

Diamond, subcalcic garnet, and mantle metasomatism: Kimberlite sampling patterns define the link

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ABSTRACT

A genetic relationship between diamond and subcalcic Cr-pyrope garnet, both being produced by a metasomatic process, can be inferred from the sampling patterns of kimberlites in the Daldyn-Alakit province, Yakutia, Russia. Pressure-temperature estimates for xenoliths and xenocrysts show a strong concentration of highly depleted rocks in a well-defined zone 140–190 km deep; diamond inclusions and diamond-bearing xenoliths show that most diamonds come from harzburgites within this layer. Xenocryst distribution curves indicate that diamondiferous kimberlites have sampled both garnet and chromite from the harzburgitic layer, but low-grade pipes have sampled only chromite. Diamond formation probably is due to the oxidation of methane-rich, silica-bearing fluids: Fe_2O_3 (in chromite) + $\text{CH}_4 \rightarrow \text{C} + \text{H}_2\text{O} + \text{FeO}$ (in chromite), accompanied by another reaction: $\text{chromite} \pm \text{olivine} \pm \text{orthopyroxene} + \text{Si, Ca (in fluid)} \rightarrow \text{low-Ca, high-Cr garnet}$. The presence or absence of diamond in kimberlites thus reflects the distribution of metasomatized fluid conduits in a lithospheric mantle that originally consisted of highly refractory harzburgites containing neither garnet nor diamond.

Keywords: diamond genesis, subcalcic garnet, mantle harzburgite, mantle metasomatism, Archean mantle.

INTRODUCTION

Diamonds provide important insights into processes in the Earth's mantle. In particular, mineral inclusions in diamonds define the rock types in which they occur at depth, prior to entrainment in kimberlitic or lamproitic magmas. Recognition of this sampling mechanism has led to a vast literature on diamond-inclusion (DI) mineralogy, beginning >30 yr ago (Meyer and Boyd, 1972; Gurney and Switzer, 1973; Sobolev et al., 1973). Whereas some diamonds may be derived from the transition zone or lower mantle (e.g., Stachel, 2001), the vast majority of DIs are consistent with derivation from the subcontinental lithospheric mantle (SCLM). DIs from individual kimberlite provinces tend to be dominated by either peridotitic or mafic (eclogitic or websteritic) parageneses. This study focuses on information derived from the peridotitic paragenesis, but has wider implications.

Peridotitic DIs define host rocks ranging from highly depleted (harzburgitic) to relatively fertile (lherzolitic), but most populations are overwhelmingly harzburgitic. The key minerals of this paragenesis are Al-poor chromite, subcalcic Cr-pyrope garnet (Fig. 1), magnesian olivine ($\text{Fo} \geq 92$), and enstatite, and it is effectively restricted to the SCLM beneath Archean cratons (Griffin et al. 2003, references therein). Subcalcic pyrope is commonly regarded as the most reliable indicator mineral in diamond exploration (e.g., Sobolev, 1971; Gurney and Zweistra, 1995).

The derivation of such depleted harzburgites from primitive-mantle compositions requires $\geq 30\%$ – 40% melt extraction (e.g., Walter 1998). The $f\text{O}_2$ of most harzburgitic and lherzolitic xenoliths is between the Ni-NiO and fayalite-magnetite-quartz (FMQ) buffers (Woodland and Koch, 2003);

under these relatively oxidizing conditions carbon behaves as an incompatible element during partial melting (e.g., Holloway, 1998). The presence of diamond in such highly depleted rocks thus is paradoxical; carbon should have been quantitatively removed during the formation of the harzburgites. This suggests that the formation of diamond in mantle peridotite involves the reintroduction of carbon by metasomatic fluids, but the nature of this metasomatic process is still unclear (Stachel et al., 2004).

Xenocryst populations (garnet, chromite) in kimberlites and other volcanic rocks can be used to map the vertical distribution of rock types and processes in the SCLM (Griffin et al., 1999a, 2003; O'Reilly and Griffin, 2006). Single-grain temperature (T) estimates (Ni in peridotitic garnet, Zn in chromite; Ryan et al., 1996) are referred to a known geotherm (derived from xenolith or xenocryst data) to derive a depth of origin for each grain, and thus to put the geochemical information from each grain in stratigraphic context. This information includes the rock type from which each garnet grain was derived (Griffin et al., 2002), and the types of depletion and metasomatism that rock has undergone. The relative proportions of garnet-bearing rock types can be expressed in a chemical tomography section for the SCLM beneath a kimberlite province (Fig. 2; O'Reilly and Griffin, 2006; see Table DR1 in the GSA Data Repository¹). Here we show how these studies can provide clues to the processes of diamond genesis in the SCLM.

KIMBERLITE SAMPLING PATTERNS

A chemical tomography section for the SCLM beneath the Daldyn kimberlite field in Yakutia (Fig. 2; see Griffin et al., 1999a) is consistent with available xenolith evidence (e.g., Boyd et al., 1997). It shows a concentration of strongly depleted rocks (harzburgites, depleted lherzolites) in a well-defined zone 140–190 km deep ($T = 850$ – 1050 °C), where they make up ~50% of the garnet-bearing rocks. At depths <140 km, harzburgites are rare and more fertile rocks dominate; below 190 km, the proportion of lherzolites increases with depth, and their trace element signatures show they have been affected by melt-related metasomatism (introduction of Fe, Ti, Zr, etc.). A section for the neighboring Alakit field (Griffin et al., 1999a) shows differences in detail, but the same pronounced harzburgite-rich layer. However, these sections, based on garnet xenocrysts, cannot show the relative abundance of garnet-bearing versus garnet-free rocks. Furthermore, such relative abundances are rarely available from studies of xenolith suites, which tend to focus on the garnet-bearing rocks for which pressure-temperature estimates can be made. Studies of DIs from these fields indicate that the diamonds originated from the 140–190 km harzburgite layer (Sobolev et al., 1973, 2004, and references therein), while diamond-bearing xenoliths from the Udachnaya pipe (Pokhilenko et al., 1991) show that dunite and harzburgite-dunite are the original host rocks of most diamonds.

Curves representing the depth distribution of garnet grains from kimberlite pipes in the Daldyn and Alakit fields (Fig. 2) show a marked dichotomy in terms of apparent sampling depth. High-grade pipes (Table 1)

¹GSA Data Repository item 2007082, Table DR1 (data for Figures 2–4), is available online at www.geosociety.org/pubs/ft2007.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, USA.

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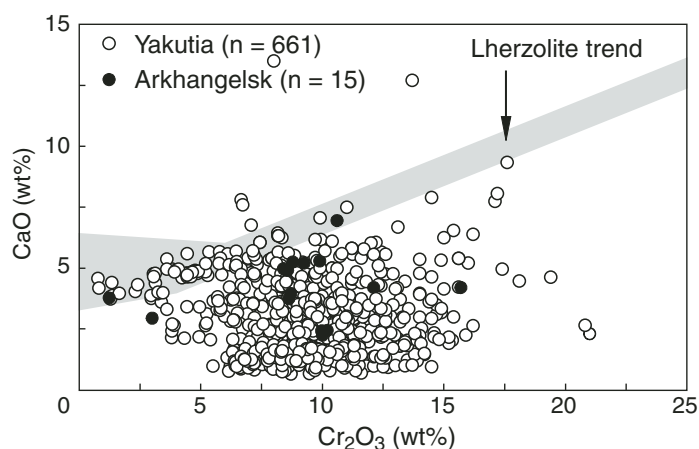


Figure 1. CaO-Cr₂O₃ data for garnet inclusions in diamonds from Daldyn-Alakit and Arkhangelsk kimberlite fields. Lherzolite trend represents garnets coexisting with clinopyroxene; lower-Ca (subcalcic) garnets are derived from highly depleted harzburgites-dunitites. Data are from Sobolev et al. (2004) and references therein.

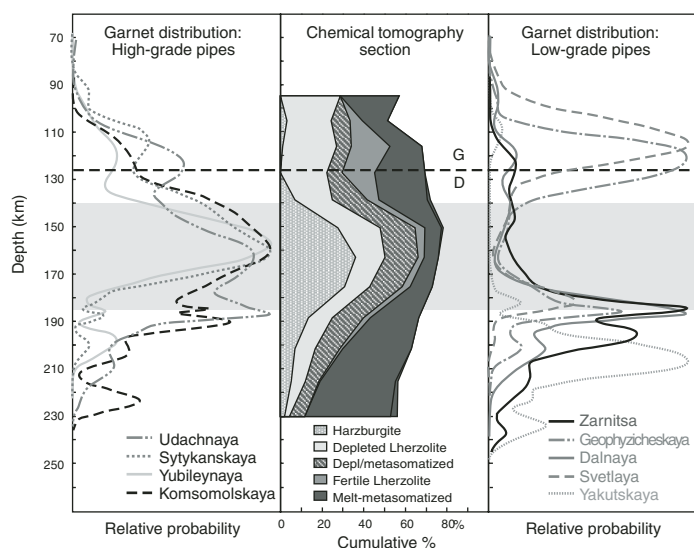


Figure 2. Chemical tomography section for Daldyn kimberlite field (9 kimberlites, 858 garnets), showing relative abundance at each depth of garnets from harzburgites, depleted lherzolites, depleted but metasomatized lherzolites, fertile lherzolites (also interpreted as metasomatic products), and lherzolites metasomatized by silicate melts (classifications of Griffin et al., 2002). Note strong concentration of harzburgitic garnets in 140–180 km depth range, marked by shaded horizontal band. Curves show distribution of sampling depths of garnet grains entrained in high-grade (left) and low-grade (right) kimberlites from Daldyn field and adjacent Alakit field. Horizontal dashed line is graphite (G)-diamond (D) phase transition.

contain a high proportion of garnets derived from the harzburgite-rich layer, while low-grade pipes contain few garnets from this depth range. Most low-grade pipes have entrained their garnets from depths below the harzburgite layer, but the Geophyzicheskaya pipe also contains a large proportion of lherzolitic garnets from shallower depths (110–135 km). These observations are consistent with a long-recognized general correlation between diamond grade, the abundance of garnet, and (especially) the proportion of harzburgitic garnets in individual pipes (Table 1).

The distributions shown in Figure 2 could record sampling of different depth ranges by the rising kimberlite magmas, perhaps reflecting differences in rheology and eruption rate. Alternatively, the sampling pat-

TABLE 1. DIAMOND GRADE, PYROPE AND CHROMITE CONTENTS AND PROPORTION OF HARZBURGITIC GARNETS IN SOME KIMBERLITIC PIPES FROM YAKUTIA AND ARKHANGELSK DIAMONDFEROUS PROVINCES

Pipe	Pyrope (g/t)	Chromite (g/t)	Diamond (ct/t)	Harzburgitic garnets (%)
Yakutia				
Udachnaya			2.1	16.4
Yubileynaya			1.28	5.7
Sytykanskaya			0.92	9.6
Komsomol'skaya			0.36	16.3
Osennaya			0.07	8.9
Zarnitsa			0.07	5.1
Dal'naya			0.05	
Massif				14.6
Breccia				0
Geophyzicheskaya			0.03	4
Yakutskaya			0.03	0
Svetlaya			0.005	6.1
Arkhangelsk				
Karpinskaya-I	1.0	40.6	1.2	5.1
Arkhangelskaya	8.6	61.7	0.95	0
Pionerskaya	4.9	46.8	0.45	8.2
Lomonosovskaya	5.7	55.2	0.4	1.1
Karpinskaya-II			0.3	
Snegurochka			0.28	
Pomorskaya	0.14	13	0.1	0
Solokha			traces	7.7

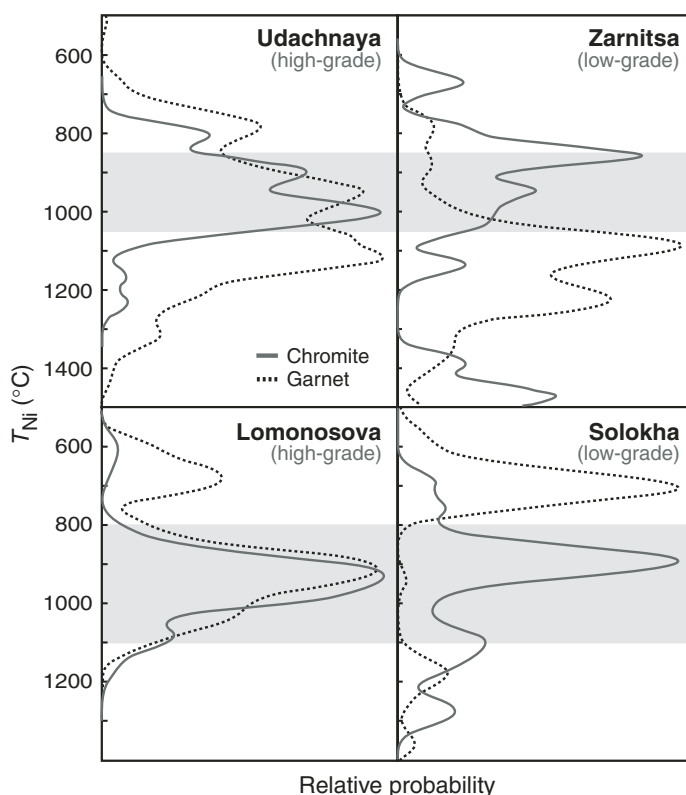


Figure 3. Temperature (depth) distribution of garnets and chromites in high- and low-grade kimberlites. A: Daldyn field, high-grade Udachnaya pipe and low-grade Zarnitsa pipe. B: Arkhangelsk field, high-grade Lomonosovskaya pipe and low-grade Solokha pipe. Shaded band shows temperature range of depleted harzburgite-rich layer beneath each field (cf. Fig. 2).

tern may reflect differences in the distribution of garnet in the SCLM. The depth distribution of chromite grains in these kimberlites (Fig. 3A) supports the second alternative. High-grade pipes such as Udachnaya have sampled both chromites and garnets from the harzburgite-rich layer of the SCLM; low-grade pipes such as Zarnitsa contain abundant harzburgitic chromites, but few garnets, from the 140–190 km depth range.

Similar observations apply to other kimberlite fields we have studied. For example, in the Arkhangelsk field (Griffin et al., 1996), garnet-bearing harzburgites are concentrated between 800 and 1100 °C (130–200 km), and diamond-inclusion studies show that most diamonds are of the harzburgitic paragenesis (Fig. 1; Sobolev et al., 1997). High-grade pipes such as Lomonosova sampled both garnets and chromites from this depth range (Fig. 3B); the low-grade Solokha pipe sampled chromites, but few garnets.

DISCUSSION AND CONCLUSIONS

The data shown in Figures 2 and 3 suggest that each kimberlite sampled the mantle over much of the depth range from 100 to >200 km, but that the mantle columns through which individual kimberlites passed had different compositions. At depths of 140–190 km beneath the Daldyn-Alakit kimberlite province, some volumes clearly contain chromite-bearing harzburgite and/or dunite, but little garnet and only rare diamond; others contain harzburgitic and lherzolitic garnet, chromite in at least some rock types, and diamond.

The genesis of the high-Cr, low-Ca garnets of the dunite-harzburgite paragenesis, found as xenocrysts, inclusions in diamonds, and in rare xenoliths, is still debated. Canil and Wei (1992) concluded that single-stage melting of peridotite does not produce residual garnets with >4% Cr₂O₃, and suggested multiple episodes of melt extraction, still leaving garnet-bearing residues. However, Cr-Al-Fe relationships in Archean peridotite xenoliths indicate that both Cr and Al were incompatible during the melt extraction that produced the harzburgites; this suggests that neither garnet nor spinel were residual phases (Griffin et al., 1998, 2003). In these very Al-depleted rocks, chromite may have exsolved from high-*T* complex orthopyroxenes on cooling.

The apparent absence of garnet in some chromite-bearing harzburgites beneath the Daldyn-Alakit area and Arkhangelsk kimberlite fields suggests that the garnet is a secondary phase, added by metasomatism. The strong correlation between harzburgitic garnets and diamonds (Table 1) allows two interpretations: (1) the garnet and the diamond are products of the same metasomatic episode, or (2) garnet formation is a necessary precursor to diamond formation. Stachel et al. (2004) argued that a metasomatic origin for low-Ca harzburgitic garnets is unlikely, because their sinuous rare earth element (REE) patterns would require a parental fluid more strongly light REE-enriched than known mantle fluids. However, trace element analyses of diamonds suggest that such a fluid may be an integral part of the diamond-forming process (Rege, 2005; Rege et al., 2006).

On average, low-grade pipes carry more oxidized chromite [i.e., with higher Fe³⁺/(Fe³⁺ + Cr + Al)] than high-grade pipes (e.g., Fig. 4), and dia-

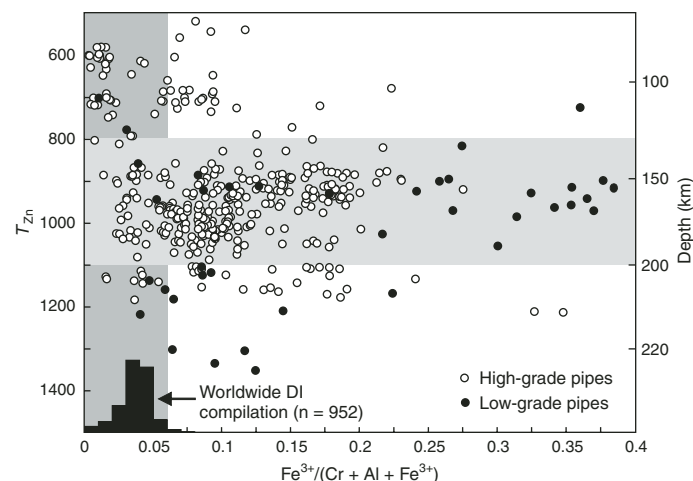


Figure 4. Fe³⁺/(Fe³⁺ + Cr + Al) of chromites vs. depth in kimberlites from Arkhangelsk field. Box shows range of values in diamond inclusions (DI) worldwide; horizontal band shows depth distribution of harzburgitic rocks.

mond-inclusion chromites have very low Fe³⁺ contents. These data suggest that the diamond formation is related to reduction of the wall rocks, which in turn implies that the metasomatic fluids were reducing. The estimated *f*O₂ of the deep lithospheric mantle is low enough to allow the existence and movement of CH₄ ± H₂O fluids (Woodland and Koch, 2003). We therefore suggest that the formation of diamond is related to the oxidation of asthenosphere-derived, methane-rich fluids: Fe₂O₃ (chromite) + CH₄ (in fluid) → C (diamond or graphite) + H₂O + FeO (chromite) (cf. Maruoka et al., 2004). The correlation between garnet and diamond suggests that another reaction proceeds roughly at the same time: chromite ± orthopyroxene + Si, Ca (in fluid) → subcalcic high-Cr garnet. These reactions may provide a direct genetic link between diamond and its most distinctive peridotitic indicator mineral.

A detailed study of a depleted harzburgite xenolith by Bell et al. (2005) described the metasomatism of such rocks by a Si-rich fluid, and showed by mass-balance calculations that such metasomatism could produce subcalcic garnet. Rege et al. (2006) suggested that fluid immiscibility is involved as well, with most diamonds crystallizing from a silicate-rich fraction, while a more carbonate-rich fraction reacts with chromite to produce garnet.

We suggest that where melts derived from below the lithospheric mantle first penetrate the harzburgitic lithosphere along fractures or other conduits, Si-bearing, CH₄-rich fluids released from them react with wall-rock harzburgite to produce subcalcic high-Cr garnet and diamond

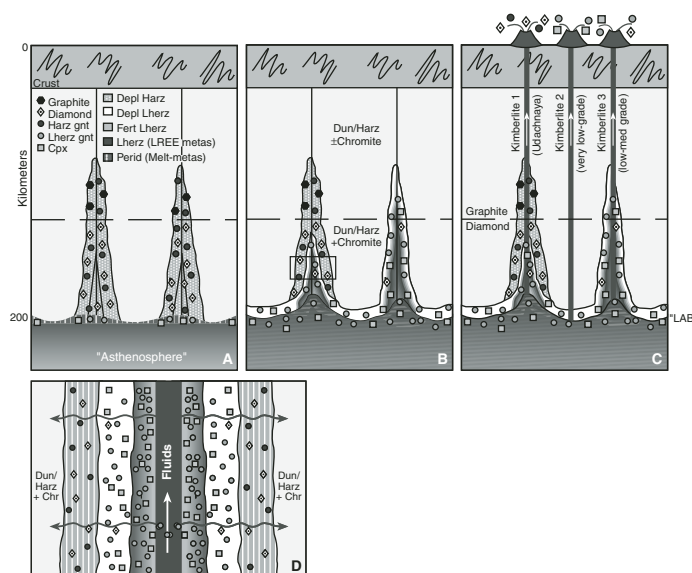


Figure 5. Cartoon showing schematic evolution of the subcontinental lithospheric mantle (SCLM) beneath the Daldyn-Alakit area. **A:** Primitive Archean SCLM, consisting of relatively oxidized harzburgite/dunite, is metasomatized by Si-bearing CH₄-rich fluids brought in by low-degree melts from the underlying “asthenosphere;” precipitation of diamond/graphite ± harzburgitic garnet near fluid conduits. Melt-related metasomatism near lithosphere-asthenosphere boundary (LAB) converts some harzburgites to fertile lherzolite by addition of Ca, Fe, Al. **B:** Continued input of melts/fluids; reduced harzburgite does not precipitate diamond/graphite; melt-related metasomatism refertilizes harzburgite to lherzolite at base of lithosphere and along conduits (weakly in left conduit, more extensively in right conduit); relict harzburgitic diamonds in lherzolites. **C:** Kimberlite eruption (Devonian); high-grade pipes sample remnants of Stage-A modified mantle. Barren pipes sample least-metasomatized mantle and lack harzburgitic garnets and diamonds; some low-grade pipes sample highly metasomatized mantle with relict diamonds. **D:** Detail of melt conduit showing progressive metasomatism of wall rocks, first by CH₄-rich fluids expelled from melts, and then by the melts themselves. Abbreviations: Dun—dunite; Harz—harzburgite; Lherz—lherzolite; Fert—(re)-fertilized; Perid—peridotite; Cpx—clinopyroxene; Gnt—garnet; Chr—chromite; LREE—light rare earth elements; metas—metasomatized

(Fig. 5A). As the asthenosphere-derived melts penetrate further into the lithosphere, metasomatism adds Ca, Al, and Fe, refertilizing depleted harzburgite to produce lherzolites (Figs. 5B, 5D). Where this refertilization process is preserved in the zoned garnets of mantle xenoliths (Griffin et al., 1999b; Pokhilenko et al., 1999) it is accompanied by reoxidation of the protolith (McCammon et al., 2001). A similar refertilization has been proposed for the genesis of lherzolitic mantle beneath Lesotho, with garnet and clinopyroxene crystallizing at the expense of olivine + orthopyroxene (Cox et al., 1987; Simon et al., 2003).

This simple metasomatic model has many implications. (1) There is a direct genetic connection between peridotitic diamond and its most common indicator minerals (subcalcic garnet, chromite). (2) The distribution of diamonds in the lithosphere should be laterally heterogeneous on relatively small scales, related to ancient structural controls. (3) The grade of a kimberlite will depend on its intersecting these previously active fluid conduits (Fig. 5C). (4) The original Archean SCLM is represented by the garnet-free dunite and/or harzburgites. (5) Existing models of Archean SCLM, which are based heavily on xenoliths from active mines, are biased toward multiply metasomatized materials, which do not represent residues of partial melting. (6) The oxidation state of pristine Archean SCLM, derived from analysis of such xenoliths, may be incorrect. These implications call for a reevaluation of the nature and origin of Archean SCLM, and the reasons for its marked differences from younger SCLM (Griffin et al., 1998, 2003).

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