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A secretive mechanical exchange between mantle and crustal volatiles revealed by helium isotopes in ¹³C-depleted diamonds

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Supplementary Information

The Supplementary Information includes:

- Sample Descriptions
- > Carbon and Nitrogen Isotope Analysis
- Helium Isotope Analysis
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- Supplementary Information References

Sample Descriptions

The Orapa diamondites originate from the largest of 23 pipes in the Orapa cluster in north-eastern Botswana, emplaced at 93.1 Ma on the western margin of the Kalahari Craton (samples described in Mikhail *et al.*, 2019). The southern African diamondites were selected from a collection at the Naturhistorische Museum Vienna, Austria. Detailed descriptions of the samples are provided by Kurat and Dobosi (2000). Similar samples were described from the Orapa, Jwaneng (Botswana; Gurney and Boyd, 1982) and Venetia kimberlites (South Africa; Jacob *et al.*, 2000). These samples likely originate from one or all these sources. The complete sample suite (Orapa + southern African diamondites) incorporates a range of silicate parageneses; five contain websteritic garnets, four contain eclogitic garnets, one shows a mixed paragenesis containing both peridotitic clinopyroxene (Mg# = 90.81) and an orange eclogitic garnet, and seven samples contain no paragenesis-defining intergrowths/inclusions (Table S-1).

Carbon and Nitrogen Isotope Analysis

The δ^{13} C-, δ^{15} N-values and N-concentrations were obtained using the custom-made fully automated Finesse machine at the Open University, UK (Verchovsky *et al.*, 1998) following the method outlined in Mikhail *et al.* (2014). Samples analysis was undertaken using platinum sample buckets (99.95 % purity platinum foil of 0.025 mm thickness). A foil strip was cut and placed in a 6 mm ID quartz tube alongside a platinum-wrapped mass of copper oxide wire, then connected to the vacuum system of Finesse in order to evacuate the tube. After this process, the tube was torched off using a hydrogen-oxygen blowtorch, collapsing the glass in order to maintain vacuum, and placed overnight in a 900 °C muffle furnace. This has the effect of liberating oxygen from the breakdown of the copper oxide wire, which serves to oxidise and remove contamination from the foil. Once removed from the muffle furnace, the tube was allowed to cool before being opened in a class 100 clean room. The sample buckets themselves were produced but cutting subsections from the foil strip and folding them into the correct shape using cleaned scissors and tweezers within the clean room. Samples were weighed directly into the buckets using a Sartorius microbalance that had been zeroed to the mass of each platinum



bucket. Tweezers were used to place fragments of the samples into the buckets, after which they were weighed, closed and gently compacted then placed into Finesse sample inlet system.

Once within the furnace, the samples were step heated. The first two steps were cleaning steps, consisting of a single pyrolysis at 1100 °C followed by combustion at 500 °C, in order to remove any residual contaminants. Subsequently, samples are combusted at 1100 °C for as many steps as were required return the system to blank levels (*i.e.* complete oxidation of diamond). Oxygen gas (O₂) was derived from the breakdown of copper oxide wire in a separate linked furnace at 850 °C, with any residual oxygen at the end of each combustion step resorbed when the temperature of the CuO furnace dropped to 600 °C. The resultant gas was then passed through to a liquid nitrogen-cooled cold finger array and a molecular sieve, collectively separating and purifying the CO₂, N₂, H₂O, and noble gases. These were then separated by a valve before the cold finger attached to a 1 Torr MKS Baratron. Non-condensable gases were subsequently pumped away, the Baratron section isolated and the cold finger warmed to release the CO₂ for measurement on the Baratron. δ^{13} C was determined by a triple-collector magnetic sector mass spectrometer operating in static mode.

Following carbon analysis, the nitrogen was desorbed from the molecular sieve and cleaned using a secondary copper oxide source in order to remove any contribution from CO. An aliquot was sent to the quadrupole mass spectrometer in order to determine the N₂-abundance and to allow the system to determine and carry out a gas splitting procedure. The remaining gas was split and sent to the nitrogen mass spectrometer, again a triple collector magnetic sector instrument operating in static mode. The gas was purified by use of another cold finger adjacent to the mass spectrometer and used to remove any condensable gases. This provided both the δ^{15} N and the nitrogen abundance through a calculation based on the intensity of ¹⁴N and the known volumes of the instrument.

Carbon and nitrogen stable isotope ratios (${}^{13}C/{}^{12}C$ and ${}^{15}N/{}^{14}N$) are expressed in delta notation relative to the Pee Dee belemnite and air standards, respectively (*e.g.*, $\delta^{13}C = ({}^{13}C/{}^{12}C$ sample / ${}^{13}C/{}^{12}C$ standard – 1)). The accuracy of the analysis determined by the analysis of the internal standards is ±<0.5 % for $\delta^{13}C$ and $\delta^{15}N$ values and <10 % for the N concentrations (2 σ). Carbon isotopic errors are determined as standard deviations of the reference materials. Nitrogen isotopic errors were determined using the statistical methodology outlined by Equations S-1 to S-3 (see Mikhail *et al.*, 2014).

Mass of nitrogen in the sample =
$$N_m - N_b$$
 Eq. S-1

$$\delta^{15} N_c = \frac{(\delta^{15} N_m) - (\delta^{15} N_b \cdot N_b)}{N_m - N_b}$$
 Eq. S-2

$$\Delta \delta^{15} N_c = \frac{1}{(N_m - N_b)^2} \cdot \sqrt{\left[(1 - \delta^{15} N_m)^2 \cdot (\Delta N_m)^2 + (1 - \delta^{15} N_b)^2 \cdot (N_m - N_b)^2 \cdot ((\Delta N_m)^2 \cdot (\Delta \delta^{15} N_m)^2 + (\Delta N_b)^2 \cdot (\Delta \delta^{15} N_b)^2) \right]}$$
Eq. S-3

where N_m and N_b refer to the mass of nitrogen in the measured gas and blank, $\delta^{15}N_m$ and $\delta^{15}N_b$ refer to the $\delta^{15}N$ value of the measured gas and blank, $\Delta\delta^{15}N_m$ refers to the measured error in per mil, $\Delta\delta^{15}N_b$ and ΔN_b refer to the standard deviation for the multiple blank analysis and $\Delta\delta^{15}N_c$ is the blank corrected precision of the $\delta^{15}N$ value of the sample.

Helium Isotope Analysis

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Helium analyses for samples from Orapa were undertaken using a ThermoFisher Scientific Helix SFT dual collector mass spectrometer. Southern African samples were analysed using a MAP 215-50 all-metal magnetic sector mass spectrometer. Both analyses were undertaken at the Scottish Universities Environmental Research Centre following methods adapted from Stuart *et al.* (2000). Sample fragments were weighed using a Satorius MSE3.6P-000-DM Cubis Micro Balance and washed with acetone in an ultrasonic bath for 15 minutes at the University of St. Andrews. Samples were then loaded into an all-metal multi-sample hydraulic crusher. The gas trapped in fluid inclusions is released by *in vacuo* crushing and then purified in an all-metal ultra-high-vacuum line equipped with two heated (250 °C) SAES GP50 getters and one activated charcoal finger (cooled to approximately -196 °C using liquid nitrogen) for 15 minutes prior to expand into the mass spectrometer. The mass spectrometer is equipped with a SAES GP50 getter (operated at room temperature) and a stainless steel finger filled with activated charcoal (cooled to *ca.* -196 °C using liquid Nitrogen) installed close to the source to minimise partial pressure of residual gases during analysis. ³He is analysed by a copper beryllium ion counting electron multiplier and ⁴He is analysed by an electrically suppressed Faraday detector. Determination of mass spectrometer sensitivity and ratio reproducibility was performed measuring multiple times each day helium standard gas HESJ (Matsuda *et al.*, 2002) with ³He/⁴He ratio of 20.63 \pm 0.10 R_A (R_A being the ³He/⁴He ratio of air 1.34 x 10⁻⁶,



Mishima *et al.*, 2018) and blank analyses were frequently run to obtain a well constrained background level of ³He and ⁴He. The average blank levels of the ThermoFisher Scientific Helix SFT are 4.21 x $10^{-12} \pm 2.89 \times 10^{-12}$ cm³ for ⁴He and 6.68 x $10^{-16} \pm 1.59 \times 10^{-16}$ cm³ for ³He. The average blank levels of the MAP 215-50 are 7.52 x $10^{-11} \pm 8.06 \times 10^{-12}$ cm³ for ⁴He and $1.17 \times 10^{-15} \pm 1.25 \times 10^{-15}$ cm³ for ³He.

The intensity of the ion beams of both ³He and ⁴He are measured simultaneously in a 20-cycle run setup using Qtegra software. The ³He and ⁴He abundance of each sample was calculated by taking the average of these 20 cycles and the uncertainty is the standard deviation. The blank corrected ³He/⁴He isotopic ratios of each sample (R) are expressed relative to the ³He/⁴He of air ($R_a = 1.34 \times 10^{-6}$; Mishima *et al.*, 2018) as R/R_a using the internal HESJ standard.



Table S-1 Full dataset used in this study.

Source of Data		Location	Туре	Paragenesis	Minerals	R/Ra New	±	⁴ He ccSTP/g	⁴ He bulk	⁴ He bulk error	³ He ccSTP/g	³ He bulk	³ He bulk error	δ¹³C	±	N at. ppm	δ¹⁵N	±
This study	DIA030	Southern Africa	Polycrystalline	Websteritic	Garnet	8.51	0.42	1.330E-07	4.456E-09	1.742E-11	1.506E-12	4.857E-14	2.714E-15	-16.6	0.2	8	+6.4	3.9
This study	DIA053	Southern Africa	Polycrystalline	Websteritic	Garnet	2.76	0.70	3.200E-08	9.075E-10	1.358E-11	1.177E-13	2.409E-15	1.691E-15	-20.8	0.5	56	+2.0	0.7
This study	DIA057B#1	Southern Africa	Polycrystalline	Unknown	None characteristic	0.06	0.05	6.790E-07	1.696E-08	6.857E-11	5.810E-14	2.107E-16	1.782E-15	-21.4	0.1	1389		
This study	DIA057B#2	Southern Africa	Polycrystalline	Unknown	None characteristic	0.14	0.05	6.900E-07	1.725E-08	5.846E-11	1.247E-13	1.676E-15	1.783E-15	-21.4	0.1	1389		
This study	DIA058B	Southern Africa	Polycrystalline	Eclogitic	Orange Garnet	3.89	0.21	7.420E-08	7.835E-09	3.339E-11	3.841E-13	4.066E-14	2.526E-15	-19.1	0.1			
This study	DIA059	Southern Africa	Polycrystalline	Websteritic	Garnet	0.53	0.20	1.420E-07	4.218E-09	1.740E-11	1.007E-13	1.676E-15	1.690E-15	-22.2	0.3	13	+5.3	6.1
This study	DIA073B	Southern Africa	Polycrystalline	Websteritic	Garnet	1.91	0.40	8.710E-08	4.008E-09	1.759E-11	2.210E-13	8.710E-15	2.461E-15	-17.4	0.1	2812	+6.9	0.5
This study	DIA077	Southern Africa	Polycrystalline	Unknown	None characteristic	7.44	0.54	1.440E-07	5.934E-09	2.211E-11	1.425E-12	5.941E-14	4.414E-15					
This study	ORF9	Orapa, Bostwana	Polycrystalline	Webesteritic	Garnet			2.441E-08						-5.3	0.1			
This study	ORF12	Orapa, Bostwana	Polycrystalline	Unknown	None characteristic			6.725E-08										
This study	ORF19	Orapa, Bostwana	Polycrystalline	Ecologitic	Orange Garnet	4.22	2.72	9.021E-08	1.911E-10	1.159E-11	5.110E-13	1.082E-15	7.265E-16	-8.0	0.1	255	+5.8	0.1
This study	ORF26#1	Orapa, Bostwana	Polycrystalline	Peridotitic & Eclogitic	Clinopyroxene & Garnet			4.220E-09	1.006E-11	3.787E-12				-14.6	0.2	38	-4.9	2.2
This study	ORF26#2	Orapa, Bostwana	Polycrystalline	Peridotitic & Eclogitic	Clinopyroxene & Garnet				1.006E-11	3.787E-12				-17.8	0.2	19	+23.2	6.6
This study	ORF28#1	Orapa, Bostwana	Polycrystalline	Unknown	None characteristic			5.368E-08						-4.3	0.1	775	+2.9	0.2
This study	ORF28#2	Orapa, Bostwana	Polycrystalline	Unknown	None characteristic				1.137E-10	7.512E-12		2.390E-16	4.448E-16	-4.9	0.2	52	+19.7	0.6
This study	ORF41	Orapa, Bostwana	Polycrystalline	Unknown	None characteristic			7.766E-08						-14.8	0.3	1146	+14.7	0.2
This study	ORF57	Orapa, Bostwana	Polycrystalline	Eclogitic	Orange Garnet	7.53	0.75	2.238E-07	1.586E-09	8.884E-11	2.259E-12	1.602E-14	1.921E-15	-16.6	0.2	17	+18.4	3.0
This study	ORF60	Orapa, Bostwana	Polycrystalline	Unknown	Chromite				0.000E+00	0.000E+00		0.000E+00	0.000E+00	-6.5	0.1	1054	+12.1	0.2
This study	ORF91	Orapa, Bostwana	Polycrystalline	Unknown	None characteristic									-20.3	0.3	401	+4.4	0.3

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This study	ORF143	Orapa, Bostwana	Polycrystalline	Ecologitic	Orange Garnet	1.58	1.12	1.027E-07	4.400E-10	2.482E-11	2.174E-13	9.317E-16	6.858E-16	-19.9	0.2	647	+10.0	0.2
Gautheron et <i>al.,</i> 2005	ORPC 1	Orapa, Botswana	Polycrystalline	Eclogitic	Orange Garnet	0.56	0.06	4.620E-07	1.983E-07	7.296E-11	1.482E-13	6.360E-14		-14.0	0.1	202	+4.5	0.5
Gautheron et <i>al.,</i> 2005	ORPC 2	Orapa, Botswana	Polycrystalline	Eclogitic	Orange Garnet	1.34	0.11	1.490E-07	9.615E-08	4.517E-11	1.717E-13	1.108E-13		-21.0	0.1	543	+15.5	0.5
Gautheron et <i>al.,</i> 2005	ORPC 3	Orapa, Botswana	Polycrystalline	Eclogitic	Orange Garnet	0.43	0.04	3.010E-07	1.655E-07	1.803E-09	9.391E-14	5.163E-14		-19.4	0.1	163	+6.7	0.5
Gautheron et <i>al.,</i> 2005	ORPC 4 #1	Orapa, Botswana	Polycrystalline	Peridotitic	Enerald-Green Diopside	0.97	0.04	6.620E-07	4.572E-07	1.520E-10	5.949E-13	4.109E-13		-9.8	0.1	1973	+8.1	0.5
Gautheron et <i>al.,</i> 2005	ORPC 4 #2	Orapa, Botswana	Polycrystalline	Peridotitic	Enerald-Green Diopside	0.15	0.05	1.090E-06	2.476E-07	9.088E-12	4.798E-14	1.090E-14						
Gautheron et <i>al.,</i> 2005	ORPC 5	Orapa, Botswana	Polycrystalline	Unknown	None Characteristic	0.86	0.08	2.180E-07	1.240E-07	1.536E-10	1.425E-13	8.108E-14		-18.3	0.1	69	+8.1	0.5
Gautheron et <i>al.,</i> 2005	ORPC 6	Orapa, Botswana	Polycrystalline	Unknown	None Characteristic	0.54	0.04	5.430E-07	4.381E-07	7.552E-09	3.153E-13	2.544E-13		-21.0	0.1	168	+6.7	0.5
Burgess <i>et al.,</i> 1998	Jwaneng 3	Jwaneng, Botswana	Polycrystalline	Unknown	None Distinctive	4.46	0.10	2.480E-06			1.476E-11			-21.7	0.1			
Burgess <i>et al.,</i> 1998	Jwaneng 5	Jwaneng, Botswana	Polycrystalline	Unknown	None Distinctive									-5.5	0.1			
Burgess <i>et al.,</i> 1998	Jwaneng F6	Jwaneng, Botswana	Polycrystalline	Unknown	None Distinctive	0.18	0.00							-23.6	0.1			
Burgess <i>et al.,</i> 1998	Jwaneng F10	Jwaneng, Botswana	Polycrystalline	Unknown	None Distinctive	0.12	0.00							-23.6	0.1			
Burgess <i>et al.,</i> 1998	Jwaneng F11	Jwaneng, Botswana	Polycrystalline	Unknown	None Distinctive	0.13	0.01							-20.1	0.1			
Burgess <i>et al.,</i> 1998	Orapa 1	Orapa, Botswana	Polycrystalline	Eclogitic	Garnet	2.00	0.05	3.300E-07			8.815E-13			-18.6	0.1			
Burgess <i>et al.,</i> 1998	Orapa 2	Orapa, Botswana	Polycrystalline	Eclogitic	Garnet	1.97	0.04	3.000E-08			7.889E-14			-5.3	0.1			
Burgess <i>et al.,</i> 1998	Orapa 3	Orapa, Botswana	Polycrystalline	Eclogitic	Garnet	0.08	0.00				7.274E-12			-20.1	0.1			
Burgess <i>et al.,</i> 1998	Orapa 4	Orapa, Botswana	Polycrystalline	Eclogitic	Garnet	1.29	0.04	2.600E-07			4.462E-13			-9.6	0.1			
Burgess <i>et al.,</i> 1998	Orapa 5	Orapa, Botswana	Polycrystalline	Eclogitic	Garnet	1.38	0.02	2.200E-07			4.050E-13			-9.7	0.1			
Burgess <i>et al.,</i> 1998	Orapa F121	Orapa, Botswana	Polycrystalline	Eclogitic	Garnet	1.08	0.07				0.000E+00			-22.5	0.1			
Burgess <i>et al.,</i> 1998	Jwaneng 1	Jwaneng, Botswana	Coated	Unknown	None Distinctive	4.25	0.20	2.560E-06			1.453E-11			-5.9	0.1			
Burgess <i>et al.,</i> 1998	Jwaneng 2	Jwaneng, Botswana	Coated	Unknown	None Distinctive	3.84	0.10	3.220E-06			1.649E-11			-6.3	0.1			
Burgess <i>et al.,</i> 1998	Jwaneng 4	Jwaneng, Botswana	Coated	Unknown	None Distinctive	5.91	0.10	2.130E-06			1.680E-11			-6.6	0.1			



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Burgess <i>et al.,</i> 1998	Jwaneng 6	Jwaneng, Botswana	Coated	Unknown	None Distinctive									-8.2	0.1	
Burgess <i>et al.,</i> 1998	Jwaneng 88	Jwaneng, Botswana	Coated	Unknown	None Distinctive	6.64	0.10	2.600E-07			2.303E-12			-5.3	0.1	
Burgess <i>et al.,</i> 1998	Jwaneng 89	Jwaneng, Botswana	Coated	Unknown	None Distinctive	7.26	0.20	1.540E-06			1.492E-11			-5.6	0.1	
Burgess <i>et al.,</i> 1998	Jwaneng 90	Jwaneng, Botswana	Coated	Unknown	None Distinctive	7.26	0.20	1.860E-06			1.802E-11			-5.4	0.1	
Burgess <i>et al.,</i> 1998	Jwaneng 112	Jwaneng, Botswana	Coated	Unknown	None Distinctive									-5.1	0.1	
Burgess <i>et al.,</i> 1998	DBP 7	DeBeers Pool, South Africa	Cloud	Unknown	None Distinctive	4.67	0.00								0.1	
Burgess <i>et al.,</i> 1998	Finsch 4	Finsch, South Africa	Cloud	Unknown	None Distinctive	3.73	0.30							-4.4	0.1	
Burgess <i>et al.,</i> 1998	Premier 2	Premier, South Africa	Cloud	Unknown	None Distinctive	3.53	0.20							-4.9	0.1	
Burgess <i>et al.,</i> 1998	Premier 4	Premier, South Africa	Cloud	Unknown	None Distinctive									-5.4	0.1	
Burgess <i>et al.,</i> 1998	Premier 8	Premier, South Africa	Cloud	Unknown	None Distinctive	0.31	0.00							-4.8	0.1	
Timmermann et al., 2018	Fin1a	Finsch, South Africa	Coated	Unknown	None Distinctive									-6.3	640	
Timmermann et al., 2018	Fin1b	Finsch, South Africa	Coated	Unknown	None Distinctive			7.310E-07	2.127E-08	1.804E-10				-6.2	750	
Timmermann et al., 2018	Fin02	Finsch, South Africa	Coated	Unknown	None Distinctive	6.94	0.15	4.140E-06	3.304E-07	2.729E-09	3.930E-11	3.136E-12	6.799E-14	-5.1		
Timmermann et al., 2018	Fin03	Finsch, South Africa	Coated	Unknown	None Distinctive	3.37	0.82	3.490E-07	1.661E-08	1.461E-10	1.610E-12	7.664E-14	1.928E-14	-3.4	30	
Timmermann et al., 2018	DBP06	DeBeers Pool, South Africa	Coated	Unknown	None Distinctive	4.85	0.10	6.510E-06	3.242E-07	2.674E-09	4.330E-11	2.156E-12	4.656E-14	-6.7		
Timmermann et al., 2018	DBP07	DeBeers Pool, South Africa	Coated		Saline HDF + Carbonate	3.99	0.08	2.050E-05	7.462E-07	6.188E-09	1.120E-10	4.077E-12	8.736E-14	-4.4	330	
Timmermann et al., 2018	DBP08a	DeBeers Pool, South Africa	Coated	Unknown	None Distinctive	5.34	0.11	1.740E-05	6.577E-07	5.405E-09	1.270E-10	4.801E-12	1.013E-13	-7.1		
Timmermann et al., 2018	DBP08b	DeBeers Pool, South Africa	Coated	Unknown	None Distinctive	5.17	0.15	1.040E-05	1.841E-07	1.512E-09	7.380E-11	1.306E-12	3.859E-14	-7.6		
Timmermann et al., 2018	DBP09a	DeBeers Pool, South Africa	Coated	Unknown	None Distinctive	3.89	0.13	1.660E-05	1.262E-07	1.034E-09	8.810E-11	6.696E-13	2.341E-14	-6.6		
Timmermann et al., 2018	DBP09b	DeBeers Pool, South Africa	Coated	Unknown	None Distinctive	5.07	0.18	7.170E-06	1.542E-07	1.273E-09	4.980E-11	1.071E-12	3.892E-14	-6.4		
Timmermann et al., 2018	DBP09c	DeBeers Pool, South Africa	Coated	Unknown	None Distinctive	4.84	0.12	1.020E-05	3.050E-07	2.509E-09	6.740E-11	2.015E-12	5.173E-14	-6.6		
Timmermann et al., 2018	DBP10a	DeBeers Pool, South Africa	Coated	Lherzolite	Olivine			3.360E-06	4.234E-08	3.843E-10				-5.1	470	



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Timmermann et al., 2018	DBP10b	DeBeers Pool, South Africa	Coated	Lherzolite	Olivine	5.05	0.14	5.700E-06	1.733E-07	1.426E-09	3.940E-11	1.198E-12	3.344E-14	-8.1	520	
Timmermann <i>et al.,</i> 2018	DBP14	DeBeers Pool, South Africa	Coated	Peridotitic & Websteritic	Olivine & Enstatite	5.01	0.10	2.490E-05	1.820E-06	1.499E-08	1.710E-10	1.250E-11	2.646E-13	-6.9		
Timmermann et al., 2018	DBP15a	DeBeers Pool, South Africa	Coated	Peridotitic	Enstatite	2.01	0.04	6.510E-05	2.038E-06	1.681E-08	1.790E-10	5.603E-12	1.233E-13	-5.9		
Timmermann et al., 2018	DBP15b	DeBeers Pool, South Africa	Coated	Peridotitic	Enstatite	4.08	0.09	1.690E-05	4.293E-07	3.531E-09	9.420E-11	2.393E-12	5.461E-14	-6.3		
Timmermann et al., 2018	DBP15c	DeBeers Pool, South Africa	Coated	Peridotitic	Enstatite	4.40	0.10	1.950E-05	3.861E-07	3.168E-09	1.180E-10	2.336E-12	5.366E-14	-5.9		
Timmermann et al., 2018	KOF17a	Koffiefontein, South Africa	Coated		Calcite	4.76	0.26	2.470E-06	3.977E-08	3.284E-10	1.610E-11	2.592E-13	1.481E-14	-4.9	800	
Timmermann et al., 2018	KOF17b	Koffiefontein, South Africa	Coated		Calcite			1.670E-06	5.828E-08	4.816E-10				-4.9	500	
Timmermann et al., 2018	KOB18a	Koffiefontein, South Africa	Coated	Peridotitic	Peridotitic Sulphide	4.97	0.18	1.970E-06	1.178E-07	9.688E-10	1.340E-11	8.013E-13	2.948E-14	-5.4	510	
Timmermann et al., 2018	KOB18b	Koffiefontein, South Africa	Coated	Peridotitic	Peridotitic Sulphide	5.90	0.67	5.460E-07	1.714E-08	1.473E-10	4.410E-12	1.385E-13	1.630E-14	-5.5	450	
Timmermann et al., 2018	KOB19a	Koffiefontein, South Africa	Coated		Saline HDF + Carbonate	5.82	0.25	1.130E-06	4.125E-08	3.405E-10	8.970E-12	3.274E-13	1.467E-14	-6.4	620	
Timmermann et al., 2018	KOF19b	Koffiefontein, South Africa	Coated		Saline HDF + Carbonate	5.22	0.66	2.530E-07	1.369E-08	1.190E-10	1.810E-12	9.792E-14	1.288E-14	-6.4	540	
Timmermann et al., 2019	409b	Jwaneng, Botswana	Coated		Eclogitic	0.16	0.00	6.710E-07	7.059E-08	7.580E-10	1.390E-13	1.461E-14	1.570E-16	-6.8	900	
Timmermann et al., 2019	410A	Jwaneng, Botswana	Coated		Eclogitic	1.92	0.54	8.830E-07	1.678E-08	4.900E-09	2.250E-12	4.283E-14	1.250E-14	-6.2	140	



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Supplementary Information References

Burgess, R., Johnson, L., Mattey, D., Harris, J., Turner, G. (1998) He, Ar and C isotopes in coated and polycrystalline diamonds. Chemical Geology 146, 205–217.

- Gautheron, C., Cartigny, P., Moreira, M., Harris, J., Allegre, C. (2005) Evidence for a mantle component shown by rare gases, C and N isotopes in polycrystalline diamonds from Orapa (Botswana). *Earth and Planetary Science Letters* 240, 559–572.
- Gurney J. J., Boyd F. R. (1982) Mineral intergrowths with polycrystalline diamonds from Orapa Mine, Botswana. Carnegie Institution of Washington Yearbook 81, 267–273.

Jacob, D.E., Viljoen, K.S., Grassineau, N., Jagoutz, E. (2000) Remobilization in the Cratonic Lithosphere Recorded in Polycrystalline Diamond. Science 289, 1182-1185.

Kurat, G., Dobosi, G. (2000) Garnet and diopside-bearing diamondites (framesites). Mineralogy and Petrology 69, 143-159.

- Matsuda, J., Matsumoto, T., Sumino, H., Nagao, K., Yamamoto, J., Miura, Y., Sano, Y. (2002) The 3He/4He ratio of the new internal He Standard of Japan (HESJ). Geochemical Journal 36, 191–195.
- Mikhail, S., Verchovsky, A.B., Howell, D., Hutchison, M.T., Southworth, R., Thomson, A.R., Warburton, P., Jones, A.P., Milledge, H.J. (2014) Constraining the internal variability of the stable isotopes of carbon and nitrogen within mantle diamonds. *Chemical Geology* 366, 14–23.
- Mikhail, S., McCubbin, F.M., Jenner, F.E., Shirey, S.B., Rumble, D., Bowden, R. (2019) Diamondites: Evidence for a distinct tectono-thermal diamond-forming event beneath the Kaapvaal craton. *Contributions to Mineralogy and Petrology* 174, 71.
- Mishima, K., Sumino, H., Yamada, T., Leki, S., Nagakura, N., Hidetoshi, O., Oide, H. (2018) Accurate Determination of the Absolute 3He/4He Ratio of a Synthesized Helium Standard Gas (Helium Standard of Japan, HESJ): Toward Revision of the Atmospheric 3He/4He Ratio. *Geochemistry, Geophysics, Geosystems* 19, 3995-4005.
- Stuart, F.M., Ellam, R.M., Harrop, P.J., Fitton, J.G., Bell, B.R. (2000) Constraints on mantle plumes from the helium isotopic composition of basalts from the British Tertiary Igneous Province. Earth and Planetary Science Letters 177, 273–285.
- Timmermann, S., Honda, M., Phillips, D., Jaques, A.L., Harris, J.W. (2018) Noble gas geochemistry of fluid inclusions in South African diamonds: implications for the origin of diamond-forming fluids. *Mineralogy and Petrology* 112, 181–195.
- Timmermann, S., Yeowa, H., Honda, M., Howell, D., Jaquesa, A.L., Krebs, M.Y., Woodland, S., Pearson, D.G., Ávila, Y.A., Ireland, T.R. (2019) U-Th/He systematics of fluid-rich 'fibrous' diamonds – Evidence for pre- and syn-kimberlite eruption ages. *Chemical Geology* 515, 22–36
- Verchovsky, A.B., Fisenko, A.V., Semjonova, L.F., Wright, I.P., Lee, M.R. and Pillinger, C.T. (1998) C, N, and noble gas isotopes in grain size separates of presolar diamonds from Efremovka. Science 281, 1165–1168.

