High water contents in the Siberian cratonic mantle linked to metasomatism: an FTIR study of Udachnaya peridotite xenoliths

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# Abstract (479 words)

The processes that control water distribution in nominally anhydrous minerals from peridotites are twofold. Melt depletion will remove water while metasomatism can potentially add water to these minerals. These processes can lead to a wide range of outcomes in water contents, which in turn could play a role in mantle rheology and long-term cratonic root stability. To examine these complexities, water concentrations in minerals from well-characterized peridotites from the Udachnaya kimberlite in the central Siberian craton were analyzed by FTIR. The peridotites span a complete top-to bottom cross-section of typical cratonic lithospheric mantle (2-7 GPa and 700-1400°C). Diffusion modeling of water content profiles across olivine grains shows that water loss during decompression is limited to the 100 μm rims of olivines; the cores preserved their mantle water contents. Water contents range from 6 to 323 ppm wt H2O in olivine, 28 to 301 ppm H2O in orthopyroxene (opx), 100 to 272 ppm H2O in clinopyroxene (cpx) and 0 to 23 ppm H2O in garnet. Melting modeling cannot reproduce the high water contents of cratonic mantle peridotites and any potential partial melting trend must have been erased by later events. The water contents of minerals, however, are correlated with modal abundances of clinopyroxene and garnet, bulk rock FeO, TiO2 and SiO2 as well as with light and middle rare earth elements in clinopyroxene and garnet. These relationships are best interpreted as interaction of residual, melt-depleted peridotites with silicate melt, which produced modal and cryptic metasomatism. Importantly, the water enrichment in the Siberian cratonic mantle took place prior to kimberlite magmatism and eruption. Water addition by metasomatism occurred from pressures > 4 GPa all the way to the base of the cratonic root below central Siberia, but was limited to shallower levels (< 5 GPa) in the Kaapvaal cratonic lithosphere. The difference in olivine water contents at the deepest levels of the Kaapvaal (< 5 ppm H2O) and Siberian (6-323 ppm H2O) cratonic roots may be linked to oxygen fugacity and resulting fluid speciation or, alternatively, to reaction with different metasomatic agents. Calculated viscosities for the deepest Udachnaya samples are similar to those inferred for the asthenosphere. If these xenoliths are representative of the deep cratonic lithosphere, water is not as important a parameter as previously thought in the strength of cratonic lithosphere, otherwise the cratonic root beneath Udachnaya would have been delaminated. Alternatively, the metasomatic xenoliths may not be representative of the Siberian cratonic root and kimberlites preferentially sample cratonic mantle lithosphere material located near, and metasomatized by, melt conduits, which served as channels for upward migration of water-rich melts and fluids including kimberlites. In that case, the cratonic root overall still may have relatively low water contents, which in addition to its less metasomatized (more refractory) and thereby buoyant nature, still play a role in making it strong enough to resist delamination by the surrounding asthenosphere.

Key words: water, mantle, xenoliths, peridotites, Siberia, craton

1. **INTRODUCTION**

Water is believed to be a key factor controlling the viscosity and hence long-term stability of the cratonic mantle lithosphere (Dixon et al., 2004; Hirth et al., 2000; 1986). Water is incompatible such that during partial melting its removal should have made the residual cratonic mantle essentially “dry” (Aubaud et al., 2004; Aubaud et al., 2008; Grant et al., 2006; Grant et al., 2007b; Hauri et al., 2006; Tenner et al., 2009). Water in the mantle primarily resides in nominally anhydrous olivine, pyroxene and garnet (Asimow and Langmuir, 2003; Bell and Rossman, 1992b; Hirth and Kohlstedt, 1996). Water enters these anhydrous minerals typically as protons attached to structural oxygen in intrinsic crystal defects (Bell and Rossman, 1992b; Rossman, 1996; Smyth et al., 1991). Olivine becomes stronger if its water content decreases (Mackwell et al., 1985). This property of olivine may be a contributing factor in explaining why cratonic mantle roots remain strong and resist delamination by the asthenosphere for billions of years (Dixon et al., 2004; King, 2005; Mackwell et al., 1985; Peslier et al., 2010; Pollack, 1986). The notion of a craton-wide dehydration was questioned by recent evidence from Kaapvaal peridotites, which appears to indicate a layer with water-poor olivines (< 1 ppm H2O) at P >6.4 GPa, while shallower mantle levels record variable water enrichments by fluids or melts (Baptiste et al., 2012; Peslier et al., 2010). It was thus suggested that the survival of the Kaapvaal mantle lithosphere was due to a mechanically resistant layer at the base of the lithosphere made primarily of “dry” olivine (Peslier et al., 2010). A recent experimental study has challenged the long-held assumption that olivine becomes weaker as water contents increase (Fei et al., 2013) such that water may not have a significant effect on the strength of olivine. If so, other factors that stabilize cratons (density, temperature, surrounding mobile belts (Jordan, 1975; King, 2005)) may be more important.

Whether or not water in olivine contributes to a rheological contrast between cratonic lithosphere and asthenosphere, it is crucial to determine whether the water distribution reported for the Kaapvaal mantle is seen in other cratons or if this observation is unique. Water distribution in the upper mantle also has potential influence on the interpretation of seismological, electrical and thermal conductivity data, and constrains the understanding of Earth’s global water cycle (Bodnar et al., 2013; Fullea et al., 2011; Jones et al., 2013; Jones et al., 2012; Karato and Jung, 1998; Li, 2011; Selway, 2014; Selway et al., 2014).

Here new data are presented on water in peridotite xenoliths from the Udachnaya kimberlite in the central Siberian craton. The xenoliths analyzed are well characterized for major and trace elements and have equilibration pressures and temperatures spanning a wide range, 2-7 GPa and 700-1400ºC. This is the largest depth range that can normally be obtained from cratonic peridotite xenoliths. Moreover, by contrast to the Kaapvaal where water data were obtained on xenoliths from five kimberlite pipes, the peridotites studied here are from a single kimberlite. Focusing on peridotites from a single mantle domain in the Siberian craton can better establish what processes control the water contents in cratonic peridotites, without regional and temporal differences complicating the picture. Finally, the role of water in the longetivity of the Siberian craton and the implications of these results to cratons in general are considered.

1. **GEOLOGICAL SETTING**

The Siberian craton is mainly covered by Paleozoic sediments and Permian-Triassic flood basalts. The Anabar and Aldan shields, exposed portions of the basement in the northern and southeastern parts of the craton (KML map), contain gneisses and granulites with ages up to 3·0-3·6 Ga (Jahn et al., 1998; Nutman et al., 1992; Zonenshain et al., 1990). The Archean and Paleoproterozoic blocks amalgamated in the mid-Proterozoic (1·8-2·0 Ga) (Rosen et al., 2005). The oldest Re-depletion ages obtained for peridotite xenoliths from the Siberian craton are 3-3.2 Ga (Pearson et al., 1995b). The ages of the oldest crustal and mantle rocks in Siberia are similar to those determined for the oldest crustal and mantle rocks in the Kaapvaal craton (de Wit and al., 1992; Pearson et al., 1995a; Walker et al., 1989). The lithosphere-asthenosphere boundary beneath the Siberian craton is at ~7 GPa (i.e. 220 km) as inferred from seismic data (Kozhevnikov and Solovei, 2010; Priestley and Debayle, 2003) and from the deepest peridotites sampled by kimberlites (Boyd et al., 1997; Goncharov et al., 2012; Ionov et al., 2010).

Over a thousand kimberlite bodies are known in the Siberian craton but few of them (mainly Udachnaya, Mir and Obnazhennaya) contain peridotite xenoliths suitable for petrologic and geochemical studies (Howarth et al., 2013; Sobolev, 1977). The Udachnaya kimberlite hosts the most abundant and varied xenolith suite of the Siberian craton (Ionov et al., 2010; Pearson et al., 1994; Pokhilenko et al., 1999; Sobolev et al., 1984). It is subdivided in two complex bodies made up of breccia, porphyritic and vein kimberlites, the smaller East and the larger West pipes (Sobolev, 1977; Spetsius and Serenko, 1990; Zinchuk et al., 1993) erupted about 360 Myr ago (Kinny et al., 1997) through the Archean-Proterozoic Daldyn block of the central Siberian craton (Rosen et al., 1994). The peridotite xenoliths in this study are from the Udachnaya-East kimberlite (66°26’ N, 112°19’E). The samples were collected in 2003–2009 in the 420–640 m depth range near the center of the pipe. They are hosted by well-preserved type-I kimberlites, which contain fresh, unserpentinized olivine and groundmass and are unaffected by syn- and post-magmatic alteration (Kamenetsky et al., 2012; Kamenetsky et al., 2008; Kamenetsky et al., 2009a; Kamenetsky et al., 2009b).

1. **SAMPLE OVERVIEW**
	1. **Sample description**

Six spinel peridotites and 16 garnet peridotites were selected among previously well-characterized Udachnaya xenoliths (Doucet et al., 2013; Doucet et al., 2012; Ionov et al., 2010). These samples span the complete pressure (P) and temperature (T) range (~2-7 GPa; 735-1341°C, Fig. 1) of the lithospheric mantle beneath Udachnaya inferred from xenolith P-T estimates and seismic data (Boyd et al., 1997; Goncharov et al., 2012; Ionov et al., 2010; Kozhevnikov and Solovei, 2010; Priestley and Debayle, 2003). They also show a representative range of textures, modal, and chemical compositions (Boyd et al., 1997; Doucet et al., 2013; Doucet et al., 2012; Ionov et al., 2010). All minerals, including garnets, have homogenous major, minor and trace element compositions. A summary of petrologic data on the xenoliths in this study is given in Table 1, details are provided in the Electronic Annex 1 (EA1).

The spinel peridotites are harzburgites with coarse-grained microstructures (Doucet et al., 2012; Ionov et al., 2010). Four have orthopyroxene abundances < 30% and low modal clinopyroxene (0 to 3.3%). They are referred to as low-opx harzburgites in this study. Two other xenoliths have much higher modal orthopyroxene (35-43%) and clinopyroxene (3-5%) and are referred to as high-opx harzburgites. The low-opx spinel harzburgites were interpreted to be pristine residues of partial melting that initially formed the Siberian cratonic roots (Doucet et al., 2012) i.e. the range of their Al2O3 and FeO contents match lines of equal degrees of melt extraction at either ~45% anhydrous batch melting or ~38% anhydrous polybaric fractional melting starting at 7-4 GPa and ending at ≤1-3 GPa (EA2 Fig. 1). The high-orthopyroxene and hence, low-olivine, spinel harzburgites are comparatively rich in SiO2 (46.5 to 47.9 %) and low in FeO (5.61 to 6.45%) but have high Mg# [(Mg/ (Mg+Fe) at] of 0.923 to 0.929 (Table 1). The origin of high-opx spinel harzburgites in cratonic mantle continues to be debated (Herzberg and Rudnick, 2012; Kelemen et al., 1998; Pearson and Wittig, 2008; Simon et al., 2007). Those from Udachnaya have been attributed to metamorphic segregation into olivine-rich and orthopyroxene-rich domains during gravity-driven upward transport of initially higher-P melting residues during accretion of cratonic nuclei (Boyd et al., 1997; Doucet et al., 2012).

The garnet peridotites range from harzburgite to lherzolite (Table 1). Based on their microstructures, they are grouped into coarse, transitional and sheared types (Ionov et al., 2010). Eleven out of 16 garnet peridotites in this study are “coarse” grained peridotites. Two are transitional (low-deformation) peridotites that contain only small amounts (1 to 10 wt.%) of olivine neoblasts and sub-grains surrounding coarse, strained olivine (Doucet et al., 2013). Three samples are sheared peridotites characterized by porphyroclastic microstructures with olivine as fine-grained neoblasts (up to 90%) and relics of strained crystals with abundant sub-grains (Agashev et al., 2013; Ionov et al., 2010). The coarse garnet peridotites were interpreted as ranging from near-pristine melting residues to metasomatized, Fe–Ti-enriched peridotites (Doucet et al., 2013). About half of the coarse peridotites are similar in modal and major compositions to low-opx Udachnaya spinel harzburgites (EA2 Fig. 1) and may have formed at similar to slightly lower melting degrees and pressures than the spinel harzburgites (Doucet et al., 2013). The remaining coarse peridotites as well as transitional and sheared peridotites experienced modal metasomatism, which precipitated clinopyroxene and garnet with enrichments in light to middle rare earth elements, and produced enrichments in Fe, Ti and Al in bulk rocks (Doucet et al., 2013). The metasomatism can be linked to a range of silicate- and carbonate-rich liquids; the last event, responsible for the deformation, appears to have occurred before the host kimberlite eruption (Agashev et al., 2013; Ionov et al., 2010).

* 1. **Thermal state and oxygen fugacity**

The thermal and redox state of the mantle beneath Udachnaya was reported and discussed in detail by Goncharov et al. (2012) (Table 1, Fig. 1) as well as Yaxley et al. (2012). In order to insure the coherence of our P-T dataset with previous studies on water in cratonic mantle, the equilibration P-T estimates for garnet peridotites were recalculated using the opx-cpx solvus thermometer (TBK90) and the Al-in-opx barometer (PBK90) of Brey and Köhler (1990). These are the most commonly used thermo-barometers for cratonic peridotites. For comparison, we also present in Table 1 the P-T estimates using the opx-cpx thermometer of Taylor (1998) and the opx-garnet barometer of Nickel and Green (1985), which are considered the most accurate in P-T estimates for cratonic mantle peridotites (Nimis and Grütter (2010).

The PBK90-TBK90 estimates (Fig. 1a) define a broad P-T range for coarse peridotites from 2.2 to 6.9 GPa and 735 to 1289°C, respectively. In contrast, deformed peridotites are only found at 6.3 to 6.6 GPa and 1281-1341°C, i.e. both deformed and coarse peridotites were present near the base of the lithosphere at the time of kimberlite eruption. The P-T values obtained from other methods and using different samples yield a generally similar P-T range (Boyd et al., 1997; Doucet et al., 2013; Goncharov et al., 2012; Ionov et al., 2010). The data scatter between the 35 and the 45 mW/m2 model conductive geotherms (Pollack and Chapman, 1977) and define no single P-T gradient.

The oxygen fugacity (*f*O2) of peridotites was calculated from Fe3+/∑Fe in garnet and spinel determined by Mössbauer analyses (Goncharov et al., 2012) using the calibration of Stagno et al. (2013) for garnet and Wood (1991) for spinel, and presented relative to the fayalite-magnetite-quartz buffer in logarithmic units (∆FMQ). For comparison we also present in Table 1 the *f*O2 of Udachnaya garnet peridotites estimated after Gudmundson and Wood (1995) for garnet. The *f*O2 values for the garnet peridotites in this study decrease from -0.9 to -3.6 ∆FMQ with increasing depth in the lithospheric mantle (Fig. 1b).

1. **METHODS**
	1. **Sample preparation**

Ninety to five hundred gramsof fresh material was taken from xenolith cores and crushed to < 5-10 mm diameter pieces in a steel jaw crusher. Twenty to thirty grains (or portions of grains) of olivine, pyroxene and garnet containing as few inclusions or cracks as possible were handpicked from sieved >1 mm size fractions using a binocular microscope. The mineral grains were set in epoxy prior to being polished on two parallel surfaces at the Astromaterials Research and Exploration Science (ARES) directorate of the NASA-Johnson Space Center, at the Department of Earth and Atmospheric Sciences of the University of Houston and at the Geology department of J. Monnet University in St Etienne. Each olivine and pyroxene grain was examined with an optical polarizing microscope to determine its orientation. Grains showing Bxo, Bxa or optic-normal interference figures in convergent polarizing light were selected for analysis by Fourier transformed infrared spectroscopy (FTIR). The thickness of the minerals was determined using a Mitutoyo digimatic micrometer and ranges from 300 to 800 µm (± 3 µm).

* 1. **FTIR**

A Hyperion 3000 microscope in combination with a Bruker Vertex 70 FTIR was used at ARES at NASA- Johnson Space Center to detect water in minerals. Areas of 10 ×10 to 100 × 100 µm (depending on the size of the area available for a clear path through the minerals) were analyzed by transmission in the mid-infrared range (wavenumber 4000-600 cm-1) using a resolution of 4 cm-1 and 128 scans for olivine and pyroxenes and 200 scans for garnet. Several spots were analyzed in the interior of each grain and at least one spot at the edge. Olivine and pyroxene grains were analyzed twice in two perpendicular orientations with the polarizer (a Zn-Se wire grid) parallel to the optical indicatrix α, β, or γ which ensures precise quantification of water in anisotropic minerals ([Libowitzky and Rossman, 1996](#_ENREF_62)). Garnets, being isotropic, were analyzed unoriented in non-polarized infrared light***.*** Non-polarized FTIR analyses were also performed on randomly oriented olivines and orthopyroxenes from 13 and 4 samples, respectively following the method of Kovacs et al. (2008) where as many grain cores as possible (> 10) were analyzed (EA1 Table 1). The baseline for each spectrum was drawn using the Nicolet Omnic software and using the spline baseline correction. This method results in systematic errors of ≤ 20% (2σ) (Peslier et al., 2002). The following expression of the Beer-Lambert law is used to obtain water contents:

[1] $C\_{H\_{2}O}=A/l'$

where *CH2O* is water concentration (in ppm weight H2O), *A* is the absorbance values (in cm-2, normalized to 1 cm thickness) which correspond to the area beneath the O-H vibration bands, and *I’* is the integral specific absorption coefficient which is mineral composition specific (in ppm-1 cm-2). For polarized FTIR analysis of olivine and pyroxenes, *A* is the sum of the area in the three perpendicular directions (Aα+Aβ+Aγ). For non-polarized FTIR analysis of olivine and orthopyroxene, *A* is three times the average of absorbance values obtained for individual grains (Kovacs et al., 2010). Absorption coefficients used for olivine are I*’* = 5.31 ± 0.01 (2σ and from hereon) ppm-1 cm-2 between 3800 and 3400 cm-1 and I’ = 2.22 ± 0.01 ppm-1 cm-2 between 3400 and 3100 cm-1 (Bell et al., 2003). For comparison we also we also used the absorption coefficient of olivine of l’ = 8.4 ± 0.4 ppm-1 cm-2 between 3800-3100 cm-1 determined by the recent study of Withers et al. (2012). Absorption coefficients used for pyroxenes are I’ = 14.84 ± 0.59 ppm-1 cm-2 between 3800 and 2600 cm-1 for orthopyroxene and I’ *=* 7.09± 0.32 ppm-1 cm-2 between 3700 and 2600 cm-1 for clinopyroxene(Bell et al., 1995). We calculate the water content in garnet using two absorption coefficients: I’ = 0.39 ± 0.14 ppm-1 cm-2(Bell et al., 1995) and I’ = 0.81 ± 0.1 ppm-1 cm-2  (Maldener et al., 2003) between 3700 and 3400 cm-1. Sources of uncertainties have been discussed by Peslier et al. (2002) and Peslier and Luhr (2006) and are included in an error propagation formula to obtain the 2σ error bars on water contents shown in figures of this paper.

1. **RESULTS**
	1. **Olivine**

Absorption bands due to O-H vibrations in olivines of twenty Udachnaya peridotite xenoliths are located between 3700 and 3100 cm-1 (EA2 Fig. 2a, c and e). These spectra are similar to published OH spectra of olivines from kimberlite-hosted mantle xenoliths from the Kaapvaal (Bell et al., 2003; Peslier et al., 2008) and Siberian cratons (Matsyuk and Langer, 2004). Most of the olivines have OH bands that may in part be caused by non-intrinsic nano-inclusions (EA2 Fig. 3): rare talc (bands at 3677 cm-1), common serpentine (3640 cm-1) and even Ti-clinohumite (3572 and 3525 cm-1), though the latter bands overlap with those caused by intrinsic OH defects (Khisina and Wirth, 2008; Khisina et al., 2008; Miller et al., 1987; Mosenfelder et al., 2006). This overlap makes it impossible to calculate separately the potential total contribution to the total water content of intrinsic versus hydrous phase inclusions. At any rate, studies on nano-inclusions in olivine megacrysts from the Udachnaya kimberlite suggest that these inclusions are formed either in mantle environments (Khisina et al., 2008) or through decompression-induced autoserpentinization during kimberlite eruption owing to interaction between the inclusion material and the host olivine matrices (Khisina et al., 2008; Matsyuk and Langer, 2004). We consequently interpret all water detected, whether intrinsic and non-intrinsic, as present in the olivines before entrapment in the host kimberlite magma. In three out of twenty samples, the olivine water contents were estimated using the non-polarized FTIR method (Uv-604/09, U501 and U71) because our grain mounts did not contain any crystals oriented parallel to the α electric vector of the incident infrared light. Nevertheless, the comparison between polarized and non-polarized FTIR analyses on nine samples in this study show similar olivine water contents within 20% (EA2 Fig. 4), which is the typical 2σ error for non-polarized FTIR (Kovacs et al., 2008). For three out of twenty samples the differences in water contents between polarized and non-polarized FTIR analyses exceed 20% (EA1 table 1; EA2 Fig. 4). These differences may be explained by undetected nano-fluid inclusions. Water in fluid inclusions result in wide absorption bands centered at 3420 cm-1. We did not observe such a band in the three samples. However, if the fluid inclusions are not numerous, such a band may be undistinguishable from the baseline. The water contents of olivine in these three samples probably lie between polarized and non-polarized FTIR estimates.

All olivines have less water at grain edges compared to their cores (Fig. 2 and EA2 Fig. 5). In half of our samples the water content decrease is limited to ≤ 200 µm from the rims while water contents are homogeneous in the cores (Fig. 2 and EA2 Fig. 5).The edges have 15 to 60 % less water compared to the cores. Water contents in the cores of olivine grains vary by 1-5% within each xenolith. The water contents (Table 2 and EA1 Table 1) calculated using absorbance from spectra obtained in the cores of olivine grains and absorption coefficient of Bell et al. (2003) range from 6.5 to 323 ppm H2O (6.5 to 47 ppm for low-opx spinel peridotites, 14 to 20 ppm for high-opx spinel peridotites, 7 to 323 ppm for coarse garnet peridotites, 212 to 305 ppm for transitional garnet peridotites and 70 ppm for the only sheared garnet peridotite analyzed (U503)). The water contents of olivine calculated using the absorption coefficient of Withers et al. (2012) are a third to half lower than those obtained with Bell et al. (2003) absorption coefficient (EA1 Table 1). The Bell et al. (2003) absorption coefficient is used preferentially in the present paper because it is the one used by most research groups working on water in olivine (Peslier, 2010), which facilitates the comparison of our data with those from the literature.

Water contents in olivine appear to be linked to Mg# in olivine and modal clinopyroxene (Fig. 3a and b). Water contents in olivine from the Udachnaya peridotites show no correlation with depth (Fig. 4a), temperature or oxygen fugacity (Fig. 5a). There are no correlations with the abundances of other major and trace elements in whole-rocks and minerals or with modal abundances of olivine, orthopyroxene and garnet (Tables of EA1).

* 1. **Pyroxenes**

Thirteen peridotite xenoliths were analyzed for water in orthopyroxene and four samples for water in clinopyroxene. Water contents could not be analyzed in pyroxenes from the remainder of our samples because of the small size of the grains and the abundance of fractures, fluid inclusions and exsolution lamellae in them (EA2 Fig. 6). These features make it impossible to analyze only intrinsic water with the current FTIR analytical protocols. The FTIR spectra (EA2 Fig. 2b,d, f and h) resemble those observed in orthopyroxene and clinopyroxene from other peridotite xenoliths found in kimberlites (Grant et al., 2007b; Mosenfelder and Rossman, 2013a; Mosenfelder and Rossman, 2013b; Peslier et al., 2012; Rossman, 1996; Skogby et al., 1990). Some orthopyroxenes have OH bands between 3683 and 3677 cm-1 that may result from non-intrinsic inclusions of amphibole (Skogby et al., 1990)(EA2 Fig. 3). Polished grain surfaces oriented perpendicular to the β indicatrix were never available in grain mounts of orthopyroxene Uv-604/09 and clinopyroxene U64. The non-polarized FTIR method could not be used because there was not enough pyroxene without fractures and inclusions in the grain mount to obtain statistically valid results (Kovacs et al., 2008). Instead, to calculate the water contents of orthopyroxene Uv-604/09 and clinopyroxene U64 we assumed the absorbance value along the indicatrix α to be similar to the absorbance value along the indicatrix β. This assumption, applied to samples for which absordances along α, β and γ are known, results in calculated water contents similar within the typical error of 20% to those with only two measured directions (EA2 Fig. 7). Orthopyroxene and clinopyroxene show no evidence for water loss at grain boundaries (EA2 Fig. 5i-j). Orthopyroxene water contents of Udachnaya peridotite xenoliths range from 28 to 301 ppm H2O, i.e. 30 ppm for a low-orthopyroxene spinel peridotite (U24), 28 to 301 ppm for coarse garnet peridotites, 298 ppm for a transitional garnet peridotite (U71), and 103 to 123 ppm for sheared garnet peridotites. Water contents in the clinopyroxenes range from 100 to 272 ppm: 208 to 272 ppm for coarse garnet peridotites and 100 ppm for sheared garnet peridotite (U503). The water contents in these pyroxenes are typical for cratonic mantle xenoliths elsewhere (Peslier, 2010). Water in orthopyroxene correlates with modal orthopyroxene in coarse garnet peridotite (R²=0.7, Fig. 3c), except for sample U1147, for which no water was detected in garnet. Water content in pyroxene does not correlate with depth (Fig. 4b and d) or other geochemical parameters (Tables of EA1). Water in clinopyroxene correlates with Ce/Yb in clinopyroxene (R²=0.9, Fig. 3d) but does not correlate with other geochemical parameters (Tables of EA1).

* 1. **Garnet**

The garnets from nine Udachnaya peridotite xenoliths were analyzed for water (Table 2). The majority of the samples could not be analyzed because of the small size of garnet grains as well as ubiquitous fractures, inclusions and alteration rims (i.e. kelyphite) (EA2 Fig. 6). Nearly all the garnets in this study have detectable water, except U1147 (EA2 Fig. 2g). These FTIR spectra are similar to those observed in garnet from earlier work on kimberlite-hosted mantle xenoliths from the Siberian craton (Matsyuk et al., 1998). The majority of our garnets have their main OH band centered at 3572 cm-1 (EA2 Fig. 2b) that may be linked to non-intrinsic inclusions of Ti-clinohumite (Miller et al., 1987). As in Kaapvaal xenolith garnets (Peslier et al., 2012), such a band is more conspicuous for garnets with >1000 ppm of Ti. The majority of garnets have homogenous water distribution and only a third shows a decrease in water content whithin 200 microns of the grain edges (EA2 Fig. 5k-l). The majority of garnets in Udachnaya collection have homogeneous major, minor and trace elements composition. However, Ionov et al. (2010) show that some of the largest garnets (2-3 mm) from the sample U267 have a range of TiO2 contents higher at the rims than in the cores. This may be linked to the thick layers of kelyphite that usually rim garnet grains. Garnets from U267 analyzed here are 0.5 to 1.5 mm in size; they have homogenous water distribution in the core of grains and show decrease in water content at the grain edge, in the same order of magnitude than for garnet from U85 (EA2 Fig. 5k) for which a complete water profile is available.

Udachnaya peridotite garnets contain 0 (U1147) to 23 ppm H2O: 0 to 11 ppm for coarse garnet peridotites, 16 ppm for a transitional garnet peridotite (U71) and 16 to 23 for sheared garnet peridotites. These ranges are similar to those previously reported for garnet from other peridotite xenoliths from the Siberian and Kaapvaal cratons (Bell and Rossman, 1992a; Matsyuk et al., 1998; Peslier et al., 2012). This is consistent with the observation of [Bell and Rossman (1992a](#_ENREF_9)) on Kaapvaal craton peridotites that garnets in deformed peridotites contain more water than those in coarse garnet peridotites. Water contents of garnet in this study correlate with a few parameters shown in Fig. 6a-f. Generally, water in garnet can be linked with REE patterns in clinopyroxene and garnet (Fig. 7). Finally, we find no correlations of water contents in garnet with depth (Fig. 4c), temperature or oxygen fugacity (Fig. 5d).

* 1. **Bulk-rock water contents**

Bulk-rock water contents of the peridotites can be estimated by mass balance using the water contents of their constituent minerals and modal abundances. This could only be done for 3 samples in this study (U64, U1188 and U71) given that pyroxene and garnet could not be analyzed concurrently in the other samples as noted above. Bulk-rock water contents range from 92 to 293 ppm H2O and show a negative correlation with Mg#Ol and positive correlation with modal clinopyroxene (Table 2 and EA2 Fig. 8). Using olivine water contents calculated with the absorption coefficient of Withers et al. (2012), the bulk rock water estimates range from 16 to 135 ppm (EA1 Table 1).

1. **DISCUSSION**
	1. **On the preservation of mantle water contents during host kimberlite ascent**

Hydrogen diffuses rapidly through nominally anhydrous minerals at magmatic temperatures (T>800°C) (Demouchy, 2010; Demouchy and Mackwell, 2003; Kohlstedt and Mackwell, 1998; Mackwell and Kohlstedt, 1990). The H2O-rich kimberlite magma could potentially add water to the xenoliths, or alternatively, hydrogen could be lost from the xenoliths via re-equilibration with degassing host kimberlite magma during decompression. To explore the distribution of water and processes controlling the water contents in the mantle it is crucial to know if the mineral grains preserved the water contents they had in the mantle prior to incorporation as xenoliths in kimberlite magmas.

* + 1. *Assessment of potential water addition by kimberlite magma*

If kimberlite magmas are water-rich (Kjarsgaard et al., 2009; Kopylova et al., 2007; Le Roex et al., 2003; Price et al., 2000), hydrogen may be added from the magma to entrapped xenoliths and their constituent minerals. Whether kimberlite magmas contain volatiles from the time they are extracted from the mantle or acquire them via hydrothermal processes near the surface is a matter of debate, as is the depths at which degassing of a rising kimberlite magma may occur (Sparks et al., 2006; Stripp et al., 2006). A predictive outcome of H addition from outside sources into the xenolith minerals are diffusion profiles characterized by more H at the edges and lower but measurable amounts in the cores, similar to what is observed in diffusion experiments (Demouchy and Mackwell, 2006; Ingrin and Blanchard, 2006). In the majority of Udachnaya xenoliths where diffusion profiles were reported, the opposite is observed, i.e. the grain rims have lower H contents than the cores. This is observed at grain boundaries of all olivines and a third of the garnets in this study, whereas orthopyroxene, clinopyroxene, and the remainder of the garnets have homogeneous water contents (EA2 Fig. 5). Moreover, hydrogen addition from the host kimberlite magma is not consistent with the broad range of water contents in minerals from different xenoliths (Table 2) especially for olivine (6.5 to 323 ppm, Fig. 3) and garnet (0 to 23 ppm, Fig. 6). One may expect all xenoliths to have similar water contents in their minerals if that water came from the kimberlite magma with given volatile contents. The absence of water addition to the xenoltihs from the host magma is best explained by the fast ascent of the magma (several hours according to estimates of ascent rate of kimberlite magma from Peslier et al. (2008) and our own data, see below) leaving no time for significant exchange of water with the entrapped xenoliths. Alternatively, the parental melt of the Udachnaya kimberlite may have had a low water content as advocated by Kamenetsky et al. (2012). The latter is consistent with the fact that xenoliths that contain small amounts (<< 5%) of interstitial phlogopite group minerals (interpreted as related to infiltration of host magmas (Sharygin et al., 2012)) are rare while xenoliths that show kimberlite infiltration material without hydrous minerals is more common (Doucet et al., 2013).

* + 1. *Water loss during decompression*

The low H contents in the olivine grains at the rims compared to the cores are interpreted to result from H diffusing out of grains during transport of the xenolith by the host magma (Demouchy et al., 2006; Peslier and Luhr, 2006; Peslier et al., 2008). The same 2D-diffusion modeling as in (Peslier et al., 2008) was applied following diffusion equation of Shewnon (1983) and diffusion parameter determined by Demouchy and Mackwell (2006) (details and are given in Electronic Annex 3).

The best fit to the data is obtained using the water content measured at the core as initial water content (solid lines in Fig. 2 and EA2 Fig. 5a-d). By contrast, using 20% higher initial water content does not reproduce the water profile in olivine grains (dotted lines in Fig. 2 and EA2 Fig. 5a-d). The major conclusion of this modeling is that the cores of the olivine grains preserved their mantle water contents.

* + 1. *Kimberlite ascent rates*

Time estimated from the best fit to diffusion profiles can be used to calculate host magma ascent rates (Demouchy et al., 2006; Peslier and Luhr, 2006; Peslier et al., 2008). If the depth of equilibration of the peridotites is used as the start of degassing, 1 to 87 m.s-1 are obtained for the ascent rates. However, it is a matter of debate at what depth volatiles start to be exsolved from the kimberlite magma and from the xenoliths it transports (Canil and Fedortchouk, 1999; Sparks et al., 2006). If we assume water starts degassing at the same time CO2 does (Wilson and Head, 2007), then a depth of 60 km can be used for the inception of H loss, the kimberlite ascent rates are 0.5-25 m.s-1.

These ascent rates are approximations because the velocity of the kimberlite magmas could have varied during the ascent to the surface, in particular due to decompression which decreases the CO2 solubility and increases the ascent rates (Russell et al., 2012). However, the ascent rates are on the same order of magnitude as estimates for kimberlite magmas derived from other methods, with the xenoliths being transported from the mantle to the surface in a matter of hours to days (Canil and Fedortchouk, 1999; Downes et al., 2006; McCallister et al., 1979; McGetchin and Besancon, 1973; Sparks et al., 2006).

* 1. **Estimates of equilibrium water distribution between peridotite phases and coexisting melts.**

The water contents measured in peridotite minerals allow (1) comparison of the inter-mineral water partition coefficients with experimentally determined data, and (2) calculation of the water content of a melt in equilibrium with these minerals using experimentally determined partition coefficients.

* + 1. *Water equilibrium between minerals*

Partitioning of water between pyroxene and melt depends on their Al contents and also is temperature dependent(Aubaud et al., 2004; Aubaud et al., 2008; Grant et al., 2007b; Hauri et al., 2006; O'Leary et al., 2010; Tenner et al., 2009). We calculated the orthopyroxene-melt and clinopyroxene-melt partition coefficients using the equations of O’Leary et al. (2010) parameterized as a function of temperature and Al and Ca contents of the pyroxenes (Table 2 of EA2). Experimental studies and thermodynamic models suggest dependence of olivine-melt water partitioning on pressure(Hirschmann et al., 2009; Kohlstedt et al., 1996; Tenner et al., 2009). However, in the absence of a specific scheme for calculating H partition coefficients for the variety of possible mantle fluids and conditions of our samples we adopt here the partition coefficients for olivine (0.0006) and garnet (0.0033) of Tenner et al. (2009). These were determined experimentally at 5 GPa and 1350 °C, which is relatively close to the equilibration conditions of most of our peridotites (Table 1). We then calculate the water partition coefficients between minerals by dividing two experimentally determined mineral-melt partition coefficients (DH2Omineral/mineral exp.). Finally, bulk peridotite partition coefficients (DH2Operidotite/liquid) were determined using the partition coefficient of each mineral and the modal abundances of our samples.

The data on Udachnaya peridotites could also provide a inter-mineral partition coefficients if the minerals are in equilibrium for water. We determined the ratio of water contents between two minerals (DH2Omineral/mineral Udachnaya), for example DH2Ool/opx being the ratio of water contents in olivine and orthopyroxene from the same sample. We then compare these values to the DH2Omineral/mineral calculated to see if the minerals are in equilibrium (Fig. 8). The inter-mineral ratios of water contents are extremely variable, ranging from 0.086 to 1.63 for DH2Ool/opx, from 1.65 to 75 for DH2Ool/gar, from 0.73 to 1.45 for DH2Oopx/cpx and from 5.3 to 85 for DH2Oopx/gar (EA1 Table 4).

Water content ratios between olivine and orthopyroxene (average DH2Ool/opx of 0.59 ± 0.02) are higher than the experimental partition coefficients (0.10 ± 0.05, EA1 Table 4)(Fig. 8a). In addition, the highest values for DH2Ool/opx are similar to recent experimental determinations (~1.67 at 6 GPa; (Withers et al., 2011)), and the average value for Udachnaya peridotite DH2Ool/opx is similar to DH2Ool/opx determined from Kaapvaal megacrysts formed at 5 GPa (Bell et al., 2004). One possible explanation for the range of DH2Ool/opx is that the water content of minerals was disturbed after major element equilibration was achieved (water addition to olivine or water loss from orthopyroxene). Our diffusion modelling of olivine water data above has demonstrated that water loss from olivine occurred only at the rim of some olivines. However, as the water content at the core of olivines were used to calculate the DH2Ool/opx , the discrepancy in DH2Ool/opx between Udachnaya data and experimental values cannot be due to water loss from olivine during xenolith entrainment by kimberlite magma. Another possibility is that the discrepancy results in part from the difficulty in determining the water partition coefficients between olivine and melt from high pressure experiments (>3 GPa) (Hirschmann et al., 2009). Most likely, however, the water contents of olivine and/or orthopyroxene could have been disturbed during metasomatism as discussed in section 6.3.2.

The opx/cpx water ratio measured in Udachnaya peridotites (average DH2Oopx/cpx Udachnaya of 1.19 ± 0.04) is two to three times higher than experimentaly determined values (average DH2Oopx/cpx Udachnaya of 0.61 ± 0.09 for samples for which water in pyroxene is available; average value of 0.46 ± 0.04 for all samples; Table 4 of EA1 )(Fig. 8c). Orthopyroxene in the Udachnaya peridotites has similar to higher water contents than clinopyroxene. This is in contrast with peridotites worldwide, in which clinopyroxene contains about twice as much water as orthopyroxene (Bell and Rossman, 1992b; Grant et al., 2007a; Peslier et al., 2002; Peslier et al., 2012; Xia et al., 2010; Yang et al., 2008).

The Udachnaya water content ratio between orthopyroxene and garnet (average DH2Oopx/gar Udachnaya of 28 ± 4) are higher than those calculated from experimental data (DH2Oopx/gar exp. = 1.74 ± 0.19, EA1 Table 4) (Fig. 8d). Moreover the Udachnaya water ratio between olivine and garnet (average DH2Ool/gar Udachnaya of 25 ± 8) is 135 times higher than that calculated from experimental data (DH2Ool/gar exp. =0.18 ± 0.08, EA1 table 4)(Fig. 8b) and 10 times higher than that measured in Kaapvaal megacrysts (4-15 (Bell et al., 2004)).

The apparent overall disequilibrium for water between Udachnaya peridotite minerals is due to either (i) inadequate experimentally-determined water partition coefficients between melt and olivine or garnet, or (ii) to metasomatic processes affecting each mineral differently as discussed below.

* + 1. *Water contents of equilibrium melts*

Calculated water contents of melts in equilibrium with olivine range from 2.3 to 51 wt% H2O (Table 3). Five samples with olivine water contents > 200 ppm yield calculated water contents of melts in equilibrium with olivine > 30%. These unrealistic melt water contents are generated by using the very low partition coefficient between olivine and melt determined by Tenner et al. (2009) (i.e. DH2Ool/melt ~0.0006) estimated for olivine-melt equilibrium at 5 GPa. The recent experimental study of Sokol et al. (2013c) on CO2-rich Udachnaya kimberlite (8.5 to 9.4% CO2) shows that olivine-melt partition coefficients could be 5 times higher at P ~7 GPa, up to DH2Ool/melt ~0.003. Ten out 15 garnet peridotites in this study are equilibrated at P ≥ 5 GPa and seven are equilibrated at 6 to 7 GPa. The effective DH2Ool/melt must be between the two estimates. The peridotite xenoliths equilibrated at P > 5 GPa have P-T conditions close to those in the experimental work of Sokol et al. (2013c) and DH2Ool/melt ~0.003 appears more adequate to calculate the water content of melt in equilibrium with olivines of those peridotites. Using the Sokol et al. (2013c) olivine /melt partition coefficient, the water content of melt in equilibrium with peridotites equilibrated at > 5 GPa ranges from 7 to 11% H2O (Table 3). These values appear more realistic and are in the range of volatile contents estimated for kimberlites (> 7% water; (Sparks et al., 2006) and carbonates (6 to 10%,(Sokol et al., 2013b). However, these values are only accurate if abundant CO2 is present in the melt in equilibrium with olivine (Sokol et al., 2013a). Sokol et al. (2013a) show that DH2Ool/melt is strongly affected by CO2. For olivine equilibrated with CO2-poor silicate-melt, DH2Ool/melt is two times higher than for olivine in equilibrium with carbonate-silicate melt. In the light of the latter study and assuming mineral-melt equilibrium, the water content of melt in equilibrium with Udachnaya peridotites equilibrated at > 5 GPa ranges from 3.5 to 5.5 % H2O. Using the Withers et al. (2012) calibration of FTIR data to estimate water content in olivine, the melt in equilibrium with the olivine contains from 0.4 to 27% H2O using Tenner et al. (2009) partition coefficients and from 0.1 to 5.4 % using those of Sokol et al. (2013c) (EA1 Table 4).

To sum up, melts in equilibrium with Udachnaya peridotite minerals (Table 3) contain 0.3-6 wt% H2O (6.0 ± 4.0 for a melt in equilibrium with the olivine, 2.3 ± 0.4 with orthopyroxene, 2.0 ± 0.2 with clinopyroxene, and 0.29 ± 0.04 with garnet). The water content of a melt in equilibrium with the calculated bulk peridotite (using modal proportions of minerals, Table 1) is 3.3 ± 0.4 wt% H2O (1.3 ± 0.5 wt% using the Wither et al. (2012) calibration). These estimates are mostly higher than those for parental magmas of mid-ocean ridge basalts (MORB) (0.05 to 1.4%) (Danyushevsky et al., 2000; Dixon et al., 1988; Johnson et al., 1994; Michael, 1995; Michael, 1988) and ocean island basalts (OIB) (0.6 to 2%)(Dixon et al., 1997; Dixon and Clague, 2001). The highest estimates for water in melts in equilibrium for Udachnaya peridotites are similar to those measured in some melt inclusions in olivine from subduction arc magmas (up to 6-8 wt% H2O) (Grove et al., 2002; Roggensack, 2001; Sisson and Layne, 1993) indicating that such melts can be considered as good candidates for the metasomatic agent, however such high values in the melt inclusions may result from fractional crystallization processes (Sobolev and Chaussidon, 1996). Melt inclusions in Mg-rich minerals (olivine, pyroxenes and spinel) from subduction arc magmas provide the best estimates of water contents in the least-modified parental magmas. These contain <1 to 3 wt% H2O (Dobson et al., 1995; Kamenetsky et al., 2002; Sobolev and Chaussidon, 1996), i.e. are similar to values estimated for melts in equilibrium with Udachnaya peridotites. The latter are also commonly lower than the estimated water contents of kimberlite magmas before eruption, i.e. ~5 wt% (Kavanagh and Sparks, 2009). Moreover the parent magmas of Kaapvaal megacryst suites, thought to represent melts crystallized at mantle depths, have the same water contents within uncertainty (2.8 ± 1 wt%) (Bell and Moore, 2004; Bell et al., 2004) as the melts in equilibrium with our Siberian peridotites.

* 1. **Metasomatic control of water contents in Udachnaya olivine and garnet**

Water contents in olivine and garnet from the Siberian cratonic mantle have been reported before in two papers with FTIR analyses of peridotite xenoliths from several kimberlite pipes, but with very limited petrographic and chemical data (Matsyuk and Langer, 2004; Matsyuk et al., 1998). Other work addressed water in olivine xenocrysts and in clinopyroxene from eclogite and granulite xenoliths in the Siberian craton, but with the interpretation focused on crystallography (Khisina and Wirth, 2010; Koch-Müller et al., 2006; Koch-Muller et al., 2004). In the present section, we seek to link water contents with the petrologic history of our peridotites inferred from comprehensive petrographic and chemical data.

* + 1. *Partial melting and water contents*

Although the tectonic settings responsible for the formation of cratons are still debated, it is generally accepted that the cratonic mantle lithosphere under ancient continental cores experienced partial melting with at least 30% melt extraction (Bernstein et al., 1998; Boyd, 1989; Boyd et al., 1997; Ionov et al., 2010; Kopylova et al., 1999; Lee, 2006; Pearson and Wittig, 2008; Simon et al., 2007). These melting events occurred during the Archean or early in the Proterozoic (Carlson et al., 1999; Pearson et al., 1995b; Wiggers de Vries et al., 2013; Wittig et al., 2010), with a range of post-melting enrichments by infiltrating metasomatic agents (Boyd et al., 1997; Ionov et al., 2010; Kopylova et al., 1999; Lee, 2006; Pearson and Wittig, 2008; Simon et al., 2007). Water partitions into the melt during partial melting leaving behind water-depleted residues (Aubaud et al., 2004; Aubaud et al., 2008; Grant et al., 2006; Grant et al., 2007b; Hauri et al., 2006; Tenner et al., 2009)*.*

The water contents of olivine and garnet of Udachnaya peridotites show co-variation trends with Mg # in olivine (Figs. 3a and 6a). The Mg#Ol in melting residues increases with increasing degrees of melt extraction (Boyd, 1989). The co-variation between Mg# in olivine and water contents of minerals could imply that water in Udachnaya peridotite is controlled by melting processes. However, Mg# below 0.91 in both olivine and whole-rock in cratonic peridotites can also be the result of post-melting mantle-melt interaction (Boyd, 1998; Kelemen et al., 1998; Lee and Rudnick, 1999). Moreover, there is no correlation between water in minerals of Udachnaya peridotites and Al2O3 in the whole-rock (EA1 Table 2), considered as a robust index of melting (Ionov et al., 2010). In addition, some Udachnaya peridotites contain several percent of clinopyroxene (up to 6 % in deformed xenoliths; Table 1) whereas cpx is exhausted in the residues after 20-25% of melting (Herzberg, 2004; Walter, 1998); this is why clinopyroxene in cratonic peridotites is commonly viewed as having a late-stage origin (Boyd et al., 1997; Simon et al., 2003).

Seven out of ten Udachnaya peridotites in this study can be interpreted as minimally metasomatized, near-pristine melting residues from modal and chemical data (Table 1). They have water contents in olivine of 6.5 to 47 ppm H2O, i.e. an order of magnitude lower than all other samples (Table 2). As a first approximation, these relatively low water contents may have originated from melting processes with minimal metasomatic disturbance.

To investigate the possible link between water distribution in cratonic lithosphere and melting processes we modeled the water content of a melting residue assuming that water behaves as an incompatible component {Shaw, 1970 #195}{Johnson, 1990 #647}{Hellebrand, 2002 #2212}. Detail of melting models are given in Electronic Annex 3.

The water content estimated for the primitive mantle (1100 ppm H2O) is first used as initial water content (Bell and Rossman, 1992b; Palme and O'Neill, 2003). Using the fractional melting equation, the residue becomes “dry” (< 0.1 ppm H2O) after 3 to 5% melt extraction at 3 GPa, and after 5 to 6 % at 5 GPa, all minerals have < 0.1 ppm H2O (Aubaud et al., 2004; Aubaud et al., 2008; Dixon et al., 2004; Grant et al., 2006; Grant et al., 2007b; Hirth et al., 2000; Pollack, 1986)*.* After 25 % of batch melting the residue still contains between 10 to 15 ppm H2O in the bulk-rock, 1 to 15 ppm in olivine, 20 to 45 ppm in orthopyroxene, 30 to 45 ppm in clinopyroxene and 2 to 8 ppm in garnet. Using a water-depleted source, for example the source region of mid-ocean ridge basalts (MORB), i.e. an initial water content of 180 ppm H2O (Danyushevsky et al., 2000; Dixon et al., 1988; Michael, 1995; Michael, 1988; Sobolev and Chaussidon, 1996), the residue contains 2 to 3 ppm H2O in the bulk rock, <0.2 ppm in olivine, 3 to 7 ppm in pyroxene and <1.3 ppm in garnet after 25 % of batch melting, which is lower than the degree of melting inferred for most Udachnaya peridotites (EA2 Fig. 1a). These values are lower than the bulk-rock and mineral water contents found in Udachnaya peridotites. Even if the partition coefficient ofSokol et al.(2013c) between olivine and melt is applied (DH2Ool/melt ~0.003, which is 5 times higher than the partition coefficient of Tenner et al. (2009), DH2Ool/melt ~0.0006), the residue becomes “dry” after 6 to 7% of fractional melting, with water contents in minerals <0.1 ppm H2O.The majority of peridotite xenoliths in kimberlites, including the Udachnaya peridotite in this study, contain at least several ppm H2O (Peslier, 2010) even though most are residual in terms of modal and major element compositions. Most water found in residual mantle peridotites must have been added after melt extraction because their measured water contents are too high compared to those predicted by simple melting models. The cratonic peridotites are affected by post-melting metasomatism, which modified their initial trace element signatures and may also affect major oxides, mineralogy and microstructures (Doucet et al., 2013; Kelemen et al., 1998; Rudnick et al., 1993; Simon et al., 2003). Metasomatism can erase the record of partial melting and may control the water distribution in cratonic mantle (Peslier et al., 2012).

* + 1. *Metasomatism and water contents*

As discussed above, the water content of Udachnaya peridotites is likely controlled by metasomatism rather than melting processes. A key observation is that the highest measured water contents in olivine (> 200 ppm) and garnet (> 10 ppm) are only observed in the xenoliths that are relatively rich in clinopyroxene (≥ 4 %) and garnet (≥ 7 %) and that have low Mg#Ol (Fig. 3b and Fig. 6c-d). Melting experiments relevant to the formation of Mg-rich (Mg# >0.92) cratonic peridotites show that cpx in the residues may be exhausted after 20–25% of melting; only small amounts of garnet (3%) can survive high-degree melting at high pressures (4-3 GPa) (Herzberg, 2004; Walter, 1998; Wittig et al., 2008). Doucet et al. (2013) argued that up to 5% of modal garnet in Udachnaya cratonic peridotite may be present in residues of high degrees of melting at > 5 GPa after they cool down to the ambient lithospheric temperatures. Only small cpx amounts (<3%) can be unmixed from high-T, (Al,Cr)-rich residual opx on cooling or produced by reaction between garnet and olivine (ol + gar 🡪 spl + opx + cpx) if a pressure drop causes garnet- to spinel-facies transition. The water content in olivine and in garnet in our samples increases with modal clinopyroxene > 3 % (Fig. 3b and Fig. 6d), and the water content in garnet increases as well for samples with modal garnet > 5% (Fig. 6c). This is interpreted here as evidence for a link between water contents in peridotite and modal metasomatism (re-fertilization), i.e. precipitation of clinopyroxene and garnet during percolation of water-bearing melts/fluids through melting residues.

Doucet et al. (2013) showed that modal metasomatism in Udachnaya peridotites produced enrichments in Fe, Ti, Al, Ca, HREE-MREE commonly accompanied by a decrease of Mg# in bulk-rocks and olivine. The water contents in olivine are low (6.5 to 42 ppm) for all but two samples with Mg#Ol > 0.92 while peridotites with Mg#Ol < 0.92 have generally higher water contents in olivine (up to 323 ppm H2O; Fig. 3a). The Mg-rich xenoliths U29 and Uv-419/09 (Mg#Ol > 0.92) have high water contents in olivine (229 and 213 ppm; Table 2), but these samples have high modal clinopyroxene ( >3%; Table 1), which indicates modal metasomatism. Peridotites with Mg#Ol > 0.92 have garnet water contents ranging from 0 to 10 ppm while peridotites with Mg#Ol < 0.92 commonly have higher garnet water contents ranging from 3.6 to 23 ppm, with a positive co-variation of Mg#Ol > 0.92 with water in garnet (R² = 0.6; Fig. 6a). The water contents in garnet also increase with higher bulk-rock and garnet TiO2 (Fig. 6b and e), LREE-MREE (Eu shown in Fig. 6f). The water contents in clinopyroxene, available only for three peridotites with a range of modal compositions (see section below), positively correlate with the Ce/Yb ratios in clinopyroxene, which could indicate an increase of MREE-HREE relative to HREE by metasomatism (Fig. 3d). The highest water contents in olivine, clinopyroxene and garnet are also found in samples with trace element patterns indicative of metasomatic enrichments such as garnets with sinusoidal or MREE-enriched patterns and high LREE/HREE ratios in clinopyroxene (e.g. U64, U501, U267 (Fig. 7) (Doucet et al., 2013; Doucet et al., 2012). The lower water contents are found in the samples with LREE-depleted garnet and with the lowest HREE in clinopyroxene (U506 in Fig.7) indicative of minimal metasomatic inputs. The high FeO, TiO2 and LREE-MREE are typically linked to metasomatic agents (Simon et al., 2007; Wittig et al., 2008), which appear to have also brought in water and crystallized water-rich clinopyroxene and garnet.

The REE-rich clinopyroxene and garnet with sinusoidal to humped REE patterns in modally metasomatized (“re-fertilized”) peridotites are interpreted to result from interaction with silicate- or carbonate-rich melt and liquids (Shimizu, 1999; Shimizu et al., 1997; Simon et al., 2003). Because these peridotites are also enriched in FeO (≥ 7%; Mg# ≤ 0.91) and have Al2O3 ≥ 1%, SiO2 > 43% (EA1 Table 2), they were likely infiltrated by silicate melts rather than fluids. The latter do not transport these elements in concentrations high enough to precipitate silicate minerals (Dvir et al., 2011; Hermann et al., 2006; Mysen and Shang, 2003), like those observed in Udachnaya peridotites in the form of late-stage clinopyroxene and garnet (Doucet et al., 2013). Moreover, orthopyroxene water contents in coarse garnet peridotites appear to correlate positively with modal orthopyroxene (R² = 0.7, Fig. 3c) if we exclude U1147, for which no water was detected in garnet. Given that orthopyroxene addition can be the result of mantle-melt interaction (Kelemen et al., 1998), this observation is consistent with enrichments of the peridotites in water by silicate melts, rather then fluids. Finally, no correlations are observed between water contents of Udachnaya peridotite minerals and oxygen fugacity (Fig. 5), indicating that the water-bearing metasomatic melts were not more oxidized than the cratonic peridotites they were infiltrating.

* + 1. *Origin of metasomatism*

Goncharov et al. (2012)argued that the broad range of T at similar P for peridotites equilibrated at 4-5.5 GPa and the vague trend from near 40 mW/m² to 45 mW/m² for peridotites equilibrated at P > 6 GPa (Fig.1a) may reflect thermal perturbation of an initial “cold” lithosphere through a combination of magmatic under-plating and intrusion of melts/fluids in the lower lithosphere accompanied by shearing*.* It follows that the Fe-, Al-, Si-, REE- and water-rich melts responsible for the metasomatism of the lower part of the cratonic mantle in central Siberia probably derived from the underlying asthenosphere. The enrichments in TiO2 > 0.1% and FeO ≥ 7% in metasomatized Udachnaya peridotites may indicate links with melts from mantle plumes rather than with depleted convecting mantle sources (Agashev et al., 2013; Doucet et al., 2013; Doucet et al., 2012; Ionov et al., 2010).

The H2O/Ce ratio in melts has been used to constrain tectonic settings of their sources (Michael, 1995; Plank et al., 2009). Because of the similar partition coefficients for H and Ce during melting and crystallisation, their ratio is assumed to be independent of these processes. Mid-ocean ridge basalts typically have H2O/Ce ratios ranging from 155 to 280 (Michael, 1995), the OIB have a wider range from 100 to 330 (Dixon and Clague, 2001), and arc lava melt inclusions record from 200 to up 20000 (Plank et al., 2009). To estimate the H2O/Ce ratios of the melt in equilibrium with Udachnaya peridotites we used the bulk-rock water contents of the three Udachnaya peridotites for which all phases have been analyzed and calculated the water content of the melt in equilibrium with those (section 6.2.2). To contraint the Ce compositions of the melt responsible for water enrichment in Udachnaya peridotite minerals we calculate the Ce content of a hypothetical melt in equilibrium with clinopyroxene for the 3 samples for which we have the whole-rock water contents. Clinopyroxenes from these samples have REE patterns interpreted to result from metasomatism. We used the trace element partition coefficient between clinopyroxene and melt Dcpx/meltCe = 0.098 calculated by McKenzie and O’Nions (1991) and currently used in the pMELT software (Ghiorso et al., 2002; Smith and Asimow, 2005), which is in the range for Dcpx/meltCe determined experimentally (Hart and Dunn, 1993; Hauri et al., 1994; Jenner et al., 1993; Johnson, 1998) and was determined for the peridotite system.

The calculated melts in equilibrium with Udachnaya peridotites have H2O/Ce ratios ranging from 100 to 400 (Fig. 9). Using bulk peridotite water contents calculated with the calibration of Withers et al. (2012), the H2O/Ce of the equilibrium melt range from 40 to 180 (EA1 Table 4). These ranges are smaller than that inferred for Kaapvaal peridotites (Peslier et al., 2012) and are closer to the OIB range and lower than the H2O/Ce in arc melts. This is generally consistent with the inference of Goncharov et al. (2012) that the metasomatic sources beneath the Siberian craton were in the asthenosphere, e.g. in a mantle plume environment (Howarth et al., 2013).

Asthenospheric upwellings underneath cratonic lithosphere are in particular believed to be responsible for kimberlite magmatism, producing localized deformation and metasomatism (Agashev et al., 2013; Becker and Le Roex, 2006; Ionov et al., 2010). Recent experimental work suggests that kimberlite magmas are derived from lithospheric domains metasomatized by carbonatic melt (Sokol et al., 2013b; Sokol et al., 2013c). The hypothesis of Udachnaya peridotite having been metasomatized by parental kimberlite melts is supported by REE estimates of melts in equilibrium with clinopyroxene and garnet from Udachnaya peridotites, which are in the range of those typical of the Udachnaya kimberlite (Doucet et al., 2013). This argues for interaction of the cratonic mante roots with asthenosphere-derived parental magmas of kimberlite.

We emphasize that water addition to Udachnaya peridotite minerals took place in the mantle prior to kimberlite magmatism and eruption. No water addition from the host kimberlite occurred during xenolith transport to the surface as shown in section 6.1.1. The water enrichment linked to modal metasomatism that affected Udachnaya garnet peridotites must have taken place in the mantle before the ascent and eruption of kimberlite magmas. Trace elements of melts in equilibrium with clinopyroxene and garnet must result from the last metasomatic event, i.e. circulation of parental magmas of kimberlite in the mantle, prior to the ascent and eruption of the evolved end-members of their differentiation.

* 1. **Comparison of water distribution between Siberian and Kaapvaal cratons**

The range of olivine water contents in this study is similar to those reported by (Matsyuk and Langer, 2004) for Udachnaya peridotite xenoliths and by (Koch-Müller et al., 2006) for olivine megacrysts from the Udachnaya kimberlite. However, this range extends to values twice as high as the highest olivine water content reported for xenoliths from the Kaapvaal, the only other craton which has not lost its mantle root and for which water data are available (Fig 4a)(Baptiste et al., 2012; Peslier et al., 2010). The distribution of water contents of olivine along a depth profile beneath Udachnaya is also distinct from that reported for the Kaapvaal craton (Fig 4a). In particular, the minerals from the base of the lithosphere (equilibrated at 6-7 GPa) show broad ranges of water contents of (26 to 323 ppm H2O in olivine, 103 to 301 ppm in orthopyroxene, 100 to 272 ppm in clinopyroxene and 3.6 to 23 ppm in garnet) and appear to have similar or even higher average concentrations than those from the shallow and middle lithospheric levels (11 to 229 ppm H2O in olivine, 28 to 202 ppm in orthopyroxene and 0 to 11 ppm in garnet) (Fig. 4). This is in contrast to what has been reported for the Kaapvaal craton where the deepest peridotites (6-7 GPa) record low olivine water contents (1 to 40 ppm H2O) (Fig. 4) (Baptiste et al., 2012; Peslier et al., 2010). In the Udachnaya peridotites, the highest water contents in olivine, orthopyroxene (≥ 300 ppm H2O) and garnets (> 15 ppm H2O) are found at the bottom of the lithospheric profile (6-7 GPa; Table 2, Fig. 4) and are higher than any measured in Kaapvaal peridotites. Another major difference is that 2/3 of the garnets measured in Udachnaya peridotites have water contents ≥ 5 ppm H2O whereas 1/2 of the garnets from the Kaapvaal craton have water contents < 5 ppm H2O or below detection limit (Fig. 4c) (Baptiste et al., 2012; Bell and Rossman, 1992a; Peslier, 2010).

Peslier et al. (2010) proposed that infiltration of asthenospheric melts and/or fluids were responsible for the low water content of olivines at the base of the Kaapvaal cratonic lithosphere and could have been water-poor compared to the craton peridotites they infiltrated (Fig. 10b). Such melts and/or fluids may have scavenged water from the deepest mantle section and re-deposited it in peridotites at lower P (< 5 GPa). Therefore, it is the “dry” metasomatism, rather than partial melting, that may be ultimately responsible for the low water in the deepest part of the Kaapvaal cratonic root.

At shallower levels of the Kaapvaal cratonic lithosphere, however, various water-bearing metasomatic agents enriched the Kaapvaal craton in water(Peslier et al., 2012), possibly those of subduction-related origin. It is genelly accepted that subduction zones played a role in craton formation (Canil, 2004; Carlson, 2005; Grove and Parman, 2004; Lee and Walker, 2006; Pearson and Wittig, 2008; Simon et al., 2007). In particular the silica (orthopyroxene) enrichment in Kaapvaal peridotite xenoliths (Boyd, 1998; Simon et al., 2007) may be the result of reaction of melting residues with silica-rich fluids and/or liquids expelled from sinking slabs (Kelemen et al., 1998; Kesson and Ringwood, 1989) as suggested by the high H2O/Ce (100 to 4000, Fig. 9) in Kaapvaal peridotites (Peslier et al., 2012). In contrast with the Kaapvaal craton, Si-rich peridotites are rare in the Siberian cratonic mantle (Ionov et al., 2010) and H2O/Ce of Udachnaya peridotite is similar to the range for OIB and lower to melt produced in subduction zones. The Kaapvaal lithosphere might have been more affected by subduction metasomatic agents than the Siberian craton.

The abundance of deformed peridotites equilibrated at P > 4-5 GPa in Udachnaya (Sobolev, 1977) could be an artifact of sampling by kimberlites, that may rise to the surface mainly through shear zones. The addition of even small amounts of water-bearing melts results in strong weakening of mantle rocks (Zimmerman and Kohlstedt, 2004) and may facilitate their deformation as shown by studies on deformation distribution in peridotite massifs (Skemer et al., 2013; Vauchez et al., 2012). Moreover, intrusion of water-rich melts may be more likely through the mechanically weak shear zones (Vauchez et al 2012).

The main difference between Siberian and Kaapvaal cratons in terms of water is that water addition by metasomatic agents occurred at pressures > 4 GPa and all the way to the base of the cratonic root below central Siberia, while this phenomenon is limited to shallower levels (< 5 GPa) beneath the Kaapvaal. The origin of the difference in water distribution at the base of the Kaapvaal and Siberian cratons is not clear. The base of the Kaapvaal craton was reported to be more reduced than in the central Siberian craton by about 1 log unit (Goncharov et al. (2012) (Fig. 1b), which may result in fluids of somewhat different compositions at these depths (Ballhaus and Ronald Frost, 1994; Goncharov et al., 2012; Holloway and Blank, 1994; Taylor and Green, 1988; Woodland and Koch, 2003).

The C-O-H spectiation, i.e. the relative proportion of H2O, CO2, CH4, CO and H2 of fluids in equilibrium with peridotites is estimated using least square method on the state equation of Belonoshko and Saxena (1992) for fixed P,T and *f*O2 (Frost and McCammon (2008) and assuming ideal mixing between gas phases. The C-O-H speciation of fluids using previous published *f*O2 for Kaapvaal and Siberian peridotites (Goncharov et al., 2012; Peslier et al., 2010) is calculated using the *f*O2 calculated with the new calibration of Stagno et al. (2013) based on the following equation:

The calibration of Stagno et al. (2013) results in *f*O2 estimates down to -3.15 Δlog *f*O2 (FMQ) for the Siberian mantle and down to -3.50 Δlog *f*O2 for the Kaapvaal mantle (Fig. 1b, EA1 Table 5 and 6). These values are higher than *f*O2 estimated using the calibration of Gudmundsson and Wood (1995) which give *f*O2 down to-3.58 Δlog *f*O2 for the Siberian mantle and -4.64 Δlog *f*O2 for the Kaapvaal mantle (EA2 Fig. 9a). The new calibration of Stagno et al. (2013) reproduced *f*O2 values from experimental work for which the oxygen fugacity is controlled and appears more robust than other calibrations for high-pressure peridotites. It follows that the mole fraction of H2O calculated for fluids in equilibrium whith peridotites is higher when using the calibration of Stagno et al. (2013) (EA1 Fig. 9b-c), with average H2O proportion of 0.90 for the Siberian mantle and 0.83 for the Kaapvaal mantle (Fig. 11).

The C-O-H estimates show that the proportion of H2O in fluids, expressed as mole fraction, beneath the Siberian craton range fom 0.73 to 0.98 (averag ~0.90), which is similar to or higher than for water in fluids beneath the Kaapvaal craton, which range from 0.76 to 0.95 (average ~0.83,) except for two samples equilibrated at between 4 and 5 GPa, with H2O proportion down to 0.67 to 0.53 (Fig. 11a). All fluids are very low in CO2 in the lithospheric mantle beneath both cratons (Fig. 11b-c), but fluid in the Kaapvaal mantle appears generally richer in CH4 (0.1 to 0.44) compared to fluids in the Siberian mantle which are CH4-poor (0 to 0.14) (Fig. 11b-c ; EA1 Table 5 and 6). The higher CH4 content in fluids beneath the Kaapvaal at P > 5.5 GPa may have resulted in lower water activity, and in turn, lower water contents in the olivines in equilibrium with these fluids in the Kaapvaal cratonic base compared to that of the Siberian craton. The origin of the more reduced nature of the fluids infiltrating the deepest levels of Kaapvaal cratonic lithosphere compared to that for Siberia is unknown, but it can be speculated that it may be linked to the depth of the lithosphere-asthenophere boundary (potentially deeper beneath Siberia) (Artemieva and Mooney, 2001; Priestley and McKenzie, 2006) or to asthenospheric upwelling from the lower mantle beneath Africa (super-plume (Ritsema et al., 1999)) which is not present beneath Siberia.

The common feature between Siberian and Kaapvaal cratons in terms of water is that in both cases, metasomatism controls the water content of the peridotites, and that the style of metasomatism (melt or fluid composition) is different in mantle regions sampled by individual kimberlite pipe.

* 1. **The role of water in the stability of cratonic lithospheric mantle**

The long-term stability of cratons is believed to be due to the presence of a thick (~200 km) layer of rigid residual peridotites underneath Archean-Proterozoic crust (Boyd, 1989; Jordan, 1975). Multiple ancient depletion events produced peridotites low in iron (Mg#>0.92) and incompatible trace elements, including radioactive isotopes. This enhances the buoyancy and the viscosity of cratonic mantle by decreasing its density and heat production (Jordan, 1975; Pollack and Chapman, 1977) compared to the convecting asthenospheric mantle. Cratonic peridotites record melting events as old as the crust above indicating that they may have been produced simultaneously (Carlson, 2005; Pearson et al., 1995a; Pearson et al., 1995b; Pearson et al., 1995c; Richardson and Harris, 1997; Walker et al., 1989). As discussed above, residual cratonic mantle is generally thought to be anhydrous (Aubaud et al., 2004; Aubaud et al., 2008; Dixon et al., 2004; Grant et al., 2006; Grant et al., 2007b; Hirth et al., 2000; Karato, 2010; Pollack, 1986) even though it can be locally rehydrated by metasomatism, as illustrated by our data on Udachnaya xenoliths. Finally, (Peslier et al., 2010) proposed, based on observations on Kaapvaal craton xenoliths, that olivine may be “dry” (< 1 ppm H2O) near the lithosphere-asthenosphere boundary (~7 GPa), i.e. that the dehydrated bottom layer may be responsible for the longevity of cratons by increasing the viscosity of the base of cratonic roots by several orders of magnitude compared to the underlying convective mantle.

The mechanical behavior of olivine is likely to control the rheology of cratonic mantle because it is by far the most abundant mineral (52-87% of the peridotites in this study, Table 1) and is less strong than orthopyroxene (Yamamoto et al., 2008). Although clinopyroxene and garnet can become weaker than olivine when they contain water, they are minor phases in peridotites (6.8 ± 4.3 and 4.5 ± 4.0 % respectively in Udachnaya peridotites), and garnet strength increases with increasing pressure at > 5 GPa (Chen et al., 2006; Hier-Majumder et al., 2005; Katayama and Karato, 2008; Mei et al., 2010; Xu et al., 2013). An olivine aggregate is consequently used as a proxy for the whole mantle in our calculation of mantle viscosity following the method of Li et al (2008) and Peslier et al. (2010) (details in Ellectronic Annex 3). Figure 12 shows the viscosity calculated for each Udachnaya and Kaapvaal peridotite sample using their measured olivine water content and plotted at their pressure of equilibration. A range of viscosity estimates for the asthenosphere is also shown for comparison (Fleming et al., 2007; Larsen et al., 2005; Pollitz et al., 1998; Sjöberg et al., 2000). The viscosity of Udachnaya peridotites ranges from 8.4 x 1016 to 1.9 x 1028 Pa.s-1 for *C*OH estimates using the calibration of Bell et al. (2003) (Fig. 12a) and from 4.0 x 1017 to 4.7 x 1028 Pa.s-1 for *C*OH estimates using the calibration of Withers et al. (2012) (Fig. 12b). Because the calibration of Withers et al. (2012) gives lower water contents in olivine, the viscosity estimates are higher than viscosity estimates using the calibration of Bell et al. (2003) (Fig. 12). Nevertheless, both viscosity estimates remain in the same order of magnitude, and the real viscosity may lie between the two estimates. The viscosity estimates obtained with the calibration of Bell et al. (2003) for olivine water contents are used in Fig. 14a to facilitate the comparison with data from the literature.

The lithospheric mantle down to ~180 km (P < 5.4 GPa) beneath Udachnaya is made up of peridotites with ηeff of 3.4 x 1021 to 1.9 x 1028 Pa.s-1, which defines a thick and rigid layer of cratonic mantle because these viscosities are a thousand times higher than the average viscosity of the asthenosphere, ηeff ≤3.4x1018 Pa.s-1 (Fig. 11). The high viscosity is primarily a function of the low equilibration temperatures of these samples, which plot near the 35-40 mW/m² conductive geotherm (Fig. 1). This buffers the viscosity of the peridotites despite their broad range of olivine water contents (11 to 229 ppm).

The viscosity of Udachnaya peridotites equilibrated at ≥190 km (P > 6 GPa) ranges from ηeff of 8.4 × 1016 to 8.0 × 1018. This range is slightly lower than or equal to that of the asthenosphere (Fleming et al., 2007; Larsen et al., 2005; Pollitz et al., 1998; Sjöberg et al., 2000)) (Fig. 12) due to the combination of high temperatures and, for some, high olivine water contents (> 200 ppm H2O) of these peridotites. If these low ηeff were representative of the whole base of the Siberian cratonic root, the latter would not have been able to resist delamination by the convecting asthenosphere for a long time (King, 2005). However, the lithospheric thickness of 210-220 km estimated at 360 Ma, which is the eruption age of Udachnaya kimberlites, from our data and previous work (Boyd et al., 1997), is the same as that inferred from geophysical data for the present central Siberian craton (Priestley and Debayle, 2003). The central craton still has a root extending to 200-250 km depth. Hence, our deepest Udachnaya xenoliths with their asthenospheric viscosities may not be representative of the entire central Siberian craton and instead come from a limited domain, otherwise the Siberian cratonic root would have been delaminated (King 2005).

In summary, our calculations show that the effects of water enrichments on peridotite viscosity are greater for xenoliths equilibrated at high pressure (i.e. > 6 GPa), because of the higher temperature at these depths than at shallower levels (conductive geotherm > 40 mW/m²; (Goncharov et al., 2012)). The coincidence of localized water enrichments, deformation and thermal perturbations, especially at the bottom of the lithosphere, may not be fortuitous but rather a result of a specific process (Goncharov et al., 2012). We speculate that the deepest Udachnaya samples may be more metasomatized than common rocks at the base of the Siberian cratonic root away from kimberlite fields. This may have been detected beneath the Baltic shield which shows lateral variations of seismic velocities in the cratonic lithosphere (Pedersen et al 2013). Some cratonic domains have homogeneous seismic velocities over the entire lithospheric profiles (down to 250 km), whereas others are characterized by a seismic velocity decrease at 150 km depth which is interpreted as reflecting variable degrees of refertilisation of cratonic roots (Pedersen et al. (2013). The Udachnaya xenoliths may represent peridotites variously hydrated due to their proximity to melt conduits in the lithosphere as suggested in section 6.3.4 (Fig. 10a). The Siberian cratonic root is still probably buoyant and strong due to the overwhelmingly refractory compositions and moderate water contents of such peridotites, which are under-represented in the xenolith populations carried by kimberlite magmas.

Alternatively, water may not be an important factor in the deformation properties of olivine (Fei et al., 2013) and hence the strength of the cratonic lithosphere is not related to water contents. In such a case, the Udachnaya xenoliths and their water contents could be representative of the base of the cratonic root in central Siberia, which could then have remained strong despite elevated water contents.

1. **CONCLUSIONS**

This is the first study of water contents in minerals from well-characterized peridotite xenoliths from the Siberian craton. We find no evidence that water in the minerals is related to interaction with the host kimberlite magmas. Water loss from minerals by diffusion during ascent of xenoliths in the kimberlite magma is very limited and diffusion modeling indicates that olivine cores preserved their mantle water contents. The Udachnaya peridotites show a broad range of water contents in olivine from 6.5 ± 1.1 to 323 ± 65 ppm H2O, much higher than those from similar peridotite xenoliths from the Kaapvaal craton for which olivine water contents range from <1 to 150±10 ppm H2O. The water contents of Udachnaya peridotite minerals were modified after the partial melting events by migrating water-rich melts, also responsible for enrichments in Fe, Ti and incompatible trace elements and the crystallization of late-stage garnet and clinopyroxene. Based on the present study of Udachnaya mantle xenoliths, the largest part of the lithospheric mantle section in the central Siberian craton down to ~180 km is primarily composed of coarse, refractory and relatively cold peridotites with low to moderate olivine water contents of 6±1 to 92±16 ppm. This volumetrically dominant part of the mantle profile may be responsible for the general buoyancy and strength of the entire lithospheric mantle in the region. By comparison, the deepest peridotites equilibrated at 180 to 220 km commonly record deformation and metasomatism locally linked to water enrichments (up to 323±65 ppm H2O in olivine). The latter probably occurred near localized shear zones above the lithosphere-asthenosphere boundary in the laterally limited regions above asthenospheric upwellings.

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**Figure captions**

**Fig. 1.** (a) Equilibration temperature (T) versus pressure calculated for Udachnaya garnet peridotites. The thermometer (TBK90) and the barometer (PBK90) of Brey and Köhler (1990) are used to facilitate comparisons with previous studies on water contents of cratonic roots. Also shown are the mantle adiabat, the graphite/diamond transition boundary (Rudnick and Nyblade, 1999) and the 30, 35, 40 and 45 mW/m-2 conductive continental geotherms of Pollack and Chapman (1977). (b) Oxygen fugacity estimated using the calibration of Stagno et al. (2013) and reported here relative to the quartz-fayalite-magnetite buffer and is given in logarithmic units (∆FMQ). ∆FMQ values were calculated from Mössbauer analyses of garnet obtained by Goncharov et al. (2012) for the Siberian craton and by Lazarov et al. (2009); Woodland and Koch (2003) compiled by Peslier et al. (2010).

**Fig.2.** Absorbance area in cm-2 in olivines i.e. area integrated beneath OH bands, along the α and γ (a) and β and γ (b) optical indicatrix versus distance across minerals grains in m. The diffusion modeling of H loss uses equation (5) from Peslier et al. (2008): hydrogen diffusion models are based on 2-dimension diffusion equations for a sample of finite size surrounded by an infinite medium containing no H (Shewmon, 1983). The H diffusion coefficient (D) are calculated at each xenolith equilibration temperature (Demouchy and Mackwell, 2006). The solid curves show the best match to the data using the absorbance measured in the core of olivine as initial value. The dashed curves show the closest match to the data assuming an initial water content 20% higher than that measured in the olivine core. Note that the solid curves are the best fit to the data. Days, hours and minutes are the time estmated for the water d to diffuse in minerals.

**Fig. 3.** (a-b)Water contents of olivine as function of Mg# [Mg/(Mg+Fe)] at in olivine (a) and modal clinopyroxene (b). Cpx = clinopyroxene.(b-c)Water contents of orthopyroxene (ppm H2O) vs modal orthopyroxene (a) and water contents of clinopyroxene (ppm H2O) versus Ce/Yb ration in clinopyroxene. Opx = orthopyroxene; Cpx = clinopyroxene. The trend line in (c) is for coarse garnet peridotites (red circles) not taking into account U1147 for which no water was detected in garnet.

**Fig. 4.** Pressure as a function of water contents of olivine (a), orthopyroxene (b), garnet (c) and clinopyroxene (d) from Udachnaya xenoliths [see (c) of Fig. 1 and (b) of Fig. 4 for symbols]. Also shown are water contents of olivine, orthopyroxene, garnet and clinopyroxene from Kaapvaal xenoliths (white circles; (Baptiste et al., 2012; Peslier et al., 2010; Peslier et al., 2012)). Opx = Orthopyroxene; Cpx = clinopyroxene.

**Fig. 5.** Oxygen fugacity (∆log*fO2*(FMQ)) versus water contents (ppm H2O) of olivine (a) orthopyroxene (b), clinopyroxene (c) and garnet (d) for Udachnaya garnet peridotites in this study. The barometer (PBK90) of Brey and Köhler (1990) are used to facilitate comparisons with previous studies on water contents of cratonic roots. fO2 values are calculated using the new calibration of Stagno et al. (2013) using the iron speciation in spinel and garnet from Goncharov et al. (2013) for Udachnaya peridotites and from Lazarov et al. (2009); Woodland and Koch (2003) for the Kaapvaal cratonic peridotites, compiled by Peslier et al. (2010).

**Fig. 6.** Water contents of garnet (ppm H2O) versus Mg# [Mg/(Mg+Fe)] at in olivine (a), TiO2 in whole-rocks (b), modal garnet (c), modal clinopyroxene (d), TiO2 in garnet (e), primitive mantle-normalized (McDonough and Sun, 1995) Eu (f) contents of the whole-rock (WR) calculated from mineral analyses and modal proportions (WR EuN).

**Fig. 7.** Primitive mantle-normalized (McDonough and Sun, 1995) REE patterns of clinopyroxene (a and b) and garnet (c and d) for coarse garnet peridotites (red profiles) and deformed garnet peridotites (orange profiles for transitional and green profiles for sheared peridotites). Also reported are the water contents of olivine and garnet. The peridotites with higher water contents in olivine and garnet (e.g. U501 and U85) tend to have clinopyroxene more enriched in LREE and garnet with higher HREE abundances than peridotites with lower water contents in olivine and garnet (e.g. U280, U283, U506). The sheared peridotite (in green) has higher Cpx REE contents and higher water content in garnet than the coarse peridotites. The shapes of the garnet REE profiles from U506, through U64 to U267 are also indicative of increasing metasomatic input (see text for details).

**Fig. 8.** Assessment of equilibrium for water between minerals by comparing ratios of water contents measured in Udachnaya peridotite minerals (DH2O Udachnaya) with partition coefficients measured experimentally (DH2O exp.) (Tenner et al., 2009; O’Leary et al., 2010). For example DH2Ool/opx Udachnaya is the ratio of water contents in olivine and orthopyroxene from the same Udachnaya xenolith. DH2O exp. is derived from experimental data For example DH2Ool/opx is the ratio of experimentally determined DH2Ool/melt and DH2Oopx/melt. DH2Ool/melt and of DH2Ogarnet/melt are from Tenner et al. (2009), of DH2Oopx/melt and DH2Ocpx/melt are calculated from major element composition of pyroxenes and experimentally constrained equations of O’Leary et al. (2010). See text for more details. Error bars represent 2σ and were calculated using a propagation error formula. Most minerals seem out of equilibrium with other minerals for water in Udachnaya peridotites.

**Fig. 9.** Calculated H2O/Ce range of melts in equilibrium with Udachnaya peridotites for which all minerals were analyzed for water contents (black arrow) compared to the range of H2O/Ce ratios for MORB (Michael, 1995), OIB (oceanic island basalts from Hawaii) (Dixon and Clague, 2001), arc melts inclusions (Plank et al., 2009) and for hypothetical melts in equilibrium with Kaapvaal craton peridotites (Peslier et al., 2012). The range of H2O/Ce of Udachnaya peridotites is lower compared to that of Kaapvaal’s, but is similar to that of OIB.

**Fig. 10.** Sketches illustrating the distribution of water in cratonic roots. (a) Scenario proposed in this study, based on Siberian cratonic xenolith data: the mantle is mainly made of coarse peridotites with moderate water contents in olivine (≤50 ppm), the water enrichments are localized in shear zones and associated deformation and metasomatism by Fe-, Al-, Si and REE-rich melts; (b) scenario based on Kaapvaal cratonic xenolith data: the base of the cratonic root is dehydrated by infiltrating reduced asthenospheric melts, fluids or an immiscible phase which deposited the water at higher levels (Peslier, et al 2010). The upper part of the cratonic mantle is also hydrated by metasomatic agents of different nature in each region sampled by each kimberlite pipe, some possibly slab-derived (thick black lines representing ancient stacked slab into cratonic lithosphere) and/or precursors of kimberlite melts (Peslier et al 2012).

**Fig. 11.** (a)C-O-H speciation of fluids in equilibrium with Siberian cratonic peridotites calculated following the state equation of Belonoshko and Saxena (1992) for fixed P,T and fO2 calculated using the calibration of Stagno et al.(2013) (Circle, H2O mole fraction; square, CH4 mole fraction). Also shown the C-O-H speciation of fluids in equilibrium calculated for the Kaapvaal mantle from the iron speciation in garnet of Kaapvaal peridotites (Peslier et al., 2010). C-O-H speciation of fluids in equilibrium with cratonic peridotites estimated by Frost and McCammon (2008) for H2O (blue dashed line), CH4 (orange dashed line) and CO2 (grey dashed line). (b-c) C-O-H speciation of fluids in equilibrium with Siberian peridotites (b) and Kaapvaal peridotites (c) for each sample as function of equilibration pressure (blue bars, H2O; grey bars, CO2; orange bars, CH4, green bars, H2).

**Fig. 12.** Viscosities calculated for olivine aggregates in dislocation creep and measured water contents versus depth (in km and GPa). We show *η*eff, calculated with two olivine water content estimates, that from the calibration of Bell et al. (2003) (a) and that from the calibration of of Withers et al. (2012) (b) combined with the equilibration pressure and temperature of the Siberian xenoliths. The Kaapvaal xenoliths data from Peslier et al. (2010) are also shown. Estimate of the cratonic viscosities versus depth for constant water content of 10 to 300 ppm H2O are also pictured as lines. The cratonic geotherm was calculated assuming a heat flow of 40 mW.m-2 (grey field) and 35mW m-2 (black dashed lines ), following the equation T = 360.73 + 9.0612*P*-0.0206*P*2+(4x10-5)*P*3, with T in K and P in Km.The range of viscosities of the asthenosphere is from (Fleming et al., 2007; Larsen et al., 2005; Pollitz et al., 1998; Sjöberg et al., 2000).