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Kankan diamonds (Guinea) III: $\delta^{13}\text{C}$ and nitrogen characteristics of deep diamonds

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Abstract Diamonds from the Kankan area in Guinea formed over a large depth profile beginning within the cratonic mantle lithosphere and extending through the asthenosphere and transition zone into the lower mantle. The carbon isotopic composition, the concentration of nitrogen impurities and the nitrogen aggregation level of diamonds representing this entire depth range have been determined. Peridotitic and eclogitic diamonds of lithospheric origin from Kankan have carbon isotopic compositions ($\delta^{13}\text{C}$: peridotitic -5.4 to -2.2‰ ; eclogitic -19.7 to -0.7‰) and nitrogen characteristics (N: peridotitic 17–648 atomic ppm; eclogitic 0–1,313 atomic ppm; aggregation from IaA to IaB) which are generally typical for diamonds of these two suites worldwide. Geothermobarometry of peridotitic and eclogitic inclusion parageneses (worldwide sources) indicates that both suites formed under very similar conditions within the cratonic lithosphere, which is not consistent with a derivation of diamonds with light carbon isotopic composition from subducted organic matter within subducting oceanic

slabs. Diamonds containing majorite garnet inclusions fall to the isotopically heavy side ($\delta^{13}\text{C}$: -3.1‰ to $+0.9\text{‰}$) of the worldwide diamond population. Nitrogen contents are low (0–126 atomic ppm) and one of the two nitrogen-bearing diamonds shows such a low level of nitrogen aggregation (30% B-centre) that it cannot have been exposed to ambient temperatures of the transition zone ($\geq 1,400$ °C) for more than 0.2 Ma. This suggests rapid upward transport and formation of some Kankan diamonds pene-contemporaneous to Cretaceous kimberlite activity. Similar to these diamonds from the asthenosphere and the transition zone, lower mantle diamonds show a small shift towards isotopic heavy compositions (-6.6 to -0.5‰ , mode at -3.5‰). As already observed for other mines, the nitrogen contents of lower mantle diamonds were below detection (using FTIRS). The mutual shift of sublithospheric diamonds towards isotopic heavier compositions suggests a common carbon source, which may have inherited an isotopic heavy composition from a component consisting of subducted carbonates.

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Introduction

Alluvial deposits in the Kankan district of Guinea (West Africa) represent one of the very few occurrences worldwide of diamonds formed over a depth range exceeding the thickness of the lithosphere. From their syngenetic mineral inclusion content it may be concluded that these diamonds originated between about 150–700 km in the Earth's mantle (Stachel et al. 2000a, 2000b). This depth range includes the mantle lithosphere (down to about 200 km), the convecting upper mantle (about 200–410 km), the transition zone (410–660 km) and the uppermost lower mantle. The restriction in maximum depth to the uppermost lower mantle is entirely based on the low aluminium content of MgSi-perovskite inclusions (Stachel et al. 2000b), therefore some lower mantle diamonds containing inclusions, e.g. of ferropericlaite only, may be derived from even greater

depth. Based on the paragenetic types of inclusions and the P–T information derived from them, we can now extend our study to the diamonds themselves and examine, as a function of depth, variations in carbon isotopic composition, nitrogen content and the nitrogen aggregation level. The combination of all these data will allow us to put better constraints on the evolution of the different diamond sources and especially the possible role of crustal protoliths.

Samples and analytical methods

Ninety-one diamonds, which were all previously studied for their syngenetic inclusion content, form the sample for the present study. The diamonds ranged in size from 2 to 6 mm and were first characterised according to their shape, colour, resorption, plastic deformation features and their paragenesis. Fragments (weight of ~1 mg) of the diamonds were then analysed for their carbon isotopic composition using the method described in Deines et al. (1984). The instrumental measurement precision is $\pm 0.05\%$. The sample gases were compared with a working reference (Iceland spar, PSU2) whose isotopic composition with respect to the original PDB reference was measured by H. Craig and R.N. Clayton (personal communication, 1971). The carbon isotopic compositions are reported on the PDB scale used by Craig (1957).

Nitrogen concentrations and aggregation levels were determined on diamond cleavage chips by Fourier transform infrared spectroscopy (FTIRS) using a Nicolet, Magna IR 550 coupled with a Spectra-Tech IR-microscope. After conversion to absorption coefficients the spectra were de-convoluted into the A, B and D components (e.g. Boyd et al. 1995) using least square techniques. Nitrogen concentrations (atomic ppm) were calculated using the absorption strength at 1,282 wave-numbers for the A-centre (Boyd et al. 1994: 16.5 ± 1) and the B-centre (Boyd et al. 1995: 79.4 ± 8). Detection limits and errors strongly depend on the quality of the cleavage chip, but typically range between 10–20 ppm and about 10–20% of the concentration respectively.

Mantle sources derived from syngenetic mineral inclusions

The mineral inclusion content of the Kankan diamonds studied here is given in Table 1. The mineralogy and chemistry of these inclusions has been described in detail by Stachel et al. (2000a, 2000b) and only a short summary is given here. Peridotitic diamonds form an unusually small proportion of the diamonds studied and are strongly dominated by the lherzolitic suite. Despite this apparent fertility of the peridotitic source, pyrope garnets of lherzolitic paragenesis are in part very rich in Cr (Cr_2O_3 up to 17 wt%), thereby indicating an extensive melt depletion event. Several stages of subsequent metasomatic re-enrichment are reflected by the REE composition of pyrope garnet inclusions.

Eclogitic inclusions are derived not only from the cratonic lithosphere but also from the underlying convecting mantle (asthenosphere and transition zone). REE in garnet and cpx inclusions indicate that the lithospheric eclogitic source has $\text{LREE}_N/\text{HREE}_N < 1$ and REE_N patterns similar to those of subducted oceanic crust, which experienced partial melting after transformation into eclogite (Ireland et al. 1994; Rapp

and Shimizu 1998). In contrast, sublithospheric majoritic garnets and coexisting clinopyroxenes indicate a source that is strongly enriched in LREE. The occurrence of small negative Eu anomalies in both lithospheric and sublithospheric inclusions may indicate a link to ancient oceanic slabs for all eclogitic Kankan diamonds. Such a link does not necessarily involve diamond formation directly within former oceanic crust, but could also be established by ascending plumes entraining such material at the core mantle boundary or close to the 660 km discontinuity.

Diamonds from the lower mantle are the dominant paragenesis at Kankan and their inclusion chemistry suggests a chemically heterogeneous source at the top of the lower mantle, which is strongly but variably enriched in incompatible elements and is comprised of both positive and negative Eu anomalies. These chemical characteristics may be taken as circumstantial evidence for a source containing recycled crustal material.

Physical and chemical diamond data

Physical characteristics

The Kankan diamonds examined were specifically selected for their inclusion content and are not necessarily representative of the total production with respect to their physical characteristics. The shape of Kankan diamonds is strongly dominated by dodecahedral resorption forms. Octahedral shapes are common only for diamonds of the peridotitic suite, where 6 out of 13 diamonds showed complete or residual octahedral morphology. Among diamonds with inclusions from the lower mantle only 3 out of 36 still showed relics of primary octahedral faces. Twins (macles) and aggregates only account for 3 and 4% of the diamond sample, respectively, and are restricted to lithospheric (peridotitic and eclogitic) parageneses. About 10% of the diamonds have irregular shapes, either because of fracturing or deep etching. More regular etching in the form of trigons or hexagons occurs but is even less frequent. Plastic deformation lines occur occasionally (7%) and are restricted to lower mantle and peridotitic diamonds. Brown body colours, which also indicate plastic deformation, are more common (16%) and occur for all parageneses except the peridotitic suite.

Carbon isotopic composition

The mode in $\delta^{13}\text{C}$ of Kankan diamonds (-3.5% , Fig. 1) is shifted towards isotopic heavier compositions by about 1% relative to diamonds from worldwide sources (-4.6% , Galimov 1991). From Fig. 1 it is apparent that this shift in mode is caused by sublithospheric diamonds only. The peridotitic Kankan diamonds form a narrow normal distribution between -5.4 and -2.2% , with a mode of -4.5% (class -5 to -4%). Eclogitic diamonds

Table 1 Physical and chemical characteristics of Kankan diamonds. Shape: *o* octahedral, *d* dodecahedral, *m* macle, *i* irregular, *a* aggregate. Colour: *c* colourless, *b* brownish. Plastic deformation (*p.d.*) *y* present, *n* absent. Inclusions: *amph* amphibole, *cpx* clino-

pyroxene, *chr* chromite, *Fe-per* ferropericlasite, *grt* garnet, *ol* olivine, *opx* orthopyroxene, inclusions connected by a hyphen form a touching pair

Sample	Shape	Colour	p.d.	$\delta^{13}\text{C}$	Type	<i>n</i> (ppm)	%B	Paragenesis	Inclusions
KK-1	i	b	n	0.89	II	0	0	Majoritic	grt, cpx
KK-2	d/m	c	y	-3.23	II	0	0	Unknown	Altered
KK-3	d/m	c	n	-4.91	IaAB	711	27	Eclogitic	cpx
KK-4	d	b	n	-0.55	IaB	260	96	Eclogitic	grt
KK-5	i/d	c	n	-0.91	II	0	0	Majoritic	grt
KK-6	d	c	n	-4.31	II	0	0	Unknown	
KK-7	d	c	n	-3.91	II	0	0	Lower mantle	Fe-per
KK-8	i/d	c	n	-4.29	II	0	0	Unknown	
KK-9	d	b	n	-0.49	II	0	0	Lower mantle	CaSiO ₃
KK-10	d	c	n	-4.03	II	0	0	Unknown	
KK-11	d	b	n	-3.96	II	0	0	Lower mantle	Fe-per
KK-12	d	c	n	-2.95	IaB	32	97	Majoritic	grt
KK-13	d	c	n	-3.54	II	0	0	Lower mantle	2 Fe-per
KK-14	d	b	y	-3.48	II	0	0	Unknown	Altered
KK-16	d	c	n	-2.57	II	0	0	Lower mantle	2 Fe-per, MgSiO ₃ , siderite
KK-18	a	c	n	-16.25	IaAB	375	26	Eclogitic	2 grt
KK-19	d	b	n	1.05	II	0	0	Unknown	
KK-20	d	c	n	-0.22	II	0	0	Unknown	Altered
KK-21	o/d	c	n	-5.05	IaAB	288	55	Peridotitic	2 ol
KK-22	o/d	c	n	-5.20	IaAB	314	34	Unknown	
KK-23	d	c	n	-4.73	II	0	0	Lower mantle	Fe-per
KK-24	d	c	n	-3.40	II	0	0	Eclogitic	grt
KK-25	d	c	n	-5.23	IaA	874	0	Unknown	
KK-26	d	c	n	-4.50	IaAB	159	48	Peridotitic	grt, 2 ol
KK-28	d/m	c	y	-4.96	IaAB	183	53	Peridotitic	grt, cpx, 2 ol
KK-29	d	c	n	-3.71	II	0	0	Lower mantle	Fe-per
KK-30	d	c	n	-3.52	II	0	0	Lower mantle	Fe-per
KK-31	d	c	n	-4.42	II	0	0	Lower mantle	Fe-per, SiO ₂
KK-32	d	c	n	-3.74	II	0	0	Lower mantle	Ca-silicates
KK-35	d	c	n	-4.38	II	0	0	Lower mantle	Fe-per
KK-36	d	b	n	-3.98	II	0	0	Lower mantle	2 Fe-per
KK-37	d/i	c	n	-3.51	II	0	0	Lower mantle	Fe-per
KK-38	d	b	y	-3.72	II	0	0	Lower mantle	Fe-per
KK-39	d/i	c	n	-1.26	II	0	0	Lower mantle	Fe-per
KK-40	m	c	n	-5.29	IaAB	757	31	Eclogitic	cpx
KK-41	o	c	n	-9.03	IaA	557	9	Unknown	Graphite
KK-42	o	c	n	-5.11	IaA	556	8	Eclogitic	3cpx
KK-43	d	c	n	-3.91	II	0	0	Lower mantle	Fe-per
KK-44	d	c	n	-4.10	II	0	0	Lower mantle	Fe-per, MgSiO ₃ , CaSiO ₃ , MgSiO ₃ -ol
KK-45	d	c	y	-3.50	II	0	0	Lower mantle	opx, K-fsp (hollandite?)
KK-46	o	c	n	-3.01	IaB	264	98	Unknown	Altered
KK-47	d	c	n	-4.68	IaA	1,122	100	Unknown	sulph
KK-48	d	c	n	-4.04	II	0	0	Lower mantle	Fe-per
KK-49	m	c	y	-5.42	IaAB	463	47	Peridotitic	ol
KK-50	o	c	n	-3.34	IaB	96	91	Unknown	
KK-51	d	c	n	-1.48	II	0	0	Unknown	3 altered
KK-52	o	c	n	-4.30	IaAB	747	11	Unknown	
KK-53	o/d	c	n	-3.39	IaAB	49	29	Unknown	
KK-54	d	c	n	-3.67	II	0	0	Lower mantle	Fe-per
KK-56	o	c	n	-3.65	IaAB	231	49	Unknown	
KK-57	d	b	n	-4.16	II	0	0	Lower Mantle	Fe-per
KK-59	d	c	n	-1.45	II	0	0	Unknown	Altered
KK-61	o/d	c	n	-0.35	II	0	0	Majoritic	2 grt, altered
KK-62	d/a	c	n	-3.55	II	0	0	Unknown	2 altered
KK-63	o/d	b	n	-3.56	II	0	0	Lower Mantle	Fe-per
KK-65	d/o	c	n	-2.21	IaAB	212	36	Peridotitic	grt, cpx-opx, 2 ol
KK-66	d	c	n	-5.79	II	0	0	Lower Mantle	2 Fe-per, 2 CaSiO ₃
KK-67	m	c	n	-9.16	IaAB	1,313	13	Eclogitic	Coesite
KK-68	i	b/c	n	-19.74	IaA	1,229	0	Eclogitic	2 rutile
KK-70	d/o	c	n	-4.58	IaAB	100	15	Peridotitic	2 grt, 2 chr
KK-71	d	c	n	-5.04	II	0	0	Lower mantle	Fe-per
KK-73	o	c	n	-4.99	IaAB	50	46	Peridotitic	2 ol
KK-74	d	b	n	-6.55	II	0	0	Lower mantle	Fe-per

Table 1 (Contd.)

KK-75	d	c	n	-8.50	IaAB	466	28	Eclogitic	cpx
KK-76	i	c	n	-15.81	IaAB	240	20	Unknown	
KK-77	o/d	b	n	-2.36	IaAB	88	11	Eclogitic	2 cpx, altered
KK-78	twin	c	n	-3.61	IaAB	17	35	Peridotitic	grt, 2 ol
KK-79	i/d	c	n	-4.86	IaB	40	100	Peridotitic	cpx
KK-80	i/d	c	n	-4.85	IaAB	923	54	Eclogitic	2 cpx
KK-81	i/d	c	n	-3.13	IaAB	126	30	Majoritic	grt, cpx
KK-82	d	b	n	-2.62	II	0	0	Lower mantle	Fe-per
KK-83	d	c	n	-3.26	II	0	0	Lower mantle	Fe-per, 3 cpx, 'ol'-Fe
KK-84	i	c	n	-3.87	II	0	0	Unknown	Fe-per, ol, Fe-per-ol
KK-86	d/a	c	n	-4.96	IaAB	819	29	Eclogitic	grt
KK-87	d	c	n	-4.73	II	0	0	Lower mantle	Fe-per, CaSiO ₃
KK-88	o/d	c	n	-3.66	II	0	0	Lower mantle	Fe-per
KK-89	d	c	n	-3.90	II	0	0	Lower mantle	Fe-per
KK-90	o	c	n	-4.97	IaA	176	1	Peridotitic	chr, altered
KK-91	i/d	c	n	-4.52	IaAB	74	15	Peridotitic	ol
KK-92	d	c	n	-3.19	IaAB	107	34	Peridotitic	2 ol
KK-93	o	c	n	-4.73	IaAB	648	13	Peridotitic	chr
KK-94	d	c	n	-3.87	II	0	0	Lower mantle	2 Fe-per
KK-95	d	c	n	-4.09	II	0	0	Lower mantle	2 Fe-per
KK-96	o/d	c	n	-5.37	IaAB	942	28	Eclogitic	grt, cpx, altered
KK-97	d	c	n	-1.54	II	0	0	Eclogitic	grt
KK-98	d	c	n	-16.03	II	0	0	Unknown	
KK-101	d	c	n	-5.78	IaAB	57	26	Eclogitic	grt
KK-103	d	c	n	-3.88	II	0	0	Lower mantle	Fe-per, MgSiO ₃
KK-104	d	b	n	-4.07	II	0	0	Lower mantle	Fe-per
KK-108	d	c	n	-3.58	II	0	0	Lower mantle	2 Fe-per, 2 MgSiO ₃ , amph
KK-109	o	c	n	-3.78	II	0	0	Lower mantle	2 Fe-per, Fe-per-ol

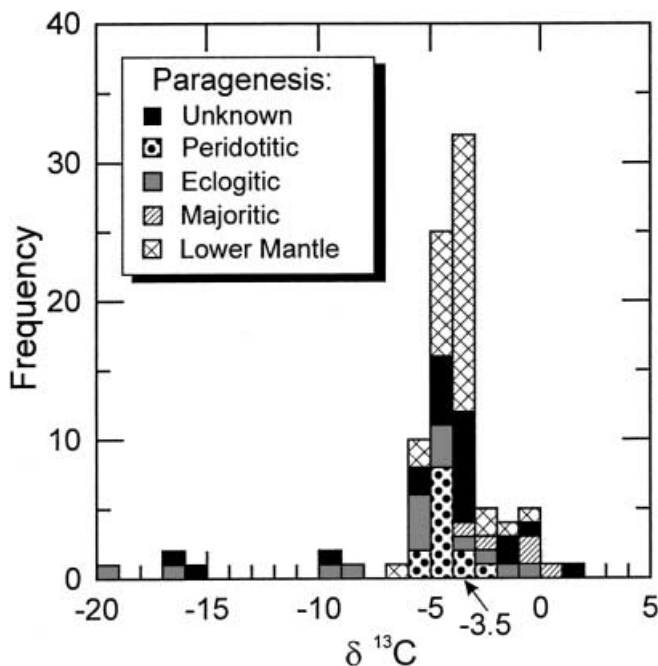


Fig. 1 Carbon isotopic composition ($\delta^{13}\text{C}$) of diamonds from Kankan (of all parageneses). Class size is 1‰; therefore, the mode for Kankan diamonds at -3.5‰ corresponds to an isotopic range of -4 to -3‰ .

show a large spread from -19.7 to -0.6 with a poorly defined mode at -5.5‰ . A much larger isotopic spread of eclogitic diamonds relative to peridotitic diamonds was first recognised by Sobolev et al. (1979) and is now

well established (reviewed in Galimov 1991; Kirkley et al. 1991).

The isotopic composition of sublithospheric diamonds, comprising those with majorite garnet (derived from the asthenosphere and the transition zone) and lower mantle inclusions both extend to unusually heavy values. The five diamonds of majoritic paragenesis (Fig. 2) range between -3.1 to $+0.9\text{‰}$ (average -1.3‰), the more frequent lower mantle diamonds ($n=36$) form a normal distribution between -6.6 to -0.5‰ (Fig. 3).

Nitrogen contents and aggregation levels

Lithospheric Kankan diamonds have a large spread in nitrogen concentrations, with some eclogitic diamonds showing rather high concentrations (from below detection up to 1,313 atomic ppm) compared with peridotitic samples (17–648 atomic ppm, Fig. 4). The small group of diamonds with majorite garnet inclusions is generally low in nitrogen (below detection to 126 atomic ppm) and all lower mantle diamonds are Type II (i.e. nitrogen below detection). Absence of detectable nitrogen impurities has already been established as a characteristic of lower mantle diamonds by Harte and Harris (1994), Hutchinson et al. (1999) and Davies et al. (1999).

Nitrogen aggregation levels (Fig. 5) are very variable and range from pure IaA to pure IaB, i.e. a full range is observed with all nitrogen in the A-centre to all nitrogen in the higher aggregated B-centre. The dependence of nitrogen aggregation on mantle residence time and ambient temperature was examined by Evans and Harris

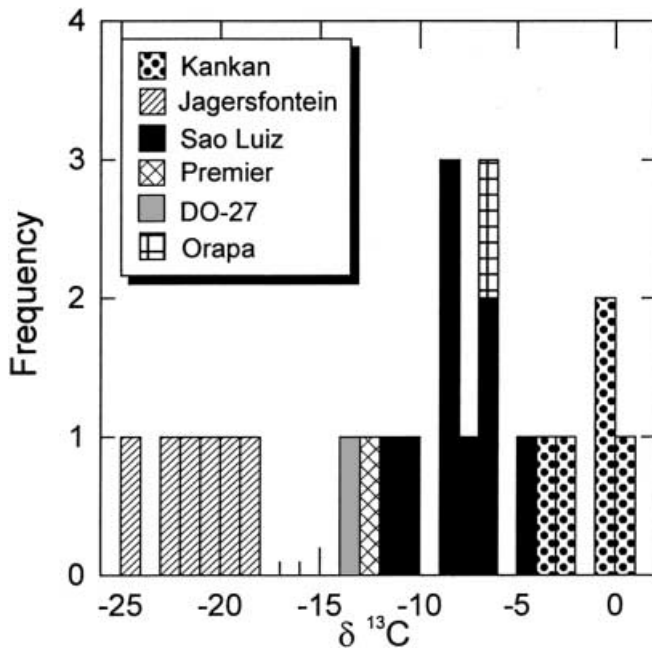


Fig. 2 Carbon isotopic composition ($\delta^{13}\text{C}$) of diamonds containing majorite garnet inclusions. Data sources: Jagersfontein, Deines et al. (1991); Sao Luiz, Wilding (1990) and Hutchinson et al. (1999); Premier, Deines et al. (1984); DO-27, Davies et al. (1999); Orapa, Deines et al. (1993). Kankan diamonds from the asthenosphere and transition have an average value of -1.3‰ ($\pm 1.7\text{‰}$, $n=5$) whereas the average for worldwide sources is considerably shifted towards isotopically lighter compositions (average $-12.9 \pm 6.4\text{‰}$, $n=18$)

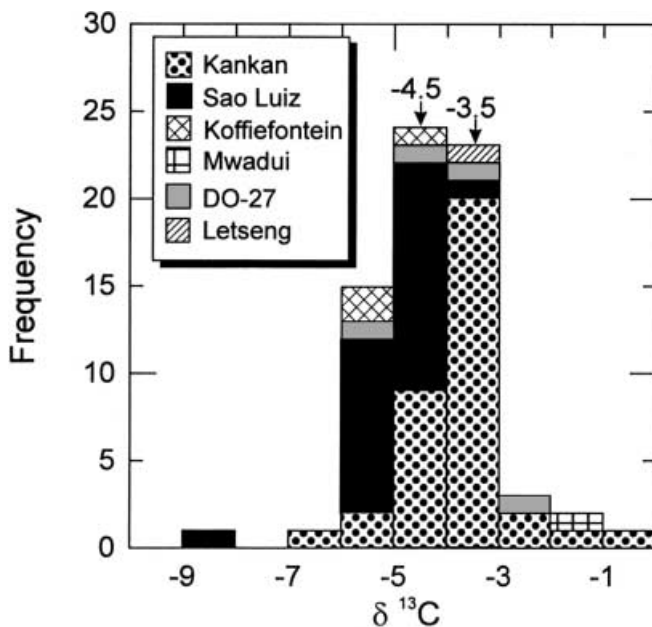


Fig. 3 Carbon isotopic composition ($\delta^{13}\text{C}$) of diamonds from the lower mantle. Data sources: Sao Luiz, Wilding (1990), Hutchinson (1997) and Hutchinson et al. (1999); Koffiefontein, Deines et al. (1991); Mwadui, own unpublished data; DO-27, Davies et al. (1999); Letseng-la-Terrai, McDade and Harris (1999). Kankan shows a mode at -3.5‰ whereas lower mantle diamonds from worldwide sources peak at -4.5‰ . The same shift is also apparent for the statistical average (Kankan: $-3.8 \pm 1.0\text{‰}$, $n=36$, worldwide sources: $-4.8 \pm 1.1\text{‰}$, $n=34$)

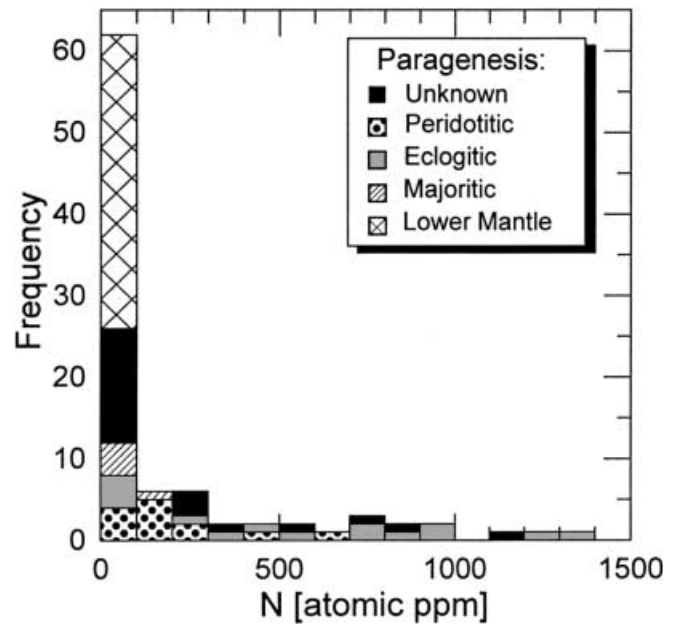


Fig. 4 Nitrogen concentrations (atomic ppm) of Kankan diamonds. The class 0–100 atomic ppm includes all the analyses with nitrogen concentrations below detection

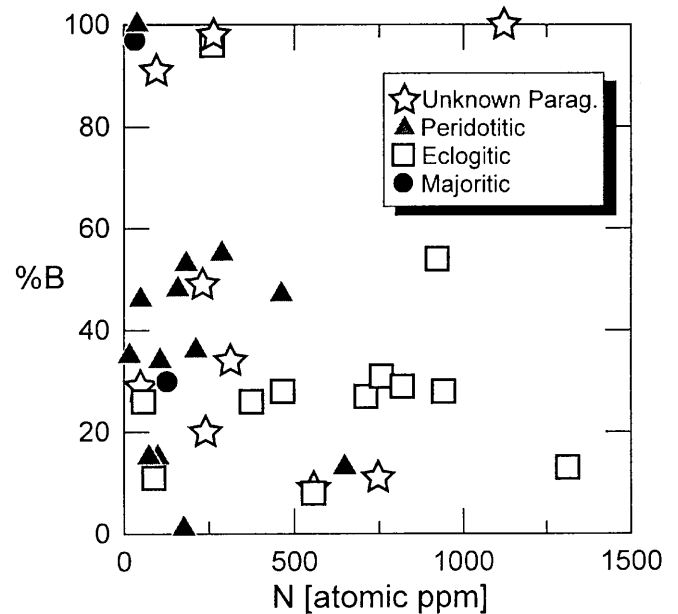


Fig. 5 Nitrogen aggregation level (relative proportion of the highly aggregated B-centre) versus total nitrogen concentration (atomic ppm). Diamonds with nitrogen below detection are excluded

(1989) and Taylor et al. (1990), but turned out to be of limited use as a thermo-chronometer because of the unquantified effects of plastic deformation. Nevertheless, from the low nitrogen aggregation levels of eclogitic diamonds rich in nitrogen relatively low mantle temperatures ($< 1,100\text{ °C}$ at 2 Ga mantle residence time) or, alternatively, mantle residence times of less than 2 Ga may be deduced. For diamond KK-96, inclusions

of eclogitic garnet and cpx indicate an equilibration temperature of 1,240 °C (Mg–Fe exchange thermometer of Krogh 1988), corresponding to a mantle residence time of this diamond of less than 5 Ma (applying the activation energy and Arrhenius constant for the transition from the A- to the B-centre obtained by Taylor et al. 1990). If the high equilibration temperature is not assumed to reflect disequilibrium between the two non-touching inclusions then significantly longer residence times may only be achieved by rapid cooling immediately after diamond formation.

A surprising observation is the low nitrogen aggregation level (30% B-centre at 126 atomic ppm N) of an asthenospheric diamond containing majorite garnet with 6.56 Si atoms per formula unit (suggesting formation in the upper part of the transition zone). Along an adiabatic mantle geotherm temperatures at that depth should exceed 1,400 °C, thus causing rapid aggregation towards the B-centre. The absence of such aggregation either implies a short mantle residence for this diamond (≤ 0.2 Ma), or formation and storage in an environment much cooler than 1,400 °C.

Figure 6 shows the relationship between total nitrogen content and carbon isotopic composition: the data plot into a triangle with nitrogen-free diamonds occurring over the entire range from -24 to $+1$ ‰ and high nitrogen contents being limited to compositions close to -5 ‰. The left hand side of this relationship, i.e. decreasing maximum nitrogen contents with decreasing $\delta^{13}\text{C}$, has already been recognised previously (Stachel and Harris 1997; Cartigny et al. 2001b), but from our data it also appears that there is a steady decrease in nitrogen content as the carbon isotopic value becomes

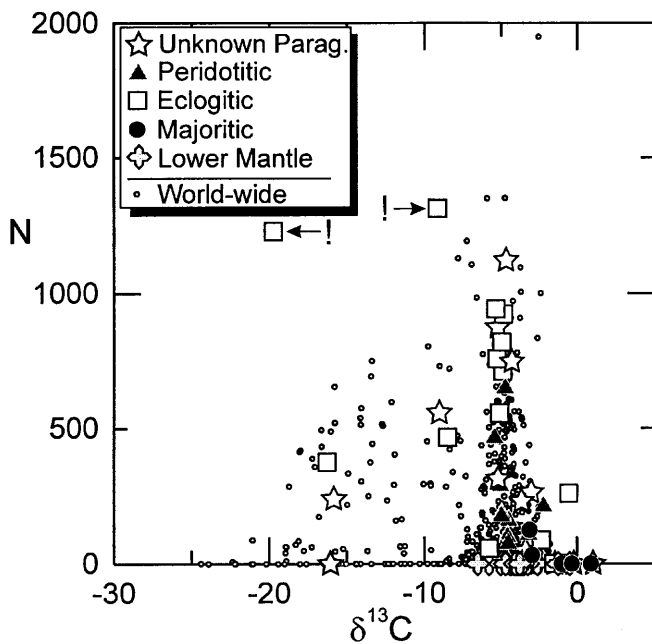


Fig. 6 Nitrogen concentration (atomic ppm) versus $\delta^{13}\text{C}$ (‰) for diamonds from Kankan (divided according to their inclusion parageneses) and from worldwide sources

progressively heavy, leading to diamonds with $\delta^{13}\text{C} > -0.5$ ‰ being Type II. Two eclogitic diamonds from Kankan with high nitrogen contents (1,229 and 1,313 atomic ppm) plot outside this triangle, indicating that these diamonds may also have formed in unusual circumstances. However, FTIRS (N-contents) and combustion mass spectrometry ($\delta^{13}\text{C}$) were done on separate diamond chips (as it is the case for almost all the samples in the worldwide database shown in Fig. 5) and thus a very inhomogeneous distribution of both nitrogen abundances and carbon isotopes may be an alternative explanation for this observation.

Correlation between inclusion chemistry and diamond characteristics

A number of studies (reviewed by Harris 1987; Galimov 1991; Kirkley et al. 1991; Hutchinson et al. 1999) have shown that the carbon isotopic composition and the nitrogen content of diamonds have certain characteristic relationships to the inclusion paragenesis. Examples are light carbon isotopic values, which are almost exclusively restricted to diamonds of the eclogitic suite or the Type II composition ($\text{N} < 10$ atomic ppm) of lower mantle diamonds. However, correlations between the chemical composition of distinct inclusion species and diamond characteristics have not been firmly established so far. With one possible exception this conclusion is also confirmed by the present study: Fig. 7 shows that

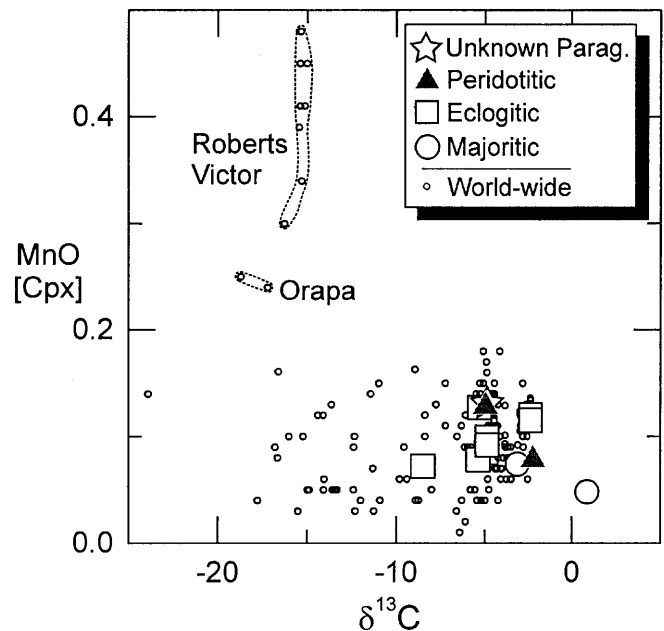


Fig. 7 MnO content of clinopyroxene inclusions versus carbon isotopic composition of their host diamonds from Kankan (divided according to their inclusion parageneses) and from worldwide sources. High manganese contents appear to be limited to eclogitic inclusions from Roberts Victor (Deines et al. 1987) and eclogitic and websteritic inclusions from Orapa (Deines et al. 1993) contained in diamonds with very light isotopic composition

clinopyroxene with $\text{MnO} > 0.2$ wt% is restricted to diamonds with $\delta^{13}\text{C} \leq -15\text{‰}$ occurring at Orapa and Jwaneng. Apparently only eclogitic (and websteritic) diamonds with very light isotopic values form in Mn enriched environments.

Discussion

Lithospheric diamonds

Cartigny et al. (2001b) suggest that the limiting sector observed in diagrams of nitrogen content versus carbon isotopic composition of diamonds (i.e. the left hand side of the triangular distribution apparent from Fig. 6), reflects the evolution of melts or fluids through fractionation by separation of a CO_2 -rich fluid species. In a peridotitic mantle, at the high pressures within the diamond stability field, CO_2 is buffered by olivine and opx, and, in consequence, a free CO_2 -fluid cannot exist. In an eclogitic environment, CO_2 is buffered by cpx at low temperatures, but decarbonation occurs at high temperatures (Luth 1993). In a system with $x(\text{CO}_2) = 1$ this reaction curve cuts the diamond stability curve at 1,400 °C and 5.5 GPa, which does not correspond to the position of realistic geothermal gradients. For CO_2 - H_2O mixtures decarbonation may be expected to occur at lower temperatures (Knoche et al. 1999), possibly in accordance with the temperature of formation of eclogitic diamonds (around 1,200 °C, see below). Introduction of such hydrous melts/fluids would induce partial melting of eclogites (see Rapp and Watson 1995), which may also provide an explanation for the LREE depleted character frequently observed for eclogitic inclusions in diamonds (Ireland et al. 1994; Taylor et al. 1996; Aulbach et al. 2001). However, oxygen fugacity in the upper mantle decreases with depth because of an increasing modal proportion of minerals which readily accommodate ferric iron (O'Neill et al. 1993; Ballhaus 1995; Wood et al. 1996) and, therefore, the prerequisite of the model of Cartigny et al. (2001b) that melts derived from the asthenosphere and transition zone contain C-H-O as oxidised species (i.e. H_2O and CO_2) is not unequivocal.

If the high nitrogen and low $\delta^{13}\text{C}$ signature of the two eclogitic diamonds that plot above the limiting sector in Fig. 6 is not caused by inhomogeneity (see before), then these diamonds imply that their light carbon isotopic composition is not caused by a fractionation process as envisaged by Cartigny et al. (2001b). Compositionally these diamonds are intermediate between 'normal' eclogitic diamonds and diamonds from ultra high pressure metamorphic rocks, which typically have light carbon (-10 to -16‰) and high N ($> 2,300$ ppm; Cartigny et al. 2001a, 2001b) and this signature may indicate crustal protoliths. Formation and storage in a 'cold' crustal protolith would also be in accordance with the very low nitrogen aggregation level (0 and 13% B-centre) of these two diamonds. A subduction origin for diamonds with light carbon isotopic signatures has previously been

proposed by a number of authors (e.g. Frank 1969; Kirkley et al. 1991 and references therein) and was mainly based on the similarity in $\delta^{13}\text{C}$ to organic matter.

Because of their 'cold' nature, subducting slabs enter the diamond stability field at a shallower depth than expected for cratonic regions where the temperature distribution follows a conductive geothermal gradient. For a typical slab (e.g. Green and Falloon 1998) conversion of graphite to diamond should occur at about 800 °C and 4 GPa. Only temperatures can be estimated from the inclusion parageneses contained in eclogitic diamonds and, therefore, pressures for the calculations have to be assumed. Using the pressure of 4 GPa expected for diamond formation in a subducting slab, the garnet-cpx thermometer of Krogh (1988) gives a mode in the temperature distribution at 1,100 °C (worldwide data), thus resulting in conditions outside the diamond stability field. This clearly suggests that eclogitic diamonds are not generally formed from organic matter within subducting slabs.

The alternative model for the genesis of eclogitic diamonds is a formation similar to peridotitic diamonds, which occurs within the cratonic lithosphere through redox reactions. In this case, the temperature distribution of eclogitic and peridotitic diamonds should be similar. For peridotitic diamonds, pressures and temperatures may be estimated simultaneously through a combination of the garnet-opx geothermobarometers of

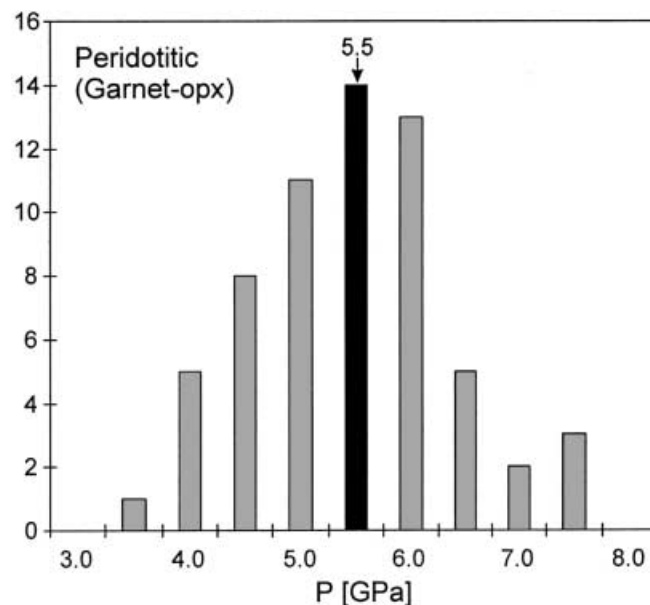


Fig. 8 Pressures of formation of peridotitic diamonds (worldwide sources) derived from iterative P-T calculations using the garnet-opx geothermobarometers of Harley (1984) and Brey et al. (1990). Class size is 0.5 GPa, a mode of 5.5 GPa therefore corresponds to a range of 5.25–5.75 GPa. Calculations are based on inclusions from 63 diamonds (results for multiple inclusion pairs are averaged), one diamond from Venetia falls outside the range shown ($P = 9.0$ GPa). References for the data base of inclusions from worldwide sources used for the calculations are given in Stachel and Harris (1997) and Stachel et al. (2000a)

Brey and Köhler (1990) and Harley (1984). This results in a fairly well-defined normal distribution with a mode at 5.5 GPa (Fig. 8, worldwide data set). This pressure of 5.5 GPa can be used to calculate temperatures for both peridotitic (using the common paragenesis garnet–olivine, O'Neill and Wood 1979; O'Neill 1980) and eclogitic diamonds (from the paragenesis garnet–cpx, Krogh 1988). Identical modes of 1,200 °C (class 1,175–1,225 °C, Figs. 9 and 10) and very similar averages and medians (peridotitic: 1,170 and 1,180 °C respectively; eclogitic: both 1,220 °C) indicate perfect agreement of the two suites, especially in view of the application of two different geothermometers. The concurrence in conditions of formation of eclogitic and peridotitic diamonds strongly supports formation of both suites within the subcratonic lithospheric mantle. This does not exclude a subduction origin of diamondiferous eclogites but requires that for the majority of cases, diamonds formed in an event separate from such possible subduction scenarios. This is consistent with the observation that $\delta^{15}\text{N}$ of eclogitic diamonds strongly supports a formation from mantle-derived fluids or melts (Cartigny et al. 1998). In addition to the before mentioned fractionation model of Cartigny et al. (1998, 2001b) the large spread in $\delta^{13}\text{C}$ of eclogitic diamonds may be attributed to a number of possible scenarios:

1. Melts or fluids from which eclogitic diamonds form may have a slab-derived component. However, it does not appear likely that such a slab-derived com-

ponent should only affect the eclogitic portions of the subcratonic lithospheric mantle to which isotopically light diamonds are limited.

2. Galimov (1991) proposed a model where lithospheric diamonds precipitate when reduced 'sub-asthenospheric' fluids interact with relatively oxidised lithosphere. Similar to the redox melting model proposed by Taylor and Green (1989), this redox process is expected to suppress the solidus of peridotite sufficiently so that partial melting occurs. In particularly oxidised portions of the lithosphere, CH_4 may partially be converted to CO_2 , and Galimov (1991) suggests that partial melting of such carbonated sources leads to the formation of basic melts from which eclogitic diamonds and their inclusions may precipitate. The large spread in carbon isotopic composition of eclogitic diamonds could thus be related to isotopic exchange in the system CO_2 – CH_4 , combined with Rayleigh distillation. However, the underlying assumption that partial melting of a carbonated peridotite at high pressures yields a magma of basaltic composition is not viable as carbonated peridotite is usually considered to yield carbonatites, melilitites and kimberlites (Brey 1978; Wyllie 1989; Gurnis et al. 1995; Dalton and Presnall 1998), i.e. carbonate is not a refractory phase, but strongly contributes to the early formed melts thereby lowering instead of rising its silica content. However, the fractionation mechanism proposed by Galimov (1991) is also applicable if pre-existing eclogite is assumed.

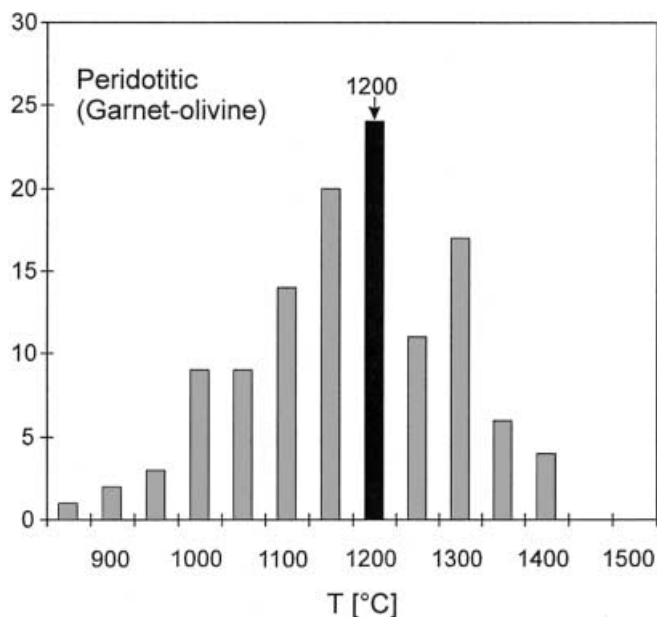


Fig. 9 Temperature of formation of peridotitic diamonds calculated from Mg–Fe exchange between garnet and olivine inclusions (O'Neill and Wood 1979; O'Neill 1980). A value of 5.5 GPa (derived from Fig. 8) was used for the pressure correction. Class size is 50 °C, a mode of 1,200 °C therefore corresponds to a range of 1,175–1,225 °C. Results are based on inclusions from 120 diamonds (results for multiple inclusion pairs are averaged), all calculated temperatures fall into the range shown

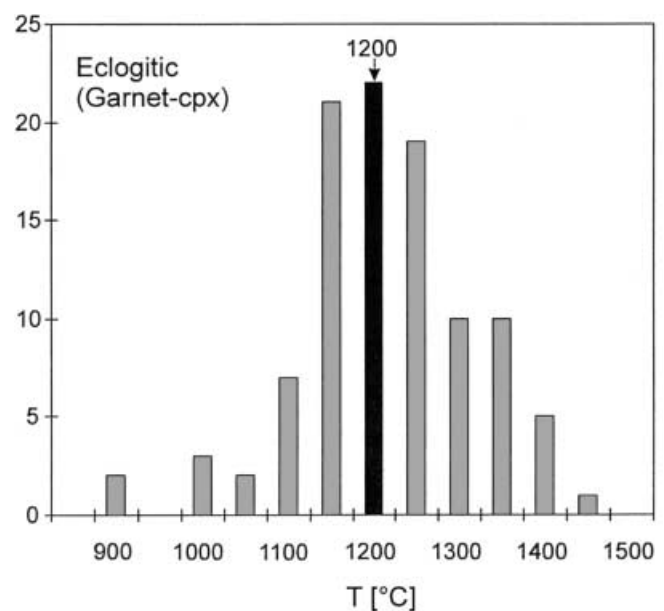


Fig. 10 Temperature of formation of eclogitic diamonds calculated from Mg–Fe exchange between garnet and cpx inclusions (Krogh 1988). A value of 5.5 GPa (derived from Fig. 8) was used for the pressure correction. Class size is 50 °C, a mode of 1,200 °C therefore corresponds to a range of 1,175–1,225 °C. Results are based on inclusions from 104 diamonds (results for multiple inclusion pairs are averaged, majorite garnet is excluded), one diamond from Argyle falls outside the range shown ($T = 1,645$ °C)

3. Galimov et al. (1989), in a study of kimberlitic websterite xenoliths, found significant contents (up to 0.09 wt%) of isotopically light carbon ($\delta^{13}\text{C}$ of -24 to -26‰) in silicate phases. Galimov et al. (1989) suggest that isotopic fractionation could be caused by the carbon in the silicates being present mostly as solitary, unbonded atoms, which, on exchange with isotopically bonded carbon, leads to significant enrichment of ^{12}C in the unbonded form. Similarly, Deines (2001b) discusses large differences (20‰) in $\delta^{13}\text{C}$ between diamond and moissanite from the same kimberlite. This suggests that thermodynamic or kinetic effects may produce significant variations in $\delta^{13}\text{C}$ (Galimov et al. 1989; Deines 2001b).

Sublithospheric diamonds

Figure 2 indicates that from the present database no clear mode in $\delta^{13}\text{C}$ can be determined for 'majoritic' diamonds. Most of the available data come from three mines, Jagersfontein (Deines et al. 1991), Sao Luiz (Wilding 1990; Hutchinson 1997; Hutchinson et al. 1999) and Kankan, which show significant differences in their isotopic composition. Compared with the mode of peridotitic diamonds at about -4.6‰ , Sao Luiz (range -11.0 to -4.6‰) and Jagersfontein (range -20.2 to -24.4‰) show a shift towards isotopically light compositions, whereas Kankan has isotopically heavy compositions (-3.1 to $+0.9\text{‰}$). In addition to the arguments used above, the isotopically light compositions of diamonds containing sublithospheric inclusions cannot be satisfactory related to a subduction scenario as it seems unreasonable to expect that organic carbon could be preserved to such depth to convert to diamond. Fractionation scenarios involving CO_2 (e.g. Cartigny et al. 1998) also are not viable as CO_2 would be buffered even by eclogitic parageneses at such great depth. Thus, thermodynamic isotope effects (see above), as suggested by Galimov et al. (1989) and Deines (2001a, 2001b), appear to be the only plausible mechanism for the observed enrichments in ^{12}C .

However, the isotopically heavy values for the diamonds from Kankan, which contain majorite garnets, can be satisfactorily explained by a subduction model. Decreasing oxygen fugacity with increasing pressure would eventually lead to carbonate reduction in a downgoing slab (Wood et al. 1996). Therefore, diamonds may form within the asthenosphere and the transition zone from former oceanic carbonates that typically have $\delta^{13}\text{C}$ values close to 0‰ (Veizer and Hoefs 1976). However, alternative explanations, such as carbon isotopic fractionation between melts and reduced C–H–O species, may equally apply. In this context it is interesting to note that the lower mantle diamonds from Kankan show a similar shift towards isotopically heavy compositions. This may be a coincidence, but it may also indicate that all sublithospheric diamonds at Kankan share a common carbon source. In this instance,

formation directly from subducted carbonates would provide one possibility but a common plume origin could equally be responsible. In terms of inclusion chemistry it appears impossible to choose between these two alternatives as a plume entraining former subducted oceanic crust (and possibly subducted carbon) would imprint a similar chemical signature (e.g. high Sr and/or negative and positive Eu anomalies).

One of the sublithospheric diamonds (KK-81), which contained a garnet with a high majorite component indicative of formation at the top of the transition zone (pressure of about 14 GPa, see Stachel et al. 2000a), has nitrogen mainly in the form of the poorly aggregated A-centre. Along an adiabatic mantle geotherm temperatures at that depth should exceed $1,400\text{ °C}$ (Brown and Shankland 1981; Ringwood 1991) and the observed aggregation level in this diamond is accomplished almost instantaneously (≤ 0.2 Ma) after formation. Longer mantle residence times are plausible only if this diamond was formed and stored in an environment much cooler than $1,400\text{ °C}$. A possible scenario is diamond formation in a 'cold' subducting slab and subsequent storage in the cratonic lithosphere. However, the very short time this diamond may have been exposed to the high ambient temperatures of the transition zone and asthenosphere suggests an exhumation mechanism operating much faster than mantle convection. This corresponds well with a previous observation made on Kankan diamonds: touching inclusions of CaSi-titanite and larnite show only incomplete reaction to CaSi-walstromite, which suggests that these Kankan diamonds were picked up by a rapidly ascending magma at pressures of 10–12 GPa (Joswig et al. 1999; Stachel et al. 2000b). The low nitrogen aggregation level found for KK-81 may now even suggest temporal proximity between diamond formation and Cretaceous magmatic activity (Banankoro kimberlites; Haggerty 1992).

Conclusions

The chemical characteristics of lithospheric diamonds from Kankan fit very well into the picture that has emerged from numerous studies on diamond sources worldwide. Peridotitic diamonds from Kankan show a narrow distribution in $\delta^{13}\text{C}$ around -4.5‰ , consistent with a formation from mantle-derived carbon during redox reactions. Eclogitic diamonds show a large spread in $\delta^{13}\text{C}$ with isotopically light compositions for some diamonds. The observation that eclogitic diamonds (worldwide) form at identical temperatures as peridotitic diamonds suggests a common origin within the cratonic lithosphere. This is not in conflict with a possible subduction origin of diamondiferous eclogites at Kankan (Stachel et al. 2000a) but requires an external carbon source. In contrast to peridotitic diamonds, the carbon for the formation of eclogitic diamonds either is chemically evolved through fractionation processes or is derived from several sources (including slab-derived melts

or fluids). However, two Kankan diamonds with light carbon isotopic composition, high nitrogen content and very low nitrogen aggregation may actually represent diamond formation in a cool, nitrogen rich and, therefore, isotopically unfractionated (Cartigny et al. 2001b) environment, consistent with possible formation directly in subducting oceanic crust.

Sublithospheric diamonds containing majorite garnet inclusions have a light carbon isotopic composition at Jagersfontein and Sao Luiz, which is not likely to represent a subduction signature because diamond formation directly from organic matter should already occur at much lower pressures. Chemical fractionation through incorporation of carbon into silicate phases (Galimov et al. 1989; Deines 2001) may provide a mechanism to explain these carbon isotopic compositions. Sublithospheric diamonds from Kankan, derived from the asthenosphere, the transition zone and the lower mantle, all show carbon isotopic signatures that are on the heavy side of the mantle value. This may indicate a contribution of subducted carbonates.

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