36.


Figure captions

Figure 1: Three-dimensional representation of tomography of the African lithospheric mantle (reproduced from Begg et al., 2009). The viewer is positioned in the mantle transition zone below the southern Atlantic ocean and is looking upwards in a NE direction towards Africa (continental outline as a white line). The up to 250-275 km deep roots of the three broad cratonic regions of Africa are labelled and defined as red coloured volumes in which the SH body-wave velocity is >1.9% than the standard model. Blue colours show volumes of unusually slow SH body-wave velocities, mainly portraying the upwelling of hot mantle below the East Africa rift system.

Figure 2: Panel (A) and (B) redrawn in colour from Figs. 18 and 19 of Condie (1993). In both panels, the coloured symbols show shale compositions. Phanerozoic and Proterozoic shales overlap in composition but Archaean shales are much richer in compatible elements Sc, Co, and Cr. The Phanerozoic and Proterozoic UCC average composition overlaps with shale data but Archaean shales are more compatible element-enriched than UCC as a whole. Panels (C) and (D) show secular evolution of UCC Ni/Co ratio reconstructed from clastic sedimentary rock compilation of Tang et al. (2016). Panel (C) plots average values with standard errors and panel (D) same as (C) with additional data from the Abitibi greenstone belt (Feng and Kerrich, 1990; Barrie, 2005).

Figure 3: Empirical and experimental data plotting modal abundance of olivine in residual peridotite vs. forsterite component in olivine. Panels (A) to (C) redrawn from Boyd (1989) showing the oceanic trend (A), off-craton harzburgites (B) and Kaapvaal craton harzburgites (C). Panel (D) shows residue mineralogy of the experiments conducted on fertile peridotite by Walter (1998) colour-coded according to experimental pressure. The cross symbols show the position of the oceanic trend of Boyd (1989). Note that the y-axis scale extends to much more magnesian olivine. Panel (E) shows the compilation of empirical peridotite compositions of Bernstein et al. (2007). Panel (F) shows the same experimental data as panel (D) but at the same scale as Boyd's (1989) figures and the position of sample 17BSK051 studied herein (orange square symbol). See text for explanations.
Figure 4: Map showing kimberlite occurrences with and without sub-calcic garnet macrocrysts and lithospheric cross-section with interpretative areas of garnet formation and metasomatic alteration. Simplified and re-drawn in colour from Nixon et al.'s (1987) figures 261 and 267.

Figure 5: Comparison of empirical (Alexo and Pyke Hill) komatiite major element chemistry with petrological predictions reproduced from Herzberg (2016). Panel (A) shows MgO vs. SiO$_2$ compositional space in which the observed LLD (open circles whole rocks and solid grey circles melt inclusions), caused by olivine crystallisation, plots at lower SiO$_2$/MgO than either batch (red cross) or accumulated fractional (green cross) melting of the melting model predictions. Panel (B) shows MgO vs. FeO compositional space in which the observed LLD of the classic Abitibi greenstone belt komatiites strongly favour batch over accumulated fractional melting of a fertile peridotite (KR4003).

Figure 6: Magnesium (MgO wt%) distribution in Archaean metavolcanic rocks from six cratons (panels A-F) for which there are more than 500 compiled analyses; a composite of four cratons with fewer data (panel G) and for the combined dataset (panel H, note logarithmic x-scale). Each panel shows the raw data as histograms of MgO wt% re-calculated to anhydrous total of 100 wt%. Superimposed on the histograms is a kernel density function (parameters listed in Table 1) as a dark blue curve with light blue fill. Linear scale diagrams highlight the dominant basaltic population at MgO 5-9 wt%, the prominent evolved population with ca. 1 wt% MgO, and the much broader ultramafic population (MgO 25-30 wt%). In all panels, the paucity of rocks with MgO of ca. 18 wt% is evident. The composite figure (panel H) with logarithmic x-scale accentuates the gap between the basaltic and ultramafic populations.

Figure 7: Magnesium (MgO wt%) and Ni (ppm) distribution in Archaean metavolcanic rocks. Panel (A) shows linear histogram of a MgO on all studied cratons with four vertical lines denoting the preferred populations from kernel density function statistics. Panels (B) to (F) all show data after logarithmic translation. Panel (B) displays Ni distribution in all compiled Archaean metavolcanic rocks, highlighting the dominant basaltic population at ca. 200 ppm and the ultramafic population centred over ca. 2,000 ppm. Panels (C) and (D) show a comparison in MgO distribution between 2.5-2.9 and >2.9 Ga metavolcanic samples. Panels (E) and (F) show the same kind of comparison for Ni distribution.
Figure 8: Stratigraphic column (panel A) of the Abitibi greenstone belt after Ayer et al. (2002). Seven volcanic dominated assemblages are unconformably overlain by two sediment-dominated assemblages at the top. Column (B) shows approximate position of mafic to ultramafic metavolcanic samples interrogated by Sproule et al. (2002) and Herzberg (2016) as well as sedimentary samples described by Feng and Kerrich (1990) and Barrie (2005). Column (C) shows evolutionary trend of plutonic samples with time recorded in the Kenogamissi complex (after Benn, 2004), starting with tonalite and grey gneisses, moving to granodiorite, biotite-bearing granodiorite and ending with proper granite.

Figure 9: Major and selected REE systematics in Abitibi and adjacent greenstone belt high-Mg basalts, picrites and komatiites (all data from Sproule et al., 2002). Panel (A) shows a histogram of MgO (wt%) content of all least-altered samples. Panels (B) and (C) show MgO (wt%) vs FeO (wt%) and SiO$_2$ (wt%) systematics, respectively. The samples are grouped into Al-undepleted, Al-depleted and Ti-depleted suites according to the Al$_2$O$_3$/TiO$_2$ criteria >25, 15-25, <15. Shown for reference are the position of bulk continental crust (from Rudnick and Gao, 2003) and the preferred modelled parental komatiite melts by Herzberg (2016) also shown on figure 5. Panel (D) plots Al$_2$O$_3$/TiO$_2$ versus CI-chondrite (after Anders and Grevesse, 1989) normalised Sm/Yb. Panels (E) and (F) show MgO (wt%) vs SiO$_2$ (wt%) systematics separated into 6 stratigraphic assemblages. Panels (G) and (H) show MgO (wt%) vs FeO (wt%) systematics separated into 6 stratigraphic assemblages (same symbols as panels (E) and (F)).

Figure 10: Selected geochemical systematics of high-P experiments relevant to komatiite petrogenesis. Panels (A) and (B) show dependence of Al$_2$O$_3$ and TiO$_2$ concentration, respectively, on extent of melting for experiments performed by Takahashi et al. (1993) and Walter (1998) on slightly different lherzolite starting compositions (also shown). Panels (C) and (D) show dependence of the Al$_2$O$_3$/TiO$_2$ ratio of liquids on Al$_2$O$_3$ and TiO$_2$ content, respectively. Same symbols as panel (A). Panel (E) is a histogram of Al$_2$O$_3$/TiO$_2$ ratios in low (<20%) and higher (>20%) degree liquids from fertile lherzolite melts from refractory mantle. Blue and green colours are from 3 and 7 GPa experiments of Walter, purple from 4.6 GPa experiment of
Takahashi et al. (1993). In each case a higher and lower degree batch melt from fertile mantle is shown with diamond and circle symbols. The hypothetical refractory melt is represented by the square symbol and corresponds to the composition of the liquid required to move from the lower degree to the higher degree melt along the tie line. The distances reflect the lever rule to preserve mass. Panel (G) shows the high-Mg basalts, picrites and komatiites from (Sproule et al., 2002) in SiO$_2$ vs MgO space with superimposed model compositions (batch and accumulated fractional melts from Herzberg (2016)) and liquids of experiments by Takahashi et al. (1993) and Walter (1998). Panel (H) shows the same data and model but with calculated hypothetical refractory melts (see examples in panel F) from previously depleted mantle. Symbols are colour coded for pressure. Note that refractory mantle melts can reach very high MgO values with highly variable SiO$_2$ content, reflecting exhaustion of pyroxene.

Figure 1: Transmitted light section scans of five garnet peridotites from the Bultfontein pans (Kimberley, Kaapvaal craton) arranged by decreasing petrographic evidence for garnet exsolution (note that these are over-thick thin sections). From the top, sample BP002 (Tomlinson et al. 2018, this study) with regular garnet lamellae in orthopyroxene; 17BSK069d with semi-regular garnet lamellae and lines of equant garnet in a single large (32 mm) orthopyroxene; CLA18 with large (~10 mm) orthopyroxene crystals, where garnet elongate and aligned is located within or adjacent to the orthopyroxene; granular peridotite CLA05, in which garnet occurs as inclusions within and adjacent to coarse (~5mm) orthopyroxene; and garnet harzburgite 17BSK051 (this study) in which garnet grains are spatially associated with coarse orthopyroxene.

Figure 12: Transmitted light scan of garnet harzburgite 17BSK051 (A) and three phase maps. Panel (B) shows a composite map with olivine in green, orthopyroxene in blue, garnet in red and phlogopite in purple. Panels (C) and (D) show isolated maps of garnet with olivine and orthopyroxene, respectively. See text for explanation.

Figure 13: CI-chondrite-normalised REE patterns (Andres and Grevesse, 1998) (panels A-F) and Onuma-type diagrams (panels G and H). Panels (A) and (C) show garnet and orthopyroxene data for megacryst BP002 and (B) and (D) equivalent data for granular harzburgite 17BSK051. Green coloured symbols denote analyses least affected by contamination and/or alteration. Red coloured symbols show variably compromised analyses.
In BP002, intergranular (black in transmitted light) contaminant (likely kimberlite) was also analysed and this was used to calculate mixing models with average most depleted garnet (panel E) and orthopyroxene (panel F). Very good coincidences with average contaminated analyses by admixing between 0.1% and 0.12% contaminant. Panel (G) shows precursor phase (blue) REE concentrations calculated from modal relative abundances of garnet (red) and orthopyroxene (green) in an Onuma-type diagram for BP002. Panel (G) shows mean most depleted garnet (red) and orthopyroxene (green) of 17BSK051 in an Onuma-type diagram.

Figure 14: Geotherms calculated by Ballard and Pollack (1988) for the Kaapvaal craton. Panels (A) and (B) depict central cratonic geotherms from models 'Present 2' (P2) and 'Archaean 2' (A2), respectively. (A) In model P2, the reduction in central cratonic heat flow to 40 mW m⁻² was achieved with a lithospheric root of 220 km depth and a reduced internal lithospheric heat production. (B) Model A2 shows the same 400 km diameter craton but at 3.2 Ga with 2.5 x higher internal heat production and the mantle potential temperature 200°C hotter than at present (stippled line is model P2 for reference). The higher internal heat production results in a Moho temperature 300°C hotter than in the present-day model and the higher mantle potential temperature elevates the mantle root geothermal overall. To achieve a similar geotherm as P2 with the higher internal heat production would require dropping the 3.2 Ga mantle potential temperature 200°C relative to present day. Panel (C) shows an alternative model for the Archaean (A2'), in which the craton is 1.5x larger in diameter than A2 and the root extents to 500 km instead of 220 km. This results in similar temperatures as present-day at depths of 120-180 km, the interval where most studied xenoliths derive from but maintains a high Moho temperature.

Figure 15: Schematic model illustrating growth and evolution of cratonic lithosphere from the Mesoarchaean (A) and the latest Neoarchaean (B) into the post-Archaean (C). In the Mesoarchaean, ancient non-convecting domains (ANCD) existed in the upper mantle as remnants of cumulates that crystallised from magma oceans and seas and from very deep melting in hot upwellings. The ANCDs are not envisaged to have been a global barrier but where they were extensive, heat flow from the lower mantle was impeded. A nascent craton with a modest mantle root is shown forming above an ANCD. Occasional upwelling of heat began to erode through the ANCD and lower-mantle derived melts may have metasomatised the cumulates. By the late Neoarchaean, the NACDs had been dislodged by repeated pulses.
of upwellings form the lower mantle. The majority of the barrier was recycled back into the asthenosphere but occasional fragments and melt residues from the NACD became entrained into the Neoarchaean 'superplumes' which fused this material with the shallower proto-cratic root. In post-Archaean events, the base of the lithosphere began to erode and shallow.
Subcalcic garnet present
No subcalcic garnet
ZIMBABWE CRATON
KAAPVAAL CRATON
CAPE FOLD BELT
NAM AQUA-NATAL
Zone of Archaean gt harzburgite affected by later enrichment
200 km
Lithosphere
100 km
Zone of 1.4 Ga enrichment
Asthenosphere
Kamber and Tomlinson Figure 4

Archaean crust
Mobile Belt
Lithosphere
Asthenosphere

Cape Fold Belt
Limpopo Belt
Kheis
Kambalda
Zimbabwe
Limpopo Belt
Kaapvaal Craton
Namaqua-Natal Belt
Superior craton (n=2845)

Western Australian cratons (n=2008)

Kaapvaal craton (n=1623)

Baltic shield (n=1658)

Dharwar craton (n=1091)

North Atlantic craton (n=702)

Zimbabwe, Tanzania, Slave & North China cratons (n=732)

All cratons

MgO wt% (log scale)
All cratons (n=10660)

2.5-2.9 Ga samples (n=1539)

>2.9 Ga samples (n=647)

Kamber and Tomlinson Figure 7
A

<table>
<thead>
<tr>
<th>Location</th>
<th>Age Range</th>
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<tbody>
<tr>
<td>Timiskaming</td>
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<tr>
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<td>Pacaud</td>
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B

C

- **biotite granite**
- **biotite-amphibole granodiorite**
- **biotite granodiorite**
- **white and grey granodiorite**
- **grey orthogneiss**
- **amphibole - biotite tonalite**

- Sproule et al. (2002)
- Herzberg (2016)
- Feng + Kerrich (1990)
- Barrie (2005)

Kamber and Tomlinson Figure 8
Kamber and Tomlinson Figure 9
Kamber and Tomlinson Figure 11
Kamber and Tomlinson Figure 13
Kamber and Tomlinson Figure 14
Asthenosphere
Continental crust
Shallow harzburgite
Deep residue
Magma sea cumulates
Magma ocean cumulates

Kamber and Tomlinson Figure 15
Table 1: Summary of population results of KDE analysis

<table>
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<tr>
<th>Region</th>
<th>MgO wt%</th>
<th>% MgO wt%</th>
<th>% MgO wt%</th>
<th>% MgO wt%</th>
<th>% MgO wt%</th>
<th>% MgO wt%</th>
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Table 2: Mineral compositions and REE concentrations (ppm)

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<th>MnO</th>
<th>CaO</th>
<th>Na₂O</th>
<th>K₂O</th>
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<td>0.00</td>
<td>17.76</td>
<td>0.83</td>
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<td>0.00</td>
<td>24.80</td>
<td>0.41</td>
<td>0.00</td>
<td>99.76</td>
</tr>
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<td>orthopyroxene (n=18)</td>
<td>58.16</td>
<td>0.00</td>
<td>0.73</td>
<td>0.01</td>
<td>35.87</td>
<td>4.22</td>
<td>0.03</td>
<td>0.04</td>
<td>5.24</td>
<td>0.00</td>
<td>0.00</td>
<td>101.45</td>
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<tr>
<td>olivine (n=5)</td>
<td>58.08</td>
<td>0.00</td>
<td>0.79</td>
<td>0.00</td>
<td>36.96</td>
<td>4.22</td>
<td>0.03</td>
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<td>5.24</td>
<td>0.00</td>
<td>0.00</td>
<td>101.13</td>
</tr>
</tbody>
</table>

Table 2. Mineral compositions and REE concentrations (ppm)
Supplemental text

Click here to download Background dataset for online publication only: Supplemental_Text_S1.docx
Supplemental data 1
Click here to download Background dataset for online publication only: Table_S1_Experimental_data_compilation with calcs.xlsx
Supplemental data 2

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