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THE PRESSURES AND TEMPERATURES OF FORMATION OF DIAMOND BASED ON THERMOBAROMETRY OF CHROMIAN DIOPSIDE INCLUSIONS

PAOLO NIMIS[§]

Dipartimento di Mineralogia e Petrologia, Università di Padova, C.so Garibaldi 37, I-35137 Padova, Italy

ABSTRACT

To date, the evaluation of the temperature (*T*) and pressure (*P*) of formation of natural diamond has generally relied on thermobarometry of rare polymineralic inclusions containing appropriate assemblages of minerals. The results are commonly ambiguous, because of potential re-equilibration of touching minerals after diamond growth and possible disequilibrium between non-touching minerals. Here, I calculate *T* and *P* for over one-hundred inclusions of chromian diopside (mostly isolated) in diamond crystals containing inclusions of peridotitic material, and for peridotite xenoliths from worldwide occurrences, using single-clinopyroxene thermobarometers. The results provide constraints on the conditions and relative timing of diamond genesis. Inclusions in diamond and xenoliths from the same source commonly yield similar *P–T* values, suggesting that diamond crystals formed when the lithospheric mantle had already attained a conductive thermal regime comparable to or even colder than that extant at the time of emplacement of the host kimberlite or lamproite. Some inclusions record thermal or metasomatic events, which can be ascribed to the ascent of hot C-rich fluids from which the diamond precipitated. In a few cases, secular cooling of the cratonic lithosphere is believed to be a possible source of scatter in *T* estimates. In general, there is no evidence for occurrences of diamond being concentrated at particular levels in the lithosphere. Where significant gaps occur in the distribution of diamond crystals with included lherzolitic material, they are associated with a scarcity of lherzolitic material in the mantle and do not necessarily correspond to a real absence of diamond crystals. The results support the use of chromian diopside thermobarometry as a complementary tool for assessment of diamond potential in exploration programs.

Keywords: diamond, thermobarometry, chromian diopside, inclusions, upper mantle.

SOMMAIRE

Jusqu'à ce point, l'évaluation de la température (*T*) et de la pression (*P*) de formation de cristaux de diamant portait en général sur la thermobarométrie de rares inclusions polyminérales contenant un assemblage approprié de minéraux. Les résultats sont en général assez ambigus, à cause du ré-équilibrage potentiel des minéraux en contact après la croissance du diamant et du déséquilibre possible parmi les minéraux qui ne sont pas en contact. Ici, je détermine la température et la pression en utilisant plus d'une centaine d'inclusions de diopside chromifère (grains isolés pour la plupart) dans des cristaux de diamant contenant des inclusions dérivées de matériau péridotitique, et des xénolithes de péridotite provenant des mêmes indices, répartis sur une échelle mondiale, en utilisant des thermobaromètres fondés sur le seul clinopyroxène. Les résultats fournissent des contraintes sur les conditions physiques et sur l'âge de la croissance du diamant. Les inclusions dans le diamant et les xénolithes provenant de la même source produisent en général des valeurs *P–T* semblables, ce qui fait penser que le diamant s'est formé quand le manteau lithosphérique avait déjà atteint un régime thermique conductif comparable à celui qui existait lors de la mise en place de la kimberlite ou la lamproïte hôte, voire même plus froid que celui-ci. Certaines inclusions ont enregistré des événements thermiques ou métasomatiques attribuables à l'ascension d'une phase fluide porteuse de carbone, à partir de laquelle le diamant s'est formé. Dans quelques cas, un refroidissement séculaire de la lithosphère cratonique serait une source possible d'écarts dans l'évaluation de la température. En général, il n'y a aucune indication que le diamant est réparti dans des niveaux particuliers dans la lithosphère. Où il existe des lacunes importantes dans la distribution du diamant contenant des inclusions de matériau lherzolitique, celles-ci seraient associées à la rareté de matériau lherzolitique plutôt qu'à une absence relative de diamant. Les résultats étayent l'utilisation du diopside chromifère dans des applications thermobarométriques comme complément dans une évaluation des ressources potentielles dans les programmes d'exploration pour le diamant.

(Traduit par la Rédaction)

Mots-clés: diamant, thermobarométrie, diopside chromifère, inclusions, manteau supérieur.

§ *E-mail address*: paolon@dmp.unipd.it

INTRODUCTION

Mineral inclusions in diamond are among the oldest samples of the Earth's interior (Boyd *et al.* 1985, Richardson *et al.* 1993). Thermobarometry of such inclusions, essential to retrieve a geochemical and thermal stratigraphy of the Earth's mantle through geological time, has been used to establish the existence of cold, thick cratonic lithospheres since the Archean (*e.g.*, Boyd *et al.* 1985). Existing thermobarometric data on inclusions of peridotitic assemblages are still fragmentary and, in many instances, ambiguous (Meyer & Tsai 1976, Tsai *et al.* 1979, Gurney *et al.* 1979, Hervig *et al.* 1980, Boyd *et al.* 1985, Jaques *et al.* 1994, Harris *et al.* 1994, Wilding *et al.* 1994, Phillips & Harris 1995, Girnis *et al.* 1999, Viljoen *et al.* 1999, Wang & Gasparik 2001). A major drawback of conventional thermobarometry is that at least two equilibrium mineral phases must be present, and their compositions must be known to obtain an estimate of *P and T* conditions. Polymineralic inclusions are uncommon, however, and even more rarely do they contain the appropriate mineral assemblage for optimum thermobarometry. Additional problems concern the "touching" or "non-touching" character of the included minerals. On the one hand, minerals that are not in contact could be entrained at different stages and *P–T* conditions during the growth of the host diamond, thus leading to incorrect thermobarometric results. On the other hand, touching minerals may have re-equilibrated to conditions different from those extant at the time of their entrapment (Meyer & Tsai 1976, Phillips & Harris 1995). Girnis *et al.* (1999) derived a method to calculate the composition of missing phases and estimate *P–T* for non-touching inclusions. Their method is affected by relatively large uncertainties if the composition of orthopyroxene is not available; it becomes highly unreliable if the composition of garnet is unknown. Although somewhat controversial (Canil 1999), combined major- and trace-element analysis of included garnet from a peridotitic paragenesis can provide useful thermobarometric indications (Ryan *et al.* 1996). It has the advantage of being applicable to isolated inclusions of garnet, provided a constant composition of olivine is assumed, but can only yield minimum estimates of pressure, making it difficult to assign unequivocal *P–T* values to individual examples of diamond. Among barometric methods, measurement of confining pressures on inclusions of olivine in diamond (Izraeli *et al.* 1999) can be helpful, but provides no thermometric information.

Here, I apply the clinopyroxene (Cpx) thermometer and barometer of Nimis & Taylor (2000) to over one-hundred inclusions of a peridotitic paragenesis in diamond from worldwide occurrences using electron-microprobe data on clinopyroxene from various sources. These inclusions are generally believed to be syngenetic with diamond, on the basis of absence of alteration and visible fractures and the typical octahedral habit imposed by the host diamond. In most published works, unfortunately, detailed descriptions of inclusion morphology are lacking; as a result, the protogenetic *versus* syngenetic nature of the inclusions remains undetermined (*cf.* Meyer 1987). Although based on the composition of clinopyroxene alone, Cpx-thermobarometry requires that clinopyroxene be in equilibrium with both orthopyroxene (Opx) and garnet (Grt). Application of this method is therefore restricted to those inclusions for which an origin from garnet peridotite can be established. Thermobarometric data for inclusions in diamond will be compared with those obtained for lherzolite xenoliths from the same kimberlitic or lamproitic host. The results will be used to constrain the depth of origin of the diamond crystals and to obtain indications on the temperature conditions of their formation. Implications on diamond-exploration strategies will be briefly discussed.

In the following, the adjective *chromian* will be used to denote diopside with more than 0.5 wt% Cr_2O_3.

CLINOPYROXENE THERMOBAROMETRY

The enstatite-in-clinopyroxene thermometer and Cr-in-clinopyroxene barometer of Nimis & Taylor (2000) enable one to retrieve *P and T* conditions of equilibration of mantle-derived chromian diopside, assumed to be in equilibrium with orthopyroxene (for *T*) and garnet (for *P*), from the composition of clinopyroxene alone. Although inclusions of the lherzolitic (*i.e.*, clinopyroxene-bearing) paragenesis are, with a few notable exceptions (Jaques *et al.* 1994, Stachel *et al.* 1998, 2000), far less common than those of the harzburgitic paragenesis, the pressure and temperature of formation of a large number of diamond crystals can be systematically evaluated using their inclusions of chromian diopside. Equilibrium with orthopyroxene and garnet can be inferred on the basis of compositional analogy with diopside from peridotitic xenoliths (*e.g.*, Ramsay & Tompkins 1994, Nimis 1998; Fig. 1). An appraisal of uncertainties on *P–T* estimates has been given by Nimis & Taylor (2000). A more detailed discussion is given here, with particular emphasis on applications to chromian diopside included in diamond.

Uncertainties in P–T estimates

The precision of the thermobarometric method can be assessed from the distribution of *P–T* estimates for grains of chromian diopside derived from the same mantle section (see Fig. 8 in Nimis & Taylor 2000). The apparent *T* range at a given *P* can be as large as 150°C, and uncertainties of ±50°C *and* ±0.3 GPa must be allowed so as to reconcile all *P–T* points with a single geotherm. The above uncertainties include propagation of errors on results of chemical analyses, but somewhat larger errors may arise from the use of published electron-microprobe data acquired at different analytical

facilities. Simple tests indicate that (i) relatively large errors (*ca.* 10% rel.) on pressure estimates can only propagate from large (10% rel.) errors in Al_2O_3 or Na_2O determinations; (ii) significant errors (>50°C) on *T* estimates can only be expected at low *T* (<900°C) in response to unusually large errors on CaO, SiO_2 (≥2% rel.) or Na_2O (≥10% rel.) determinations.

An overall indication of the robustness of the method is provided by the fact that *P–T* estimates obtained by Nimis & Taylor (2000) for eleven graphite-associated samples of diopside of various provenance are within the stability field of graphite, whereas those for about one hundred diamond-associated samples of diopside, analyzed at several different laboratories, are either within the stability field of diamond or within ±50°C and ±0.3 GPa of the graphite–diamond boundary. Application of the same method to experimentally equilibrated clinopyroxene compositions in peridotitic assemblages that were not used in the calibration of the thermobarometer provides a further test. Nimis & Taylor (2000) showed that the thermometer reproduces well (standard error of estimate: 40°C) experimental temperatures for clinopyroxene in the CMAS and NCMAS systems in the range 900–1615°C. An analogous test of the barometer, using high-*P* experimental data on peridotitic systems, is illustrated in Figure 2. The barometer reproduces experimental conditions for experiments at 2.8–3.3 GPa and 1468–1538°C by Robinson & Wood (1998) and experiments at 3–5 GPa and 1500–1680°C by Walter (1998) to within ±0.3 GPa, whereas Walter's experiments at 6 and 7 GPa (1670–1820°C) are underestimated by *ca.* 0.6 GPa and 0.6–1.0 GPa, respectively.

From the above, it appears that a total of ±50°C and ±0.3 GPa can be considered an adequate error-allowance for *P–T* estimates in proximity of the graphite–diamond boundary. For pressures above 5 GPa, uncertainties on barometric estimates are potentially larger, and pressures are probably progressively underestimated, at least at very high temperatures. However,

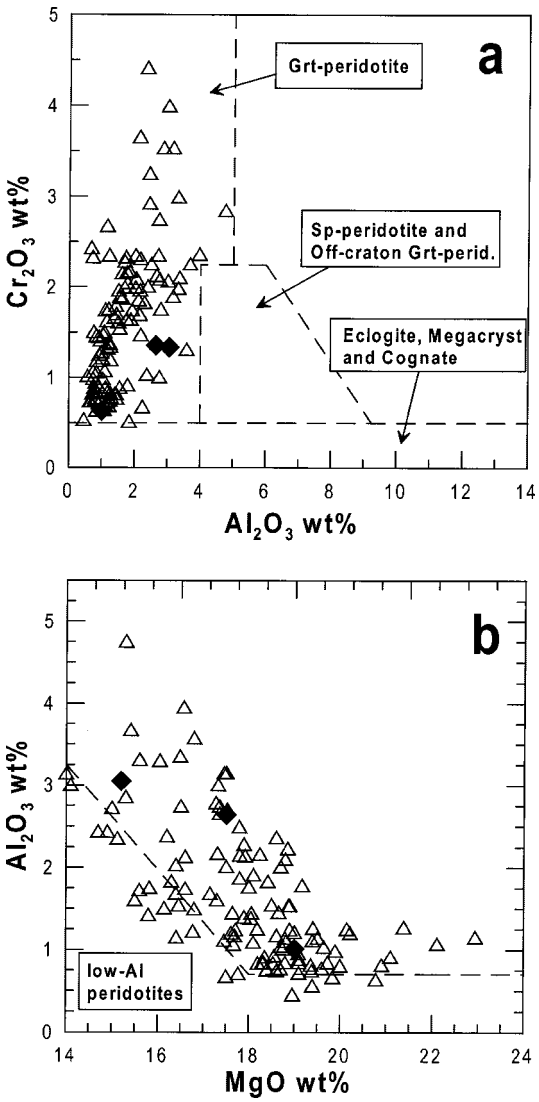


FIG. 1. Chromian diopside included in diamond (triangle) and intergrown with diamond (solid diamond) in the discriminant diagrams of (a) Ramsay & Tompkins (1994) and (b) Nimis (1998). The sources of data are listed in Table 1.

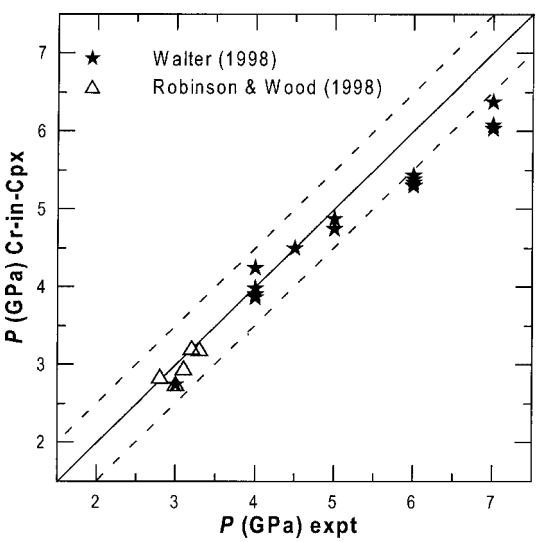


FIG. 2. Test of the Cr-in-clinopyroxene barometer against experiments on peridotite systems (lherzolitic and wehrlitic assemblages of minerals near the solidus).

the possible systematic error is not sufficient to produce a significant bias in interpretations of thermobarometric results. Geotherms based on clinopyroxene thermobarometry may show an excessive $\Delta T/\Delta P$ gradient at $P > 5$ GPa, yet their relative significance will be maintained. The possible bias on estimates of *P* of diamond formation produced by uncertainties on *T* is fairly limited. First, the temperature dependence of the barometer, which is based on clinopyroxene–garnet equilibria, is minimal, corresponding to ~0.1–0.25 GPa for a 50°C increase in temperature (Nimis & Taylor 2000), a gradient similar to that of the graphite–diamond boundary. Second, orthopyroxene and olivine can only accommodate minor amounts of the *P*-sensitive cations Cr, Na and (for olivine) Al, and the Al distribution between garnet-facies pyroxenes is reasonably insensitive to *T* (*e.g.*, Nickel *et al.* 1985). Pyroxene thermometry is in turn virtually insensitive to barometric uncertainties (*ca.* 20°C for a 1 GPa increase).

Isolated inclusions of clinopyroxene that can be deemed to represent former portions of garnet lherzolite guarantee the best indication of *P* and *T* at the time of encapsulation, because the absence of other phases prevents any subsequent chemical re-equilibration. Polymineralic inclusions containing the assemblage clinopyroxene ± garnet ± olivine should also yield sound estimates, because the composition of the clinopyroxene in orthopyroxene-free assemblages would be little affected by a variation in temperature (*cf.* Nimis & Taylor 2000). If one assumes negligible movement of the mantle within the lithospheric keels of cratons, both the orthopyroxene-free polymineralic inclusions and the isolated inclusions of clinopyroxene should yield *P–T* values close to those extant at the time of (if syngenetic), or shortly before (if protogenetic), their entrapment in the host diamond. Polymineralic inclusions containing both pyroxenes would instead easily respond to temperature changes and would therefore reflect the ambient thermal state of the mantle at the time of eruption.

Taking into account all the above considerations, the possible bias on *P* estimates for both mono- and polymineralic inclusions should be restricted in most cases to a few tenths of a GPa, with possible systematic underestimation at very high *P*. Only for the rare cases of touching, orthopyroxene-bearing, garnet-free inclusions may underestimation of *P* exceed 1 GPa, owing to potential re-equilibration over a few hundred degrees (*e.g.*, Griffin *et al.* 1993) and the consequent incorrect assumption of *T* at the *P* calculated.

Geotherm assessment

In applications to well-equilibrated lherzolite xenoliths, the Nimis & Taylor (2000; NT00) combined enstatite-in-Cpx – Cr-in-Cpx method usually yields *P–T* estimates that are in reasonable agreement with those yielded by the widely used Brey & Köhler (1990) T_{BKN}–P_{BKN} thermobarometric pair. Minor recognized systematic deviations are as follows: (i) NT00 estimates of temperature are systematically lower by *ca.* 50°C; (ii) at $P_{BKN} > 5$ GPa, the two barometers commonly show poor agreement, which can be ascribed in part to systematic underestimation of P_{NT00} at high pressure and in part to poor pyroxene–garnet equilibration in many high-*P–T* samples. The temperature dependence of the BKN barometer (and of any Al-in-Opx barometer) is such that errors in *T* estimates will move calculated *P* roughly along theoretical geotherms (Brey & Köhler 1990). The weaker *T*-dependence of the NT00 Cr-in-Cpx barometer will generally increase the scatter of *P–T* estimates around theoretical geotherms, thus amplifying the uncertainty in the determination of model heat-flows at the surface. In order to maintain internal consistency, *P–T* estimates have been calculated for both inclusions in diamond and xenoliths using the NT00 thermobarometric scheme. Given the possible systematic errors, best-fit logarithmic "geotherms" have been computed (Figs. 3–5) rather than fits of xenolith *P–T* data to theoretical conductive mantle curves. Average differences between estimated *T* of diamond formation at *P* and geotherm *T* at the same *P* are given in Table 1. In addition, the widely used theoretical geotherms of Pollack & Chapman (1977), although in part questionable (Smith 1999), are used throughout as reference curves to facilitate comparison with results given elsewhere.

Data selection

For the present study, only clinopyroxene inclusions showing Cr_2O_3 *versus* Al_2O_3 relations consistent with the garnet peridotite field of Ramsay & Tompkins (1994) were selected (Fig. 1a). In the Al_2O_3 *versus* MgO plot, all inclusions selected fall above or immediately below the line used by Nimis (1998) to separate diopside in garnet lherzolites from that in low-Al, garnet-absent, metasomatized peridotites (Fig. 1b). If such low-Al diopside were indeed part of garnet-free assemblages, its pressure of equilibration could be highly overestimated. Most of the cases, however, yield *P–T* estimates consistent with those produced by other inclusions from the same localities, which argues for an origin from garnet peridotite. Therefore, rather than discarding all low-Al inclusions, those that produce apparently anomalous *P–T* estimates will be pointed out where appropriate. To minimize uncertainties in *P* estimates, compositions for which the *P*-dependent parameter $Cr - 0.81 \cdot Na \cdot [Cr/(Cr + Al)]$ is <0.003 atoms per formula unit, or for which the Cr content is greater than 5 wt% Cr_2O_3, have not been considered (as recommended by Nimis & Taylor 2000). About one-tenth of the chromian diopside inclusions previously selected on the basis Cr_2O_3 *versus* Al_2O_3 relations were rejected on these criteria (Table 1). Most of the available compositional data on inclusions in diamond pertain to diamond from the classic kimberlite fields of the Kaapvaal (southern Africa) and Siberian cratons (Figs. 3, 4). These two

TABLE 1. NUMBER OF SAMPLES USED FOR THERMOBAROMETRIC STUDY, NUMBER OF SAMPLES REJECTED, SOURCES OF DATA, AND COMPARISON OF TEMPERATURE OF DIAMOND FORMATION AND LOCAL GEOTHERM BASED ON XENOLITHS

Locality	1	2	3	4	5	6	7	8	9
Kaapvaal									
Premier	33	2	–	–	39	3	–56°C*	Richardson *et al.* (1993, unpubl. data), Tsai *et al.* (1979), Gurney *et al.* (1985, 1986)	Danchin & Boyd (1976), Pearson *et al.* (1994), Smith (1999), P. Nimis (unpubl. data)
Kimberley	3	–	–	–	58	6	+39°C	Wilding *et al.* (1994)	Cox *et al.* (1987), Luth *et al.* (1990), Boyd & Nixon (1975), Nimis & Taylor (2000), F.R. Boyd (unpubl. data)
Venetia	2	–	–	–	7	–	+111°C	Viljoen *et al.* (1999)	Stiefenhofer *et al.* (1999)
Koffiefontein	4	1	–	–	–	–	n.c.	Rickard *et al.* (1989)	
Roberts Victor	2	1	2	–	15	1	+23°C	Gurney *et al.* (1984), Viljoen *et al.* (1994)	Viljoen *et al.* (1994), T. Stachel (unpubl. data)
Dokolwayo	1	2	–	–	–	–	n.c.	Daniels & Gurney (1989)	
Jagersfontein	1	–	–	–	8	–	n.c.	Meyer *et al.* (1977)	Cox *et al.* (1987), Luth *et al.* (1990), Pearson *et al.* (1994), F.R. Boyd (unpubl.)
Monastery	1	–	–	–	1	–	n.c.	Moore & Gurney (1989)	Boyd & Nixon (1975)
Mothae	–	–	1	–	12	–	n.c.	Dawson & Smith (1975)	Nixon & Boyd (1973), Cox *et al.* (1987), Luth *et al.* (1990)
Finsch	–	–	2	–	10	–	n.c.	Shee *et al.* (1982)	Gurney *et al.* (1979), Skinner (1989)
uncertain	–	–	–	1	–	–	n.c.	Boyd & Nixon (1970)	
Siberia									
Udachnaya	2¶	–	1	–	42	10	n.c.	Sobolev (1977), Boyd & Finnerty (1980)	Boyd *et al.* (1976, 1997), Sobolev (1977), Pokhilenko *et al.* (1991), Solovjeva *et al.* (1997), Griffin *et al.* (1996), Taylor & Nimis (in prep.)
Malo Botuobiya (Mir–Sputnik)	17	1	–	2	32	6	+39°C	Sobolev *et al.* (1976, 1997a), Sobolev (1977)	Boyd *et al.* (1976), Sobolev (1977), Roden *et al.* (1997), Taylor & Nimis (in prep.)
Western Australia									
Argyle	–	–	3	–	65	–	n.c.	Jaques *et al.* (1990)	Atkinson *et al.* (1984), Jaques *et al.* (1990), Taylor & Nimis (in prep.)
Ellendale	2	1	–	–	45	11	+55°C	Jaques *et al.* (1989, 1994)	Jaques *et al.* (1984), Taylor & Nimis (in prep.)
King George River	1	–	–	–	–	–	n.c.	Sobolev *et al.* (1989)	
Eastern Europe									
Urals	1	–	–	–	–	–	n.c.	Sobolev (1977)	
Arkhangel'sk	2	1	–	–	–	–	n.c.	Sobolev *et al.* (1997b)	
Western Africa									
Birim, Ghana	5	–	–	–	–	–	n.c.	Stachel & Harris (1997a,b)	
Kankan, Guinea	3	–	–	–	–	–	n.c.	Stachel *et al.* (2000)	
Central Africa									
Mwadui, Tanz.	7	–	–	–	3	–	–23°C	Stachel *et al.* (1998, 1999)	Nixon (1987)
Wyoming–Colorado									
Sloan	1	–	–	–	94	19	+141°C	Otter & Gurney (1989)	McCallum & Eggler (1976), Eggler & McCallum (1976), Hearn & McGee (1984), Kirkley *et al.* (1984), McCandless *et al.* (1995), D.H. Eggler (unpubl.)
George Creek	1§	–	–	–	–	–	n.c.	Chinn (1995)	
Sino-Korean craton									
Liaoning	10	1	–	–	–	–	n.c.	Harris *et al.* (1994), Wang & Gasparik (2001)	
Shandong	1	–	–	–	–	–	n.c.	Meyer *et al.* (1994)	
Guyana–Guapore									
São Luiz, Brazil	1	–	–	–	–	–	n.c.	Hutchison (1997)	

Column headings: 1. Inclusions selected. 2. Inclusions rejected, being outside the field of garnet peridotite (*cf.* Fig. 1) of Ramsay & Tompkins (1994), >5 wt% Cr_2O_3, or pressure-dependent parameter $[Cr - 0.81 \cdot Na \cdot Cr/(Cr + Al)] < 0.003$. 3. Diamond-bearing xenoliths. 4. Intergrowths with diamond. 5. Xenoliths, xenocrysts, loose grains. 6. Xenoliths, loose grains rejected, having >5 wt% Cr_2O_3, $[Cr - 0.81 \cdot Na \cdot Cr/(Cr + Al)] < 0.003$, *i.e.*, low-Al according to Nimis (1998, *cf.* Fig. 1), or unacceptable P–T estimates. 7. Average ΔT, defined as the average difference between estimated T of diamond formation at P and xenolith geotherm at the same P (n.c.: not calculated owing to lack of data). 8. Source for inclusions, diamond-bearing xenoliths and intergrowths with diamond. 9. Source for xenoliths, xenocrysts and loose grains from heavy- mineral concentrates. * Six high-T inclusions in diamond excluded from the calculation. ¶ Probably wehrlitic. § Probably websteritic.

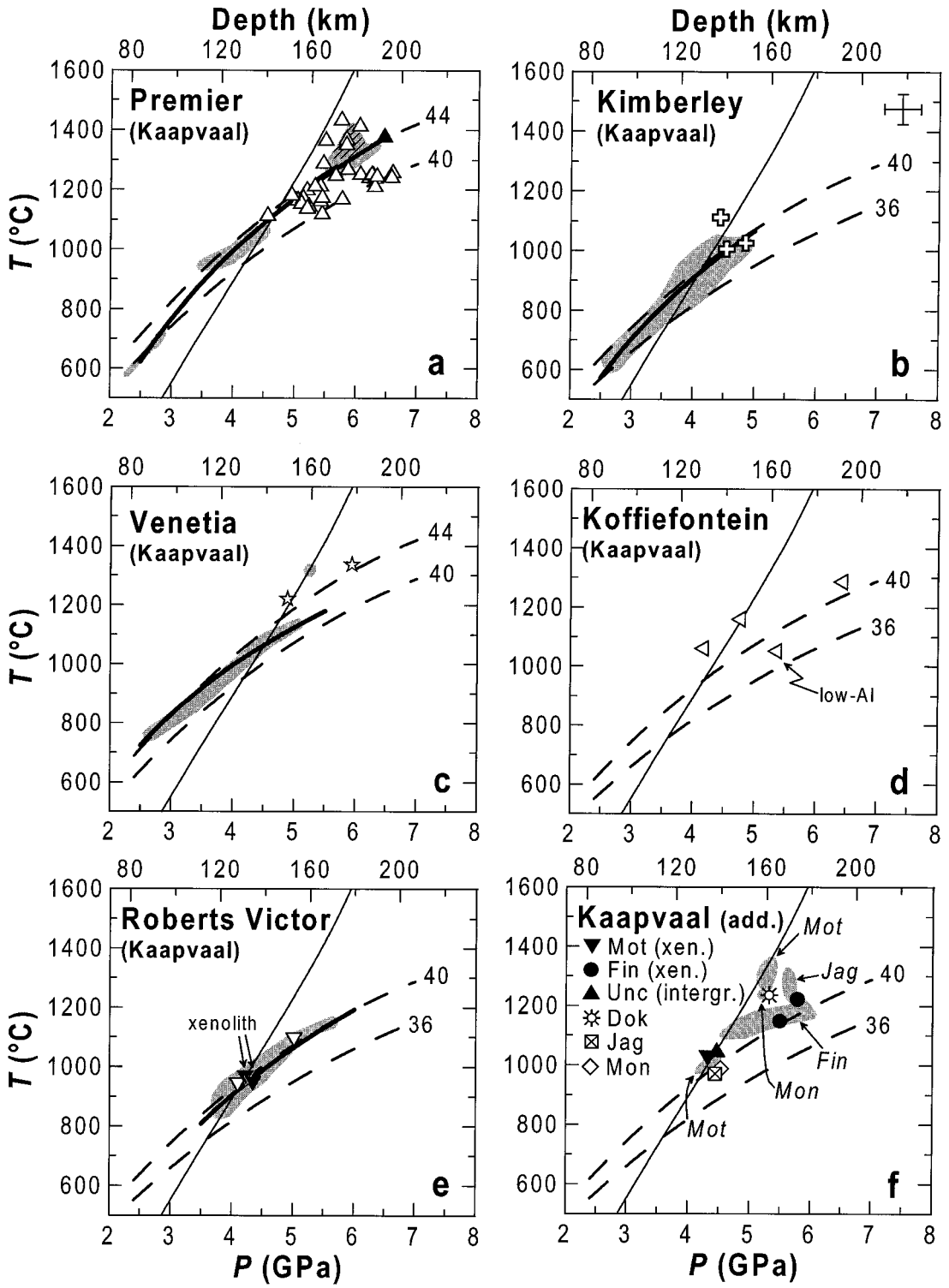

Depth (km)
80 120 160 200
Premier
(Kaapvaal)
44
40
a
Kimberley
(Kaapvaal)
40
36
b
Venetia
(Kaapvaal)
44
40
c
Koffiefontein
(Kaapvaal)
40
36
low-Al
d
Roberts Victor
(Kaapvaal)
xenolith
40
36
e
Kaapvaal (add.)
Mot (xen.)
Fin (xen.)
Unc (intergr.)
Dok
Jag
Mon
Mot
Jag
40
36
Fin
Mon
Mot
f
T (°C)
P (GPa)
1600
1400
1200
1000
800
600
2 3 4 5 6 7 8

FIG. 3. *P–T* estimates for inclusions of chromian diopside in diamond from the Kaapvaal craton according to single-clinopyroxene thermobarometry. *Shaded fields* encompass *P–T* conditions for lherzolitic xenoliths, and for xenocrysts and concentrates of clinopyroxene from the same kimberlite calculated using the same method. *Ruled fields* refer to sheared-type, high-*T* xenoliths. *Solid curves* are best fits through xenolith data (high-*T* sheared-type xenoliths excluded). *Open symbols* refer to orthopyroxene-free inclusions and reflect absolute *P* and minimum *T* at the time of their entrapment in the growing diamond. *Solid symbols* refer to orthopyroxene+garnet-bearing inclusions and, where indicated, to diamond-bearing xenoliths and intergrowths with diamond, and reflect ambient *P–T* conditions at the time of eruption. *Crossed symbols* refer to orthopyroxene-bearing, garnet-free inclusions and reflect *T* conditions at the time of eruption and minimum *P* at the time of entrapment (see text). The graphite–diamond boundary (steep solid curve) is after Chatterjee (1991) and Kennedy & Kennedy (1976). *Dashed curves* labeled 36 to 44 are reference conductive geotherms for different model heat-flows (mWm^{-2}) at the surface, after Pollack & Chapman (1977). Error bars are estimated errors on *P* and *T* in proximity of the graphite–diamond boundary. Errors on *P* can be larger at higher pressures. Dok: Dokolwayo; Mot: Mothae; Fin: Finsch; Jag: Jagersfontein; Mon: Monastery; Unc: uncertain locality. Data sources are listed in Table 1.

groups will be discussed in greater detail, with emphasis on orthopyroxene-free inclusions. Data from other cratons are shown in the same format in Figure 5 for ease of comparison.

RESULTS

Single-clinopyroxene thermobarometry shows that in several cases, inclusions in diamond equilibrated under apparently normal conductive regimes that are usually within error of those extant at the time of eruption as recorded by single-clinopyroxene thermobarometry of xenoliths from the same host (Figs. 3–5, Table 1). Such examples are represented by inclusions in diamond from Tanzania, China and the Roberts Victor pipe, Kaapvaal, South Africa. Alignment of *P–T* data along geotherms resembling theoretical steady-state geotherms gives support to the assumption of equilibrium among clinopyroxene, orthopyroxene and garnet. Inclusions from several other sources follow at least in part an analogous distribution. Nonetheless, inclusions from some localities (*e.g.*, Mir–Sputnik, Siberia; Ghana, Western Africa; Koffiefontein, Kaapvaal) show a significant scatter of thermobarometric data. The scatter may reflect diachronous growth of diamond during secular cooling of the lithosphere (*cf.* Phillips & Harris 1995), or thermal perturbations, or disequilibrium growth of clinopyroxene shortly before or during diamond formation. In the case of Koffiefontein, the low-

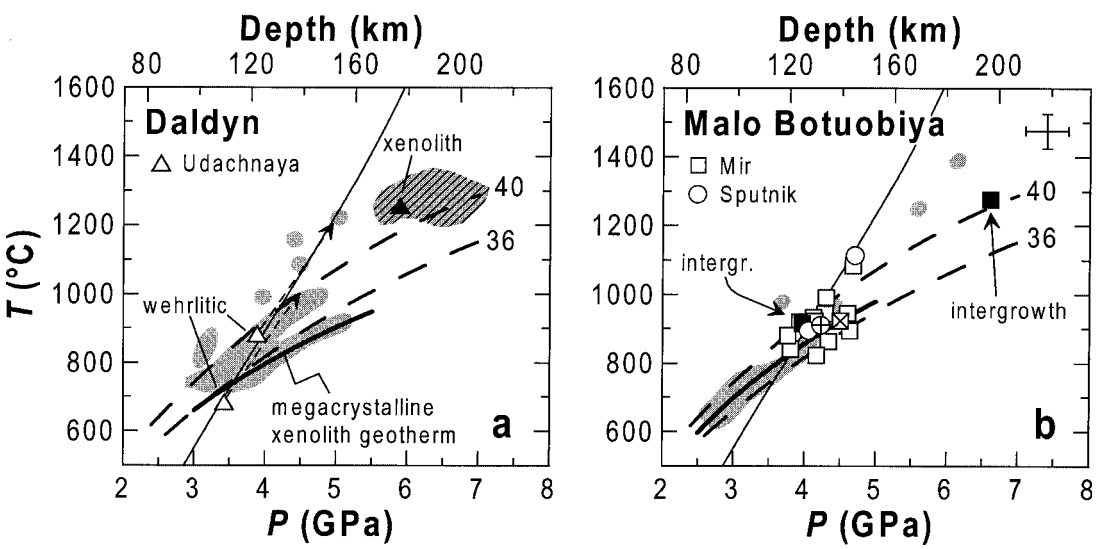
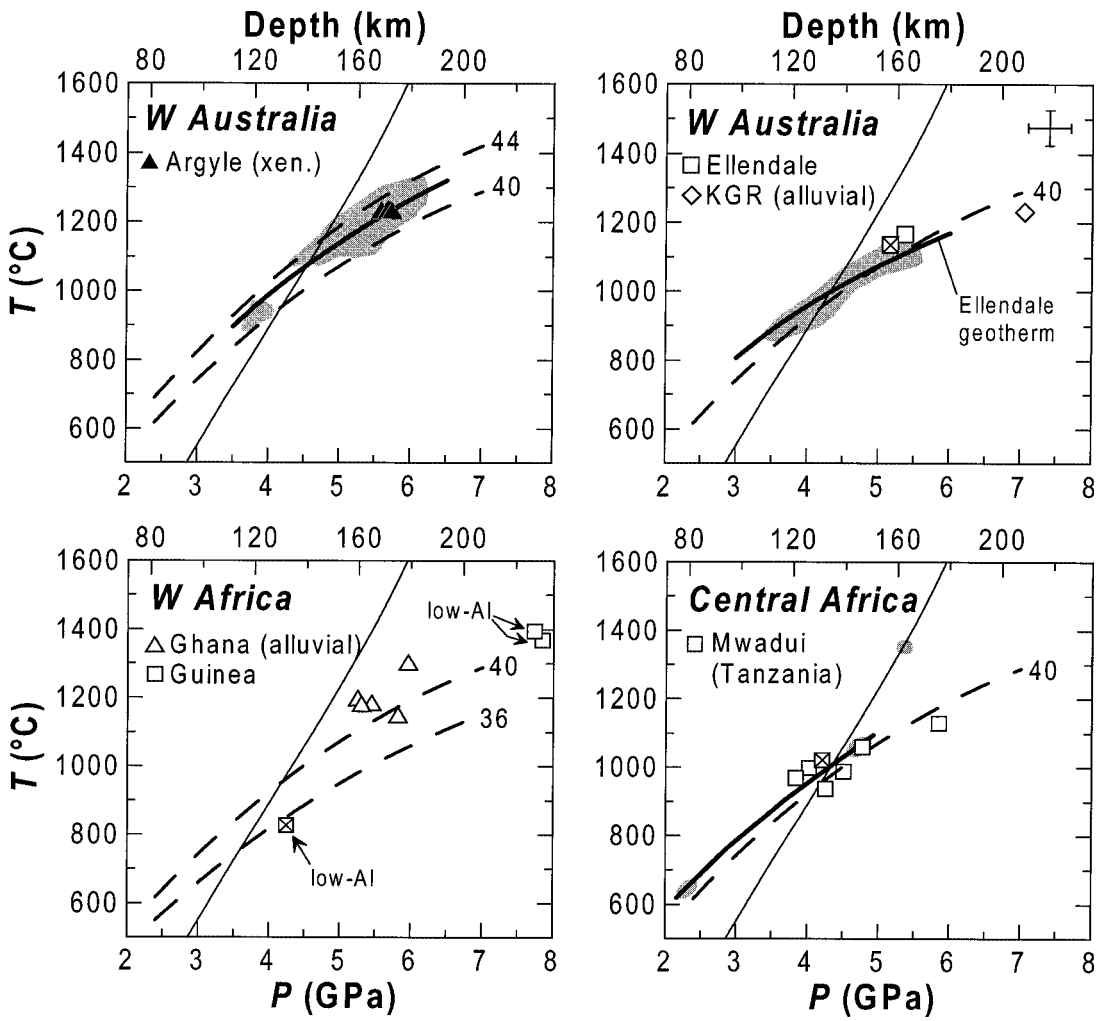


FIG. 4. *P–T* estimates for inclusions of chromian diopside in Siberian occurrences of diamond according to single-clinopyroxene thermobarometry. Symbols, fields and reference lines as in Figure 3. For the two cases of included clinopyroxene derived from a wehrlitic assemblage from Udachnaya, equilibrium with orthopyroxene is unlikely, and *T* estimates can be highly underestimated; the qualitative effect of *T* uncertainties on *P* estimates is represented with arrows. The geotherm relevant to coarsely crystalline xenoliths is based on data from Pokhilenko *et al.* (1991) and data for concentrates of coarse clinopyroxene (Taylor & Nimis, in prep.). The sources of data are listed in Table 1.

Al composition of one of the inclusions argues for possible derivation from garnet-free metasomatized peridotite, and consequent overestimation of pressure. If this is true, the apparent scatter in *P–T* estimates would not be significant. The hypotheses of secular cooling, thermal perturbation or disequilibrium growth may also account for a few sparse examples from Venetia and Kimberley pipes, which record a somewhat higher *T* compared with mantle xenoliths from the same host. The best evidence for an ancient, localized heating and metasomatic event, probably related to an episode of diamond formation, is provided by the suite of inclusions from Premier (Kaapvaal). This suite forms the largest available group of inclusions from an individual pipe and deserves further comment.

Inclusions at Premier, with one exception, are orthopyroxene-free. Most inclusions plot along a steady-state ~42 mWm^{-2} conductive geotherm, which is similar to or slightly colder than the local xenolith-based geotherm (calculated average difference in T = –56°C; Table 1), and extends to depths of ~200 km (Fig. 3a). Six of the thirty-two orthopyroxene-free inclusions, excluded from the above calculation, fall off the general trend and plot on the high-*T* side, suggesting a temperature increase of as much as 250°C at deep levels in the lithosphere. In the *P–T* plot, these high-*T* inclusions produce a tail that mimics that described by the high-*T* sheared xenoliths, which are usually interpreted as portions of the deep lithosphere that were heated and refertilized by rising melts shortly before kimberlite eruption (Smith *et al.* 1993). A relation between high-*T* inclusions and high-*T* xenoliths is challenged, however, by the fact that high-*T* inclusions at Premier are characteristically depleted in Ti, Al and Na (*cf.* LZCPX2 group



in Richardson *et al.* 1993). The lack of refertilization suggests that these high-*T* inclusions precipitated from or re-equilibrated with a hot, C-rich fluid, which could also be responsible for the thermal disturbance and diamond growth, rather than from a typical silicate melt (*cf.* Haggerty 1999). Interestingly, similar significant depletion in Na and Al, albeit without appreciable variation in Ti content, has been reported by Taylor *et al.* (2000) in clinopyroxene inclusions in an eclogitic diamond interpreted to have grown by input of C-rich fluid. Emplacement of the adjacent Bushveld complex at ~2.05 Ga, several hundred Ma before kimberlite eruption (~1.18 Ga), was also a possible source of significant thermal perturbation in the Premier area. The age of the Bushveld event is only 100 Ma older than Sm–Nd ages determined on inclusions in Premier diamonds, suggesting a link between this important event and diamond formation [Richardson *et al.* (1993); but see Navon (1999) for criticism of Sm–Nd dating of diamond inclusions]. The apparent lack of xenoliths recording conditions similar to those of the majority of the inclusions from the greatest apparent depths (*P* in the range 6.1–6.7 GPa, *T* in the range 1218–1265°C; Fig. 3a) implies that intense thermal perturbations, whatever their origin, have occurred in the lithosphere *after* encapsulation of these inclusions in their host diamond.

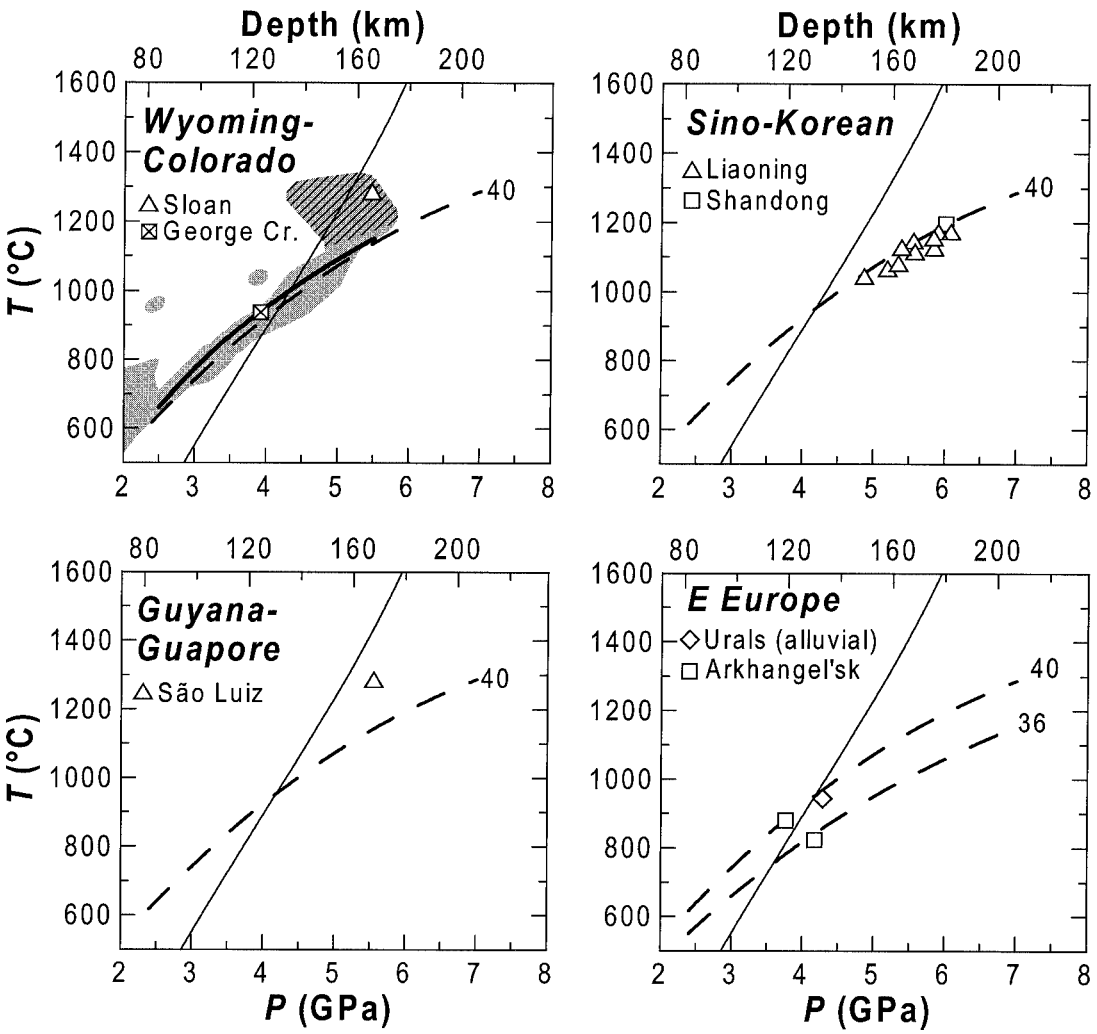


FIG. 5. *P–T* estimates for inclusions of chromian diopside in diamond from various cratons according to single-clinopyroxene thermobarometry. Symbols, fields and reference lines as in Figure 3. KGR: King George River. The sources of data are listed in Table 1.

With regards to the Siberian samples, most examples of diamond containing lherzolitic material appear to have formed near the graphite–diamond boundary, at depths less than 150 km (<5 GPa) (Fig. 4), in line with the abundance of lherzolitic material in this mantle section (Griffin *et al.* 1993, 1996, Boyd *et al.* 1997). A feature common to both the inclusions in diamond and the xenoliths is the scatter of thermobarometric data. The similarity of *P–T* patterns for inclusions in diamond, intergrowths with diamond and xenoliths may reflect lack of resolution of the thermobarometer (*ca.* ±50°C and ±0.3 GPa), but it is also compatible with hypotheses for a young origin of the diamond crystals, as proposed by Pearson *et al.* (1995), Shimizu & Sobolev (1995) and Shimizu *et al.* (1999). However, evidence also exists that many Siberian examples are ancient (Richardson & Harris 1997) and formed under conditions of progressively decreasing temperature (Sobolev & Efimova 1998). This inference suggests a link between diamond formation and one or more multistage thermal or metasomatic perturbations that have affected the Siberian mantle since *ca.* 3.2 Ga, the age of the oldest, coarsely crystalline peridotites (Pokhilenko *et al.* 1991), until shortly before eruption 350 Ma ago (*cf.* Griffin *et al.* 1993, 1996, Pearson *et al.* 1995, Shimizu *et al.* 1999). Expanding slightly the discussion given in the paper by Griffin *et al.* (1996), the scatter of xenolith and inclusion *P–T* values could be due to short-term, possibly repeated, heating–cooling events, which were probably at least in part connected to episodes of diamond formation. In this view, the existence of a few "cold" diamond inclusions from Mir plotting on the same geotherm as that defined by the 3.2-Ga-old coarsely crystalline xenoliths (Fig. 4) is consistent with an earlier proposal that diamond crystals up to 3.2 Ga old may be present in the coarsely crystalline Siberian peridotites not reworked during later thermal events (*cf.* Richardson & Harris 1997). Although it is still tenable that in some cases, diamond crystals may have formed as a consequence of late-stage melt- or fluid-assisted reactions, this hypothesis is unlikely to apply to these examples of very low-temperature diamond.

Conclusions

Most inclusions of lherzolitic origin in diamond record *P–T* conditions apparently within error of "normal", conductive thermal regimes in the mantle. Estimated *P–T* conditions are also commonly within error of those under which xenoliths from the same source last equilibrated (Table 1), apparently indicating that in several cases, diamond with inclusions of lherzolitic derivation formed when the lithospheric mantle had already attained a thermal regime comparable to or even colder than that extant at the time of emplacement of the host kimberlite or lamproite. Even where data on mantle xenoliths are not available, and particularly for the Eastern European, Western African and Sino-Korean cratons, *P–T* estimates for diamond inclusions remain broadly consistent with the relatively cool thermal regimes expected for typical cratonic lithospheres. The abundance of such "cold" inclusions indicates that either thermal conditions were often maintained near the conductive geotherm during the growth of diamond, perhaps following short-lived, localized, minor heating events, or most analyzed inclusions are protogenetic and record ambient conditions prior to the growth of diamond. The first hypothesis is supported by the "cold" signature of some unequivocally syngenetic inclusions described in Sobolev *et al.* (1976, 1989, 1997b) and by the overall prevalence of syngenetic inclusions in diamond worldwide (*e.g.*, Sobolev *et al.* 1989, Prinz *et al.* 1975).

Some high-*T* inclusions formed during ancient thermal or metasomatic events, which can be ascribed to advection of C-rich fluids from which the host diamond precipitated. Apparent differences in temperature between steady and thermally disturbed states can be as high as 250°C. Although measured differences may in part be artifacts due to potential disequilibrium growth of clinopyroxene, these results are qualitatively in line with independent thermometric data on some inclusions of garnet of harzburgitic derivation, which suggest formation of diamond during short-term heating events and temperatures fluctuations over up to *ca.* 400°C (Griffin *et al.* 1993) or *ca.* 200°C (Canil 1999), depending on the preferred calibration of the Ni-in-garnet thermometer. In a few cases, secular cooling of the cratonic lithosphere is believed to be a possible source of scatter in temperature estimates.

The distribution of diamond containing inclusions of a lherzolitic paragenesis in cratonic lithospheres seems to be closely related to the distribution of lherzolitic material in the mantle. Inferred depths of origin do not generally exhibit significant clustering at particular levels in the lithosphere and, where a statistically significant population of inclusions is available (*cf.* Premier, Kaapvaal), diamond with inclusions derived from such lherzolitic assemblages appears to have formed throughout the lithosphere within its stability field. Substantial gaps may locally occur (*cf.* Siberia) in close association with a scarcity of lherzolitic material in specific mantle sections. The hypothesis of a uniform distribution of diamond in lherzolitic portions of cratonic lithospheres, if confirmed on a large scale by a more extensive dataset on inclusions, can be extended to diamond containing all types of peridotitic inclusions, including the much more common harzburgitic ones.

The above results may have some implications on diamond-exploration strategies. Clinopyroxene thermobarometry has been proposed as a tool for assessment of diamond potential of areas affected by kimberlitic and lamproitic magmatism (Nimis & Taylor 2000, Taylor & Nimis, in prep.). The method is complementary to those based on other indicator minerals, such as garnet, chromite or ilmenite (*cf.* Gurney & Zweistra 1995, Grif-

fin & Ryan 1995), and can be applied to mantle-derived pyroxene recovered from stream sediments or direct sampling of kimberlite. Large proportions of chromian diopside crystals derived from well within the diamond window and showing no sign of thermal or metasomatic perturbations that could have led to resorption of diamond would constitute an ideal target. That most examples of diamond containing inclusions derived from a lherzolitic mantle formed when the mantle had attained a thermal regime comparable to that at the time of eruption ensures that thermobarometry of mantle-derived clinopyroxene is relevant to diamond exploration. A uniform distribution of diamond *and* lherzolite in the lithosphere should, however, be a basic prerequisite if diamond potential must be evaluated from clinopyroxene thermobarometry alone. Although the present study provides no evidence of the concentration of diamond at specific levels in the lithosphere, the possible heterogeneous distribution of lherzolitic material in some cratonic lithospheres should be taken into account.

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