MINERALOGICAL MAGAZINE

VOLUME 43 NUMBER 330 JUNE 1980

The uranium content of garnet lherzolite xenoliths from kimberlites

D. A. CARSWELL

Department of Geology, University of Sheffield, Mappin Street, Sheffield, S1 3JD

AND

C. M. RICE

Department of Geology and Mineralogy, University of Aberdeen, Marischal College, Aberdeen, AB9 1AS

SUMMARY. Uranium contents ranging from 0.03-1.18 ppm have been determined by delayed neutron activation analysis in a suite of 19 ultrabasic xenoliths (mostly of the common garnet lherzolite type) from the kimberlites of southern Africa. Consideration of these and previous data lead us to conclude that the U content of uncontaminated garnet lherzolite in the uppermost mantle is on average roughly 0.04-0.05 ppm, levels which are sufficient to account for the observed U contents of fresh basalts. Higher U contents in mantle-derived ultrabasic xenoliths are the result of secondary alteration processes, most probably related to infiltration metasomatism stemming from contact with kimberlitic or carbonatitic magma fractions. This introduced U is concentrated along the boundaries of the primary mineral grains and is mostly readily removed by acid leaching. A partial analysis is given for a secondary uraniferous calcium phosphate phase observed in kelyphitic rims around garnets.

IT is generally accepted that the garnet lherzolite xenoliths brought up by kimberlites are representative of the dominant rocks in the uppermost 100-200 km of the mantle (Rickwood, 1969; Carswell and Dawson, 1970; Nixon and Boyd, 1973; Sobolev, 1977, etc.). As such mantle depths probably also correspond to the source region for basaltic magmas (Ringwood, 1975) it is important to decide whether garnet lherzolite xenoliths represent pristine mantle capable of yielding basaltic magmas through feasible partial melting and fractionation processes (O'Hara and Yoder, 1967) or residual mantle depleted in basalt yielding constituents (Ringwood, 1975).

Major element contents of lherzolite xenoliths

suggest (Carswell, 1980) that there are significant lateral and vertical variations in the basalt yielding potential of the uppermost mantle rocks. With this in mind it is of interest to determine the U contents of such rocks, especially as they will profoundly influence the heat generating capacity of the uppermost mantle.

Previous studies. Analyses of U contents in rock samples of garnet lherzolites from kimberlites are sparse, surprisingly being fewer in number than analyses of U in griquaite (mantle-derived eclogite) xenoliths which clearly represent a more specialized and subordinate component of the upper mantle, U contents determined in some 17 ultrabasic xenoliths (peridotites) from kimberlites, mostly from localities in southern Africa, range from 0.052-1.2 ppm (Holmes and Paneth, 1936; Heier, 1963; Wakita et al., 1967; Akimov et al., 1968; Fisher, 1970; Morgan and Lovering, 1971; Manton and Tatsumoto, 1971; Kresten, 1974). Discounting the high early values of Holmes and Paneth (1936) and other high values attributable to later alteration processes, the best previous values for the primary U content of garnet peridotite (lherzolite) xenoliths appear to be in the range 0.052-0.276 ppm. On this basis Kresten (1974) suggested that unaltered garnet peridotite xenoliths from kimberlites contain roughly 0.20 ppm U.

By contrast many more analyses are available of the U contents in spinel lherzolite xenoliths brought up in magmas of the alkali basaltbasanite-nephelinite suite. The mean U content determined in 57 such xenoliths is 0.0425 ($\sigma n = 0.0450$)—data from Wakita *et al.* (1967); Green *et al.* (1968); Kleeman *et al.* (1969); Fisher (1970); Nishimura (1972); Haines and Zartman (1973); Dostal and Capedri (1976). The mean value for the 22 determinations by neutron activation analysis (0.0385) is reasonably close to, but nevertheless slightly lower than, that for the 35 determinations by fission track analysis (0.0450). Despite some overlap in their U contents, previous analyses thus appear to indicate that spinel lherzolite xenoliths in alkali basalt suite magmas typically have lower U contents than garnet lherzolite xenoliths in kimberlites.

Analytical methods. The U contents of powdered rock and mineral samples were determined by delayed neutron activation analysis using a system directly developed from that of Amiel (1962). Five replicate determinations on sample BD 1133 indicate a precision at 1σ of 5%. The analyses were carried out by Mr T. Herrington, Analytical Chemistry Group, MOD (PE), at the Atomic Weapons Research Establishment, Aldermaston, Berks., England.

For fission track radiography the samples received a total neutron irradiation of $I \times 10^{16}$ n cm⁻². Lexan plastic was used as a detector and the prints were etched in 6N NaOH at 90 °C for ten minutes.

Uranium contents in rocks. The U contents determined in a suite of 19 ultrabasic xenolith samples from kimberlites are given in Table II, along with notes on the petrography of these samples. The majority of samples are of the common garnet lherzolite type (Carswell and Dawson, 1970; Carswell, 1980) taken to represent the dominant rock type in the uppermost 150-200 km of the mantle. We have classified such xenoliths as garnet lherzolites as long as they contain the 4-phase primary assemblage of olivine + orthopyroxene + clinopyroxene+garnet, despite the fact that the contents of garnet and clinopyroxene are typically < 10%. Some of the xenoliths contain minor amounts of phlogopite interpreted as primary on the basis of its apparent textural and chemical equilibration with the 4-phase garnet lherzolite assemblage (Carswell, 1975). The group of xenoliths studied from the Matsoku pipe in northern Lesotho contains two xenoliths (BD 1354 and 1360) with much higher modal contents of garnet and clinopyroxene. These xenoliths also have lower 100 Mg/(Mg + Fe) ratios and belong to the distinctive banded peridotite-pyroxenite series of xenoliths interpreted to be of cumulate origin (Cox et al. 1973; Gurney et al. 1975). The group of xenoliths from the Kimberley area mines contains one residual type harzburgite xenolith (BD 1200) totally devoid of garnet and clinopyroxene. Major element analyses of all the BD xenolith samples are given by Carswell and Dawson (1970), whilst analyses for the remaining xenolith samples are given in Table I.

The U contents in these 19 xenolith samples are decidedly variable ranging from 0.03-1.18 ppm, with a mean value of 0.178 ppm (on 0.254 ppm). In these xenoliths no apparent correlation exists between U content and major element chemical parameters such as 100 Mg/(Mg + Fe) and 100 Cr/ (Cr+Al) ratios, which are useful indices of the 'fertile' or 'depleted' chemical character of the mantle in terms of its basalt yielding potential. Instead the variation in U content appears to reflect variation in the type and degree of secondary alteration which has affected these xenolith samples. The dramatically reduced U contents in three xenolith samples after overnight leaching at room temperature in 2N HCl indicate that in those samples with highest U contents much of it is in labile sites.

Uranium contents of mineral phases. The U contents determined in mineral separates from certain of our xenolith samples are listed in Table III, along with previous determinations (mainly by fission track analysis) of the U contents of the primary mineral phases in other ultrabasic xenoliths from kimberlites. The U levels determined in our study are more in line with the results obtained by Akimov et al. (1968) and Mitchell and Aumento (1974) than with those of Kleeman and Lovering (1973). Whilst Mitchell and Aumento (1974) determined orthopyroxenes as having the highest U contents of the 4-phase garnet lherzolite assemblage, our results for the only xenolith sample for which we have determined U levels in all four mineral phases are in agreement with those of both Akimov et al. (1968) and Kleeman and Lovering (1973) to the extent that U appears to be regularly distributed between the phases and decreases in the order garnet > clinopyroxene > orthopyroxene \geq olivine.

Discussion. Mass balance calculations for those xenoliths for which we have determined the U contents of the mineral phases (making reasonable assumptions for U levels in orthopyroxene and olivine in three out of four cases) yield calculated U contents in rocks reasonably in line with the observed values. However, this is not so if the average U contents for the minerals are used to calculate rock U contents in certain other xenoliths. Fission track mapping on lexan prints shows that in such xenoliths much of the element is concentrated along the boundaries of the primary mineral phases and it is probably this U that is being removed by acid leaching. The precise nature of these secondary U-enriched phases is not entirely clear although the

	CB1	CB2	CB4	CK2	СК3	CK4	СК33
Si0,	45.50	45.55	49.77	44.79	45.09	45.07	45.41
TIO2	0.07	0.04	0.09	0.00	0.06	0.04	0.02
A1203	1,85	1.36	2.56	1.90	2.18	2.06	1.68
cr203	0.31	0.23	0.42	0.27	0.31	0.32	0.28
Fe ₂ O ₃	2.08	1.82	0.27	1.80	0.97	2.07	2.09
Fe0	4.37	4.07	5.04	4.83	5.63	4.56	3.53
MnO	0.10	0.09	0.11	0.11	0.11	0.10	0.09
NIC	0.27	0.28	0.23	0.26	0.26	0.25	0.30
MgO	41.05	42.68	37.73	41.33	40.66	40.92	41.56
CaO	1.19	0.89	0.89	1.14	1.21	1.20	0.51
Na ₂ O	0.16	0.10	0.12	0.22	0.36	0.17	0.10
K-0	0.10	0.10	0.06	0.14	0.26	0.16	0.05
P.05	0.06	0.04	0.04	80.0	0.11	0.06	0.03
· H_0+	3.14	2.62	1.81	2,90	2.95	2.89	4.25
c0 ₂	0.29	0.25	0.14	0.21	0.24	0.23	0.30
Total	100.54	100.12	99.28	99.98	100.40	100.10	100.20
100 Cr/(Cr + A1)	10.1	10.2	9.9	8.7	8.7	9.4	10.1
100 Mg/{Mg + Fe_}}	92.1	93.0	92.7	91.9	91.8	91.9	93.2

TABLE I: Additional Analyses of Garnet Lherzolite Xenoliths from Kimberlites

Analyst: V.A. Somogyi, Department of Geology, University of Sheffield, using a combination of atomic absorption, spectrophotometric and gravimetric techniques.

TABLE II: Uranium Contents in Ultrabasic Xenoliths from Kimberlites

Locality	Sample Number	Uppm	Features
	BD 1355	0.06	Coarse equigranular textured garnet lherzolite ('Common Peridotite' xenolith type, Cox et al., 1973)
	BD 1356	0.041	Similar to BD 1355, but with more phlogopite - mostly in rims around garnets
Matsoku North Lesotho	BD 1354	0.03	Coarse textured garnet and clinopyroxene rich garnet lhersolite from banded peridotite-pyroxenite series xenoliths (Cox et al. 1973) interpreted as cumulates
	BD 1360	0.18 (0.009)*	Similar to BD 1355 but with 4 vol.% phlogopite - both as rims around garnets and in metasomatic veins with rutile
	BD 1364	0.06	Similar to BD 1354 but with even more clinopyroxene and less olivine
	BD 1140	0.1	Porphyroclastic textured garnet lherzolite. Th 0.26 ppm
	BD 1143	1.18 (0.034)*	Coarse textured garnet lherzolite fairly heavily veined with serpentine
Kimberley	BD 1150	0.10	Coarse textured garnet lherzolite with abundant phlogopite mostly along grain boundaries but also some discrete primary textured plates
Area Mines South Africa	BD 1200	0.041	Coarse textured harzburgite with 'fingerprint' spinels intergrown with ortho- pyroxene
	BD 1201	0.041	Coarse equigranular textured garnet lher2olite
	СК 2	0.27	Coarse textured garnet lherzolite with scarce primary textured phlogopite and secondary amphibole rims around garnets
	скз	0.41 (0.23)*	Coarse textured garnet lherzolite with scarce primary textured phlogopite and secondary amphibole and phlogopite rims around garnets
	CK 4	0.20	Coarse textured garnet lherzolite with primary and secondary textured phlogo- pite (Carawell 1975)
	СК 33	0.034	Porphyroclastic textured garnet lherzolite with very scarce clinopyroxene
	СВ 1	0.19	Coarse textured garnet lherzolite with minor primary and secondary textured phlogopite
	CB 2	0.13	Porphyroclastic textured garnet lherzolite with minor secondary phlogopite rims around garnets
	СВ 4	0.11	Coarse textured garnet lherzolite with minor secondary phlogopite as rims around garnets and clinopyroxene with unusual distinctly poikilitic habit (Carswell 1975)
Jagersfontein	BD 1127	0.053	Mosaic - porphyroclastic textured garnet lherzolite
Mine South Africa	BD 1133	0.154	Coarse textured garnet lherzolite with considerable secondary phlogopite. Range in five uranium determinations 0.143-0.161 ppm

*Values in parenthesis obtained after samples leached in 2N HCl at room temperature overnight Analyst: J. Herrington

TABLE III: Uranium Contents (in ppm) in the Mineral Phases of Ultrabasic Xenoliths from Kimberlites

Sample	ence	Mineral	Garnet	Clinopyroxene	Orthopyroxene	Olivine	Analytical • Method
This Study	BD	1354	0.07	0.04	-		Delayed Neutron
	BD	1364	0.28	0.04	-		Activation
	BD	1140	0.5 ± 0.05	0.3 ± 0.05	0.1	<0.05	Mineral
	BD	1150	0.21	0.07	-	-	Separates
Ak	imov ((196	et al. 8)	0.129 (Aver.3)	0.069 (Aver.3)	0.029 (Aver.2)	0.030 (Aver.3)	Fission Track Analysis
Kleema	n and (197	Lovering 3)	0.002-0.14 (Range)	0.007-0.030 (Range)	<0.001-0.0072 (Range)	<0.0003	Fission Track Analysis
Mitche	ell an (197	d Aumento 4)	0.12 (Aver.8)	0.072 (Aver.10)	0.88 (Aver.20)	0.06 (Aver.17)	Fission Track Analysis
Kra	amers	(1979)	-	≤0.006-0.045 (Range)	-	-	Isotope Dilution Mass Spectrometry on Separates

fact that the sample with the highest U content (BD 1143) also has the highest CO_2 content suggests that at least some of it might be carbonate.

Kimberlites themselves have appreciably higher U contents than the included xenoliths. Thus the average U in 159 determinations in kimberlites from southern Africa is 3.13 ppm-data from Morgan and Lovering (1971); Gurney and Hobbs (1973); Kresten (1974); Fesq et al. (1975); and Kramers (1977). This suggests that infiltration metasomatism emanating from the enclosing kimberlite, such as is apparently responsible for the development of secondary micas around garnets (Carswell, 1975), may well be responsible for the elevated U contents in certain xenoliths. However, there is only a poor positive correlation between the U contents of the xenoliths and their K₂O contents which might be taken as a possible index of the degree of contamination from the kimberlite. As carbonatites appear to have typically even higher U contents than kimberlites (Dawson and Gale, 1970; Masaits and Symslov, 1977) and carbonated kimberlites higher U contents than basaltic kimberlites (Kresten, 1974) it is possible that it is the degree of contamination due to a carbonatitic. rather than a strictly kimberlitic, component which is responsible for the elevated U contents in certain xenoliths (cf. Brookins et al., 1979).

Some of the xenoliths, notably the CK and CB samples, appear to show a positive correlation between their U and P contents. In view of the suggestion by Fesq et al. (1975) and Kable et al. (1975) that primary upper mantle apatite might be the principal source for the concentrations of both of these elements in kimberlites, it is important to establish whether apatite exists as a primary phase, albeit in minor amounts, in garnet lherzolite xenoliths. Kleeman et al. (1969) observed that primary apatite in small amounts was an important Ubearing phase (with about 35 ppm U) in certain spinel lherzolite xenoliths from Victorian basalts. However, we have not been able to detect primary apatite grains in these xenoliths, either optically or with the microprobe. Microprobe analyses of wt. % P_2O_5 in the mineral phases of xenolith CK4 gave the following values: garnet (0.034), clinopyroxene (0.020), orthopyroxene (0.0043), olivine (0.018), and phlogopite (0.0034). The fact that in the primary mineral assemblage garnet has the highest contents of both U and P may therefore help to explain the positive correlation between these elements in many of these xenoliths. However, the primary mineral phases in CK4 contain collectively less P_2O_5 than the rock content of 0.06 wt. %, indicating the presence of an additional P-enriched phase. Microprobe examination showed that the kelyphite rims around the garnets in this xenolith contain small grains (maximum dimension about 10 μ m) of a uraniferous calcium phosphate phase. A partial analysis (average of five areas) of this phosphate gave P₂O₅ 33.7, CaO 48.7, TiO₂ 0.04, Y₂O₃ 0.08, La₂O₃ 0.35, Ce₂O₃ 0.59, UO₂ 0.032, all in wt.%. We think it unlikely that sufficient, if any, primary apatite occurs in most garnet lherzolite xenoliths to significantly affect the U budget.

Five of our garnet lherzolite samples plus four samples previously studied by Akimov *et al.* (1978), Fisher (1970), and Morgan and Lovering (1971) have U contents in the range 0.034–0.075 ppm, the mean value for these 9 samples being 0.053 ppm. We consider such levels to represent the true background levels of U in garnet lherzolite xenoliths unaffected by contamination associated with secondary alteration processes. Such levels are closely in line with the U levels determined in mantle-derived spinel lherzolite xenoliths from alkali basalt suite magmas, for which a mean value of 0.0425 ppm U for 57 such xenoliths was quoted earlier in this paper.

It is then pertinent to consider whether a U content of the order of 0.04-0.05 ppm in the uppermost mantle is sufficient to account for the levels observed in basaltic magmas if these are taken to be partial melts of such garnet lherzolite source rocks. It is clear that U can be expected to be strongly partitioned into partial melts relative to residual silicate phases. Certainly in the light of U_{Cpx}/U_{melt} partition coefficients most likely to be within the range 0.001-0.024 (Onuma et al., 1968; Kleeman et al., 1969; Kleeman and Lovering, 1973; Seitz, 1973; Seitz et al., 1974; Dostal and Capedri, 1976), there seems to be little problem in generating partial melts with the U levels observed in fresh ridge or intraplate basalts $(0.078 \pm 0.048 \text{ ppm U} \text{ in } 68)$ samples) (Fisher, 1979) from upper mantle source rocks with 0.04-0.05 ppm U.

On the basis of xenolith studies (Heier, 1963; Wakita et al., 1967; Green et al., 1968; Morgan and Lovering, 1971; and BD 1140 Table II) Th/U rations in the uppermost mantle are likely to be within the range 2-5. Undoubted contamination affects from the enclosing kimberlite make it extremely hazardous to fix upper-mantle K levels on the basis of the K concentrations observed in garnet lherzolite xenoliths. However, best estimates of K/U ratios in the uppermost mantle appear to be within the range $3 \times 10^3 - 1 \times 10^4$ (Wakita et al., 1967; Green et al., 1968; Fisher, 1970, 1979). With these considerations in mind a subcontinental uppermost mantle with on average ~ 0.05 ppm U, ~ 0.19 ppm Th, and \sim 500 ppm K as estimated by Birch (1965) on the basis of heat flow data requirements seems fairly realistic. On the other hand, as emphasized recently by Fisher (1979), if such concentrations are

also valid for the sub-oceanic mantle then they are clearly inadequate to explain observed heat flows in those regions on the basis of radiogenic heat generation alone.

Acknowledgements. D. A. Carswell acknowledges support from the Royal Society and the Natural Environmental Research Council.

REFERENCES

- Akimov (A. P.), Berzina (I. G.), Gurvich (M. Y.), and Lutts (B. G.), 1968. Akad. Nauk. SSSR, Dokl. 181, 1245-8.
- Amiel (S.), 1962. Anal. Chem. 34, 1683-92.
- Birch (F. R.), 1965. Geol. Soc. Am. Bull. 76, 133-53.
- Brookins (D. G.), Della Valle (R. S.), and Bolivar (S. L.), 1979. In Boyd (F. R.) and Meyer (H. O. A.) (eds.), Kimberlites, Diatremes, and Diamonds: Their Geology, Petrology and Geochemistry. Amer. Geophys. Union, 280-8.
- Carswell (D. A.), 1975. Phys. Chem. Earth, 9, 417-29. — 1980. Lithos 13, 121-38.
- Cox (K. G.), Gurney (J. J.), and Harte (B.), 1973. In Nixon (P. H.) (ed.), *Lesotho Kimberlites*. Lesotho Nat. Devel. Corp. Maseru, Lesotho, 76-92.
- Dawson (J. B.) and Gale (N. H.), 1970. Chem. Geol. 6, 221-31.
- Dostal (J.) and Capedri (S.), 1976. Contrib. Mineral. Petrol. 54, 245-54.
- Fesq (H. W.), Kable (E. J. D.), and Gurney (J. J.), 1975. Phys. Chem. Earth, 9, 687-707.
- Fisher (D. E.), 1970. Geochim. Cosmochim. Acta, **34**, 630-4. — 1979. Ibid. **43**, 709-16.
- Green (D. H.), Morgan (J. W.), and Heier (K. S.), 1960. Earth Planet. Sci. Lett. 4, 155-66.
- Gurney (J. J.), Harte (B.), and Cox (K. G.), 1975. Phys. Chem. Earth, 9, 507-23.

— and Hobbs (J. B. M.), 1973. Int. Conf. on Kimberlites, Ext. Abstracts, 143-5.

- Haines (E. L.) and Zartman (R. E.), 1973. Earth Planet Sci. Lett. 20, 45-53.
- Heier (K. S.), 1963. Geochim. Cosmochim. Acta, 27, 849-60.

- Holmes (A.) and Paneth (F. A.), 1936. Proc. R. Soc. Lond, A, 154, 385-413.
- Kable (E. J. D.), Fesq (H. W.), and Gurney (J. J.), 1975. Phys. Chem. Earth, 9, 709-34.
- Kleeman (J. D.), Green (D. H.), and Lovering (J. F.), 1969. Earth Planet. Sci. Lett. 5, 449-58.
- ----- and Lovering (J. F.), 1973. Int. Conf. on Kimberlites, Ext. Abstracts, 189-90.
- Kramers (J. D.), 1977. Earth Planet. Sci. Lett. 34, 419-31. — 1979. Ibid. 42, 58-70.
- Kresten (P.), 1974. Lithos, 7, 171-80.
- Manton (W. L.) and Tatsumoto (M.), 1971. Earth Planet. Sci. Lett. 10, 217-26.
- Masaits (V. L.) and Symslov (A. A.), 1977. Geochem. Intern. 14, 129-40.
- Mitchell (W. S.) and Aumento (F.), 1974. J. Geophys. Res. 79, 5529-32.
- Morgan (J. W.) and Lovering (J. F.), 1971. In Brunfelt (A. O.) and Steinnes (E.) (eds.), Activation Analysis in Geochemistry and Cosmochemistry, Universitetsforlaget, Oslo, 445-54.
- Nishimura (S.), 1972. Chem. Geol. 10, 211-21.
- Nixon (P. H.) and Boyd (F. R.), 1973. In Nixon (P. H.) (ed.), Lesotho Kimberlites, Lesotho Nat. Devel. Corp. Maseru, Lesotho, 48-56.
- O'Hara (M. J.) and Yoder (S. J.), 1967. Scott. J. Geol. 3, 67-117.
- Onuma (N.), Higuchi (H.), Wakita (H.), and Nagasawa (H.), 1968. Earth Planet. Sci. Lett. 5, 47-51.
- Rickwood (P. C.), 1969. Geol. Soc. S. Africa, Special Publ. 2, 395-416.
- Ringwood (A. E.), 1975. Composition and Petrology of the Earth's Mantle. McGraw Hill, New York, 618 pp.
- Seitz (M. G.), 1973. Carnegie Inst. Wash. Year Book, 72, 581-6.
- ----- Burnett (D. S.), and Bell (P. M.), 1974. Ibid. 73, 451-4.
- Sobolev (N. V.), 1977. Deep seated inclusions in kimberlites and the problem of the composition of the upper mantle. Amer. Geophys. Union, Wash. 279 pp.
- Wakita (H.), Nagasawa (H.), Vyeda (S.), and Kuno (H.), 1967. Geochem. J. 1, 183-98.

[Manuscript received 25 October 1979]