Carbonatites, isotopes and evolution of the subcontinental mantle: An overview

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1. Introduction

Carbonatites, made up of at least 50% primary carbonates, are the products of low-degree partial melting of a carbonate-bearing mantle. Found on all continents, carbonatites are well suited for tracking the secular evolution of the sub-continental mantle because they span ages from 3.0 Ga to the present and because they are extremely enriched in Sr, Nd, and other REEs (Bell et al., 1982; Bell and Blenkinsop, 1987a; Nelson et al., 1988). Of more than 500 known carbonatite occurrences, only three are from oceanic islands (Cape Verdes, Canary Islands, and the Kerguelens; Woolley and Kjarsgaard, 2008). This raises questions about the role of continental lithosphere in carbonatite genesis. Are carbonatites formed by partial melting of metasomatized lithosphere? Or does the continental lithosphere simply act as an impermeable barrier trapping uprising volatiles from the asthenosphere below? Globally, the abundance of carbonatite occurrences appears to increase through time, and some parts of the Earth's crust (e.g., the Canadian Shield, the East European Craton, the Tanzanian Craton, and West Greenland) have witnessed repeated carbonatite magmatism over billions of years. This apparent increase in carbonatitic magmatism with time has been attributed either to an increasingly metasomatized mantle by volatiles from below (Woolley, 1989; Blichert-Toft et al., 1996; Woolley, 2003) or it may simply reflect a probable preservation bias (Veizer et al., 1992).

The origin of carbonatites remains controversial. Most carbonatites are spatially associated with much larger volumes of ultramafic and alkaline silicate rocks of similar age, implying petrogenetic relationships between the silica-undersaturated and alkaline silicate, and carbonate magmas. Although still debated, proposed models for the origin of carbonatitic melts include: 1) immiscible separation or fractional crystallization of parental carbonated silicate magmas; and 2) low-degree partial melting of carbonated mantle peridotite below 75 km (see reviews in Bell, 1989; Bell and Rukhlov, 2004). Arguments for origins within the lithosphere are partly based on the repeated intrusion of carbonatites into the same parts of the continental crust (e.g., the Canadian Shield, the East European Craton, the Tanzanian Craton, and West Greenland) over billions of years (Woolley, 1989; Larsen and Rex, 1992; Bailey and Woolley, 1995; Yang and Woolley, 2006; Woolley and Bailey, 2012). An in situ lithospheric source, however, is difficult to reconcile with the primitive isotopic signatures of noble gases (He, Ne, Ar, Kr, and Xe) and nitrogen found in some carbonatites from Brazil, Canada, and Russia, which indicate derivation from a relatively undegassed mantle (Sasada et al., 1997; Marty et al., 1998; Dauphas and Marty, 1999; Tolstikhin et al., 2002). The Sr, Nd, and Pb isotopic compositions of young (<200 Ma) carbonatites worldwide also cover the same range as those of oceanic island basalts (OIBs) involving high-$^{238}U/^{204}Pb$ or μ (HIMU), enriched mantle 1 and 2 (EM1 and EM2), and 'FOcus ZOne' (FOZO) mantle components (e.g., Bell and Simonetti, 1996; Bell and Tilton, 2001). Although several oceanic signatures are associated with carbonatites, it seems that depleted MORB mantle (DMM) played little, if any, role in generating carbonated melts. In many isotope ratio diagrams, particularly $^{87}Sr/^{86}Sr$ vs. $^{206}Pb/^{204}Pb$ and $^{143}Nd/^{144}Nd$ vs. $^{206}Pb/^{204}Pb$, the DMM signature is quite different to any of those involved with mixing, with the result that DMM is spatially isolated from them as well as the mixing patterns (Hoernle and Tilton, 1991; Bell and Simonetti, 1996; Bell and Tilton, 2001, 2002; Ignacio et al., 2006). For an overview of mantle components from oceanic settings, we refer the reader to Hofmann (2014). Isotopic signatures similar to OIBs suggest a sub-lithospheric source for carbonatitic melts and imply generation from mantle plumes or asthenospheric upwellings (e.g., Gerlach et al., 1988; Nelson et al., 1988; Simonetti et al., 1995; 1998; Marty et al., 1998; Dauphas and Marty, 1999; Bell, 2001; Bizzarro et al., 2002; Tolstikhin et al., 2002; Bell and Rukhlov, 2004; Kogarko et al., 2010). Plume-generated carbonatite magmatism is also consistent with the observation that many carbonatites are related to large igneous provinces (LIPs), characterized by high-volume, short-duration, intraplate magmatism including flood basalts and feeder systems manifested by regional radiating mafic dike swarms (e.g., Ernst and Bell, 2010).

Carbonatites provide isotopic insights into mantle evolution not offered by other rocks because they contain extremely high amounts of Sr and Nd, which buffer Sr and Nd isotopic compositions from changes due to contamination, and the fact that they are widely distributed, and span 3 Ga of Earth history. Although some carbonatites have relatively high Hf contents (up to 71 ppm, or ~15 times greater than in continental crust), low values (~5 ppm) are more typical (e.g., Woolley and Kempe, 1989; Bizimis et al., 2003; Chakhmouradian, 2006). Previous studies show that initial Sr, Pb, Nd, and Hf isotope ratios in carbonatites and alkaline rocks from the Canadian and Baltic shields and Greenland trace the evolution of depleted subcontinental mantle over at least 3 Ga (Bell et al., 1982; Bell and Blenkinsop, 1987a; Nelson et al., 1988; Kwon et al., 1989; Tilton and Kwon, 1990; Kramm, 1993; Tilton and Bell, 1994; Rukhlov et al., 2001; Rukhlov and Bell, 2003; Bell and Rukhlov, 2004; Tappe et al., 2007, 2008; Kogarko et al., 2010; Tichomirowa et al., 2006, 2013). Initial $^{86}Sr/^{87}Sr$ and $^{206}Pb/^{204}Pb$ ratios suggest that this widespread, depleted reservoir formed ~3 Ga ago, assuming a reference reservoir of bulk silicate Earth (BSE; DePaolo and Wasserburg, 1976; Kwon et al., 1989). Most Archean carbonatites, however, have positive ε_{Nd} and ε_{Hf} values indicating that depleted mantle existed long before 3 Ga. These data are consistent with a depletion event, recorded in some of the oldest terrestrial materials, that must have taken place during the Hadean (>4 Ga; e.g., Amelin et al., 1999, 2000, 2011; Blichert-Toft and Arndt, 1999; Blichert-Toft et al., 2004; Harrison et al., 2005; Hoffmann et al., 2010; Kemp et al., 2010; Caro, 2011; Puchtel et al., 2013).

Here we re-examine mantle evolution using new Sr, Pb, Nd and Hf isotopic data from several carbonatite occurrences, mainly from the northern hemisphere, along with published global data (Fig. 1). This isotopic evidence lends support to a model in which carbonatite magmas are generated by plumes that originate from a widespread, relatively primitive source in the deep mantle.

2. Samples

Samples for this study were collected from 42 carbonatite complexes worldwide, spanning ages from 3.0 to 0.1 Ga. Most of the complexes are in the Baltic and Canadian shields (Fig. 1, Table 1). We performed whole-rock and mineral fraction analyses, using amphibole, ankerite, apatite, baddeleyite, calcite, dolomite, kimzeyite, siderite, and zircon. Rukhlov and Bell (2010) reported results from most of the samples for U-Th-Pb (apatite, calcite, dolomite, and whole-rock) and U-Pb (baddeleyite, kimzeyite, and zircon).

3. Methods

Rock crushing and mineral separation were carried out in the Department of Earth Sciences at Carleton University in Ottawa (for details, see Rukhlov and Bell, 2010). Fresh rock chips were pulverized in a stainless steel mill to obtain samples for whole-rock analyses. Following density and magnetic separation, pure fractions of mineral grains or fragments were hand-picked under a binocular microscope.

3.1. Rb-Sr, Sm-Nd and U-Th-Pb isotopic analyses

Sr, Nd and Pb isotopic compositions and Rb-Sr, Sm-Nd and U-Th-Pb isotope dilution analyses of carbonates, apatite and whole-rocks were performed in the Department of Earth Sciences at Carleton University. Samples spiked with mixed ^{87}Rb–^{84}Sr, ^{149}Sm–^{145}Nd, and ^{235}U–^{230}Th–^{205}Pb tracers were analyzed for Rb/Sr, Sm/Nd, Th/Pb, U/Pb and Pb isotopic composition using conventional chemical separation techniques. Both Finnigan MAT 261 and Finnigan Triton TI multi-collector thermal ionization mass spectrometers (TIMS) were used, operating in the static mode. Unspiked samples were analyzed for Sr and Nd isotopic compositions using conventional separation techniques. For details of the sample spiking, dissolution, chemical separation, mass spectrometry, and data reduction procedures the reader is referred to Rukhlov and Bell (2010) and Stoppa et al. (2014). The Sr and Nd isotopic ratios were normalized for instrumental mass fractionation relative to $^{88}Sr/^{86}Sr = 8.37500$ and $^{146}Nd/^{144}Nd = 0.7219$, respectively, using an exponential law. The $^{87}Sr/^{86}Sr$ ratios in samples are adjusted to the conventional value of 0.710245 for the NBS 987 standard. The $^{143}Nd/^{144}Nd$ ratios in samples are adjusted to the conventional value of 0.511850 in La Jolla standard. Repeated analysis of NBS 987 and La Jolla standards (over 6 years) yielded $^{87}Sr/^{86}Sr = 0.710283 \pm 0.000073$ and $^{143}Nd/^{144}Nd = 0.511855 \pm 0.000031$, respectively (errors given at the 2σ level). USGS reference material BCR-1 analyzed during this study yielded $^{87}Sr/^{86}Sr = 0.705030 \pm 0.000013$, $^{87}Rb/^{86}Sr = 0.4042 \pm 0.0030$, $^{143}Nd/^{144}Nd = 0.512635 \pm 0.000009$, and $^{147}Sm/^{144}Nd = 0.1384 \pm 0.0003$ (2σ errors, n = 1). The procedural blanks of <7.8 ng Sr, <0.15 ng Rb, <3.0 ng Nd, and <0.29 ng Sm (n = 20) are negligible.

Initial $^{87}Sr/^{86}Sr$ ratios and $\varepsilon_{Sr}(T)$ values were calculated using a decay constant of ^{87}Rb of 1.3968×10^{-11} a^{-1} (Rotenberg et al., 2012) and the bulk Earth values of DePaolo and Wasserburg (1976). Initial $^{143}Nd/^{144}Nd$ ratios and $\varepsilon_{Nd}(T)$ values were calculated using a decay constant of ^{147}Sm of 6.539×10^{-12} a^{-1} (Lugmair and Marti, 1978) and the chondritic uniform reservoir (CHUR; after Jacobsen and Wasserburg, 1980; Hamilton et al., 1983). Initial $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios and $\gamma_{Pb}(T)$ values were calculated using decay constants of ^{238}U of 1.55125×10^{-10} a^{-1} (Jaffey et al., 1971) and ^{232}Th of 4.9475×10^{-11} a^{-1} (Le Roux and Glendenin, 1963) and the bulk silicate Earth (BSE) parameters of Allègre and Lewin (1989).

3.2. Lu-Hf isotopic analyses

Individual grains or fragments of large crystals of baddeleyite, kimzeyite and zircon were analyzed either by isotope dilution or laser ablation, multi-collector inductively coupled plasma mass spectrometer (MC-ICPMS). Using isotope dilution, U-Pb and Lu-Hf were analysed at the Geological Survey of Canada (GSC) in Ottawa. Details of dissolution and the U-Pb isotopic

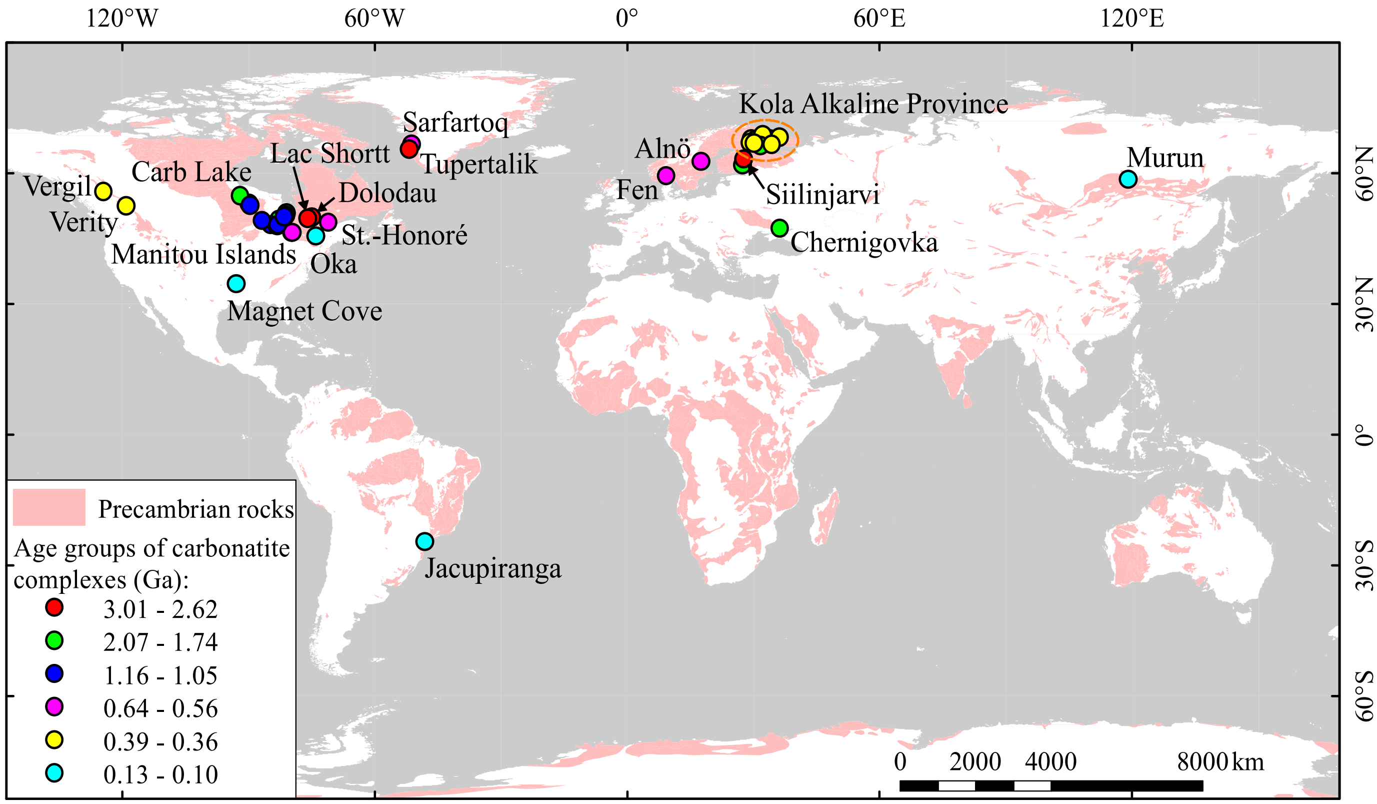

Fig. 1. Exposed Precambrian rocks (after Chorlton, 2007) and location of studied carbonatite complexes.

data can be found in Rukhlov and Bell (2010). The samples were spiked with a mixed ^{176}Lu–^{180}Hf tracer, and Lu and Hf were separated using a standard chemical separation procedure for Zr-bearing minerals modified at the GSC. Hafnium isotopic composition was measured using a Nu Plasma MC-ICPMS with DSN-100 desolvating nebulizer. Lutetium was analyzed on a single rhenium filament with graphite using a Finnigan Triton TI TIMS. The Hf isotopic ratios were normalized for instrumental mass fractionation relative to ^{179}Hf/^{177}Hf = 0.7325 using an exponential law. The ^{176}Hf/^{177}Hf ratios in samples are adjusted to the conventional value of 0.28216 in JMC-475 standard. Repeated analysis (n = 43) of JMC-475 standard yielded ^{176}Hf/^{177}Hf = 0.282164 ± 0.000019, ^{178}Hf/^{177}Hf = 1.46723 ± 0.00006, and ^{180}Hf/^{177}Hf = 1.88629 ± 0.00024 (errors given at the 2σ level). The procedural blanks of <1.3 pg Hf and <0.04 pg Lu (n = 9) are negligible. Details of the Lu-Hf isotope dilution analytical procedures can be found in Amelin et al. (2011). For laser ablation MC-ICPMS analyses, zircon and baddeleyite grains were mounted into polished epoxy mounts and analyzed for Lu-Hf isotopic composition using an Excimer (193 nm) ArF laser ablation coupled with IsoProbe Micromass MC-ICPMS in the Geotop laboratory at the Université du Québec à Montréal (Geotop-UQAM). Zircon reference material 91500 analyzed during this study (n = 13) yielded mean values (±2σ) of ^{176}Hf/^{177}Hf = 0.282288 ± 0.000052, ^{178}Hf/^{177}Hf = 1.46775 ± 0.00031, ^{180}Hf/^{177}Hf = 1.88824 ± 0.00103, ^{176}Lu/^{177}Hf = 0.00033 ± 0.00018, and ^{176}Yb/^{177}Hf = 0.0112 ± 0.0080. Details of the Lu-Hf laser ablation protocol are outlined in Machado and Simonetti (2001).

Initial ^{176}Hf/^{177}Hf ratios and ε_{Hf}(T) values were calculated using a decay constant of ^{176}Lu of 1.867 x 10^{-11} a^{-1} (Söderlund et al., 2004) and the chondritic uniform reservoir (CHUR) of Iizuka et al. (2015). Uncertainties (2σ) of ε_{Hf}(T) values, propagated using the program of Ickert (2013), include errors associated with age, measured ^{167}Lu/^{177}Hf and ^{176}Hf/^{177}Hf ratios, decay constant, and CHUR parameters.

4. Results and discussion

Below we show new and published radiogenic isotope data from carbonatites in a series of isotope ratio diagrams that reflect the isotopic composition of mantle sources as a function of time. We also compare the data from the Kola Alkaline Province, Russia (ca. 380 Ma), one of the largest and among the best studied of alkaline provinces, with global carbonatites with ages of <200 Ma and oceanic mantle components (Table 2) in ε_{Sr}(T) vs. ε_{Nd}(T), ε_{Sr}(T) vs. γ_{Pb}(T), and γ_{Pb}(T) vs. ε_{Nd}(T) diagrams. Because isotope ratios are only strictly comparable for samples of the same age, we use epsilon and gamma values, which are the relative difference in parts per 10^4 (epsilon) and 10^2 (gamma) between a sample and a reference reservoir (i.e. CHUR, BSE) at a given time (e.g., Ickert, 2013), to compare the isotopic signatures for samples of different ages.

4.1. Sr isotopic evolution

Bell et al. (1982) reported initial ^{87}Sr/^{86}Sr ratios in 1.9 to 0.1 Ga carbonatite and syenitic complexes from the Canadian Shield

Table 1. Samples.

Sample	Rock	Fraction	Complex	Age (Ma)	Method	Longitude	Latitude	Location	Provided by	Age reference
MB-8cc	carbonatite	cc (2)	Murun	134	Rb-Sr, Sm-Nd, U-Th-Pb	119.067	58.400	Aldan Shield	A.N. Zaitsev	Makhotkin (1991)
AFR-BMA2	carbonatite	zr (core, rim)	Afrikanda	381	Lu-Hf (LA)	32.800	67.417	Baltic Shield	A.R. Chakhmouradian	Wu et al. (2013)
AFR-ZAZ1	carbonatite	zr	Afrikanda	381	Lu-Hf (LA)	32.800	67.417	Baltic Shield	A.R. Chakhmouradian	Wu et al. (2013)
SOK-1	phoscorite	cc	Sokli	380	Rb-Sr, Sm-Nd, U-Th-Pb	29.450	67.800	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SOK-Stg2	carbonatite	cc	Sokli	380	Rb-Sr, Sm-Nd, U-Th-Pb	29.450	67.800	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SOK-CoreIV	carbonatite	cc-ap	Sokli	380	Rb-Sr, Sm-Nd, U-Th-Pb	29.450	67.800	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SOK-IVmet	silicocarbonatite	ap, bd (3)	Sokli	380	ap Rb-Sr, Sm-Nd, U-Th-Pb; bd Lu-Hf (ID, 2 LA)	29.450	67.800	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SOK-Stg4	carbonatite	cc-ap	Sokli	380	Rb-Sr, Sm-Nd, U-Th-Pb	29.450	67.800	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SV-49/225	carbonatite	sid	Sallanlatvi	372	Rb-Sr, Sm-Nd, U-Th-Pb	29.167	66.950	Baltic Shield	A.N. Pilipiuk	Zaitsev et al. (2004)
SV-50/185	carbonatite	ank	Sallanlatvi	372	Rb-Sr, Sm-Nd, U-Th-Pb	29.167	66.950	Baltic Shield	A.N. Pilipiuk	Zaitsev et al. (2004)
SV-53/190	carbonatite	wr	Sallanlatvi	372	Rb-Sr, Sm-Nd, U-Th-Pb	29.167	66.950	Baltic Shield	A.N. Pilipiuk	Zaitsev et al. (2004)
SV-54/236	carbonatite	wr	Sallanlatvi	372	Rb-Sr, Sm-Nd, U-Th-Pb	29.167	66.950	Baltic Shield	A.N. Pilipiuk	Zaitsev et al. (2004)
SV-56/139.7	carbonatite	wr	Sallanlatvi	372	Rb-Sr, Sm-Nd, U-Th-Pb	29.167	66.950	Baltic Shield	A.N. Pilipiuk	Zaitsev et al. (2004)
N.64.18C	carbonatite	cc	Turiy Peninsula	377	Rb-Sr, Sm-Nd, U-Th-Pb	34.450	66.583	Baltic Shield	E.A. Spencer	Rukhlov and Bell (2010)
C.18.30	phoscorite	cc-ap (2), bd	Turiy Peninsula	377	cc-ap Rb-Sr, Sm-Nd, U-Th-Pb; bd Lu-Hf (ID)	34.450	66.583	Baltic Shield	E.A. Spencer	Rukhlov and Bell (2010)
C.23.300	carbonatite	cc	Turiy Peninsula	377	Rb-Sr, Sm-Nd, U-Th-Pb	34.450	66.583	Baltic Shield	E.A. Spencer	Rukhlov and Bell (2010)
C.XC.5	carbonatite	cc	Turiy Peninsula	377	Rb-Sr, Sm-Nd, U-Th-Pb	34.450	66.583	Baltic Shield	E.A. Spencer	Rukhlov and Bell (2010)
T-19/115	carbonatite	zr (3)	Turiy Peninsula	377	Lu-Hf (ID, 2 LA)	34.450	66.583	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
HCK286/88.5	carbonatite	cc	Vuorijarvi	377	Rb-Sr, Sm-Nd, U-Th-Pb	30.117	66.800	Baltic Shield	A.N. Pilipiuk	Bayanova (2006)

Table 1. Continued.

Sample	Rock	Fraction	Complex	Age (Ma)	Method	Longitude	Latitude	Location	Provided by	Age reference
HCK228/67.5	carbonatite	cc, dol	Vuorijarvi	377	Rb-Sr, Sm-Nd, U-Th-Pb	30.117	66.800	Baltic Shield	A.N. Pilipiuk	Bayanova (2006)
HCK364/73.8	carbonatite	cc	Vuorijarvi	377	Rb-Sr, Sm-Nd, U-Th-Pb	30.117	66.800	Baltic Shield	A.N. Pilipiuk	Bayanova (2006)
KOV-105	carbonatite	cc-ap, bd	Kovdor	379	cc-ap Rb-Sr, Sm-Nd, U-Th-Pb; bd Lu-Hf (ID)	30.483	67.567	Baltic Shield	A.N. Zaitsev	Amelin and Zaitsev (2002)
KOV-127/85	carbonatite	zr	Kovdor	379	Lu-Hf (ID)	30.483	67.567	Baltic Shield	A.N. Zaitsev	Amelin and Zaitsev (2002)
KOV-783/141	carbonatite	ap, cc	Kovdor	379	Rb-Sr, Sm-Nd, U-Th-Pb	30.483	67.567	Baltic Shield	A.N. Zaitsev	Amelin and Zaitsev (2002)
KOV-9/626.7	carbonatite	cc	Kovdor	379	Rb-Sr, Sm-Nd, U-Th-Pb	30.483	67.567	Baltic Shield	A.N. Zaitsev	Amelin and Zaitsev (2002)
KOV-BDT	carbonatite	bd (2)	Kovdor	379	Lu-Hf (LA)	30.483	67.567	Baltic Shield	A.N. Zaitsev	Amelin and Zaitsev (2002)
KZ303/86	volcanic carbonatite	cc	Kontozero	380	Rb-Sr, Sm-Nd, U-Th-Pb	36.117	68.133	Baltic Shield	A.A. Arzamastsev	Arzamastsev and Belyatsky (2000)
KZ320/86	volcanic carbonatite	cc	Kontozero	380	Rb-Sr, Sm-Nd, U-Th-Pb	36.117	68.133	Baltic Shield	A.A. Arzamastsev	Arzamastsev and Belyatsky (2000)
W-440	calcite phonolite	cc	Kandalaksha	380	Rb-Sr, Sm-Nd, U-Th-Pb	32.338	67.098	Baltic Shield	A.S. Rukhlov	Claesson et al. (2000)
P1516	calcite nephelinite	cc	Pinozero	380	Rb-Sr, Sm-Nd, U-Th-Pb	32.499	67.316	Baltic Shield	A.S. Rukhlov	Claesson et al. (2000)
P-409A	alnöite	am	Kandalaksha	380	Rb-Sr, Sm-Nd, U-Th-Pb	32.446	67.124	Baltic Shield	A.S. Rukhlov	Claesson et al. (2000)
P-412	alnöite	wr	Kandalaksha	380	Rb-Sr, Sm-Nd, U-Th-Pb	32.446	67.123	Baltic Shield	A.S. Rukhlov	Claesson et al. (2000)
W-401	aillikite	am	Kandalaksha	380	Rb-Sr, Sm-Nd, U-Th-Pb	32.424	67.129	Baltic Shield	A.S. Rukhlov	Claesson et al. (2000)
W-513A	alnöite	am	Kandalaksha	380	Rb-Sr, Sm-Nd, U-Th-Pb	32.416	67.102	Baltic Shield	A.S. Rukhlov	Claesson et al. (2000)
SB333/581	carbonatite	wr	Sebljavr	378	Rb-Sr, Sm-Nd, U-Th-Pb	32.133	68.717	Baltic Shield	A.N. Pilipiuk	Bayanova (2006)
SB339/363	carbonatite	cc-ap	Sebljavr	378	Rb-Sr, Sm-Nd, U-Th-Pb	32.133	68.717	Baltic Shield	A.N. Pilipiuk	Bayanova (2006)
SB340/297	carbonatite	dol	Sebljavr	378	Rb-Sr, Sm-Nd, U-Th-Pb	32.133	68.717	Baltic Shield	A.N. Pilipiuk	Bayanova (2006)

Table 1. Continued.

Sample	Rock	Fraction	Complex	Age (Ma)	Method	Longitude	Latitude	Location	Provided by	Age reference
SB340/55	carbonatite	wr	Sebljavr	378	Rb-Sr, Sm-Nd, U-Th-Pb	32.133	68.717	Baltic Shield	A.N. Pilipiuk	Bayanova (2006)
SB340/94	carbonatite	cc, dol	Sebljavr	378	Rb-Sr, Sm-Nd, U-Th-Pb	32.133	68.717	Baltic Shield	A.N. Pilipiuk	Bayanova (2006)
931-4	calcite ijolite	wr	Kandaguba	386	Rb-Sr, Sm-Nd	32.145	67.100	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
951-9	calcite ijolite	ap (2), cc (3), wr	Kandaguba	386	wr Rb-Sr, Sm-Nd; ap, cc Rb-Sr, Sm-Nd, U-Th-Pb	32.145	67.100	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
KM-122	carbonatite	wr, cc, dol	Kandaguba	386	wr Rb-Sr, Sm-Nd; cc, dol Rb-Sr, Sm-Nd, U-Th-Pb	32.145	67.100	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
KM-2/13	carbonatite	wr, cc	Kandaguba	386	wr Rb-Sr, Sm-Nd; cc Rb-Sr, Sm-Nd, U-Th-Pb	32.145	67.097	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
KM-3/70.4	carbonatite	wr	Kandaguba	386	Rb-Sr, Sm-Nd	32.145	67.097	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
KM-31	carbonatite	wr (2)	Kandaguba	386	Rb-Sr, Sm-Nd	32.145	67.097	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
KM-74	zircon-apatite vein	ap, zr (4)	Kandaguba	386	ap Rb-Sr, Sm-Nd; zr Lu-Hf (ID, 3 LA)	32.145	67.097	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
KM-78	carbonatite	ank, wr (2)	Kandaguba	386	wr Rb-Sr, Sm-Nd; ank Rb-Sr, Sm-Nd, U-Th-Pb	32.145	67.097	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
KM-8	carbonatite	wr	Kandaguba	386	Rb-Sr, Sm-Nd	32.145	67.097	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
KM-9	carbonatite	wr	Kandaguba	386	Rb-Sr, Sm-Nd	32.145	67.097	Baltic Shield	A.N. Pilipiuk	Rukhlov and Bell (2010)
FE1	carbonatite	dol-ap	Fen	578	Rb-Sr, Sm-Nd, U-Th-Pb	9.283	59.300	Baltic Shield	I. Hornig-Kjarsgaard	Dahlgren (1994)
FE2	carbonatite	cc	Fen	578	Rb-Sr, Sm-Nd, U-Th-Pb	9.283	59.300	Baltic Shield	I. Hornig-Kjarsgaard	Dahlgren (1994)
FE5	carbonatite	ap, cc	Fen	578	Rb-Sr, Sm-Nd, U-Th-Pb	9.283	59.300	Baltic Shield	I. Hornig-Kjarsgaard	Dahlgren (1994)
P2-003	carbonatite	cc (2)	Alnö	583	Rb-Sr, Sm-Nd, U-Th-Pb	17.500	62.500	Baltic Shield	J. Gittins	Rukhlov and Bell (2010)
P1-997	carbonatite	cc	Alnö	583	Rb-Sr, Sm-Nd, U-Th-Pb	17.500	62.500	Baltic Shield	J. Gittins	Rukhlov and Bell (2010)

Table 1. Continued.

Sample	Rock	Fraction	Complex	Age (Ma)	Method	Longitude	Latitude	Location	Provided by	Age reference
P2-036	carbonatite	ap (2), bd (4), cc (3), zr (2)	Alnö	583	ap, cc Rb-Sr, Sm-Nd, U-Th-Pb; bd Lu-Hf (ID, Hf, 2 LA); zr Lu-Hf (LA)	17.500	62.500	Baltic Shield	J. Gittins	Rukhlov and Bell (2010)
P2-037	carbonatite	ap, bd (2), cc (2)	Alnö	583	ap, cc Rb-Sr, Sm-Nd, U-Th-Pb; bd Lu-Hf (ID, LA)	17.500	62.500	Baltic Shield	J. Gittins	Rukhlov and Bell (2010)
H-1E	carbonatite	cc, zr (2)	Halpanen	1792	cc Rb-Sr, Sm-Nd, U-Th-Pb; zr Lu-Hf (ID, LA)	27.433	61.767	Baltic Shield	D.L. Konopelko	Rukhlov and Bell (2010)
H-1S	carbonatite	cc	Halpanen	1792	Rb-Sr, Sm-Nd, U-Th-Pb	27.433	61.767	Baltic Shield	D.L. Konopelko	Rukhlov and Bell (2010)
TSH146/96-107	carbonatite	ap (2), cc	Tiksheozero	1999	Rb-Sr, Sm-Nd, U-Th-Pb	31.667	66.283	Baltic Shield	N.A. Frantz	Corfu et al. (2011)
TSH154/210-220	carbonatite	cc, zr (3)	Tiksheozero	1999	cc Rb-Sr, Sm-Nd, U-Th-Pb; zr Lu-Hf (ID, 2 LA)	31.667	66.283	Baltic Shield	N.A. Frantz	Corfu et al. (2011)
TSH154-15/127.4	carbonatite	cc (2)	Tiksheozero	1999	Rb-Sr, Sm-Nd, U-Th-Pb	31.667	66.283	Baltic Shield	A.N. Pilipiuk	Corfu et al. (2011)
TSH158-25/248.2	carbonatite	cc	Tiksheozero	1999	Rb-Sr, Sm-Nd, U-Th-Pb	31.667	66.283	Baltic Shield	A.N. Pilipiuk	Corfu et al. (2011)
TSH169/142-152	carbonatite	ap (2), dol, zr (2)	Tiksheozero	1999	ap, dol Rb-Sr, Sm-Nd, U-Th-Pb; zr Lu-Hf (ID, LA)	31.667	66.283	Baltic Shield	N.A. Frantz	Corfu et al. (2011)
4-ALV	carbonatite	ap, cc, zr (2)	Chernigovka	2074	ap, cc Rb-Sr, Sm-Nd, U-Th-Pb; zr Lu-Hf (ID, Hf)	36.250	47.233	Ukrainian Shield	V.M. Zagnitko	Rukhlov and Bell (2010)
4-BEF	carbonatite	dol	Chernigovka	2074	Rb-Sr, Sm-Nd, U-Th-Pb	36.250	47.233	Ukrainian Shield	V.M. Zagnitko	Rukhlov and Bell (2010)
4-SOV	carbonatite	ap, cc, zr (2)	Chernigovka	2074	ap, cc Rb-Sr, Sm-Nd, U-Th-Pb; zr Lu-Hf (ID, Hf)	36.250	47.233	Ukrainian Shield	V.M. Zagnitko	Rukhlov and Bell (2010)
SIL101	carbonatite	ap, cc	Siilinjarvi	2617	Rb-Sr, Sm-Nd, U-Th-Pb	27.733	63.133	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SIL102	carbonatite	zr (4)	Siilinjarvi	2617	Lu-Hf (ID, 3 LA)	27.733	63.133	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SIL103	carbonatite	cc	Siilinjarvi	2617	Rb-Sr, Sm-Nd, U-Th-Pb	27.733	63.133	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SIL104	carbonatite	cc (2)	Siilinjarvi	2617	Rb-Sr, Sm-Nd, U-Th-Pb	27.733	63.133	Baltic Shield	K. Bell	Rukhlov and Bell (2010)

Table 1. Continued.

Sample	Rock	Fraction	Complex	Age (Ma)	Method	Longitude	Latitude	Location	Provided by	Age reference
SIL106	carbonatite	ap (2), cc (2)	Siilinjarvi	2617	Rb-Sr, Sm-Nd, U-Th-Pb	27.733	63.133	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SIL2	carbonatite	zr	Siilinjarvi	2617	Lu-Hf (ID)	27.733	63.133	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
SIL2000	carbonatite	cc	Siilinjarvi	2617	Rb-Sr, Sm-Nd, U-Th-Pb	27.733	63.133	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
ST2800	carbonatite	cc	Siilinjarvi	2617	Rb-Sr, Sm-Nd, U-Th-Pb	27.733	63.133	Baltic Shield	K. Bell	Rukhlov and Bell (2010)
MC-1	carbonatite	kz	Magnet Cove	95	Lu-Hf (ID)	-92.867	34.450	Gulf Coastal Plain	M. Howard	Baksi (1997)
MC-115	carbonatite	ap	Magnet Cove	95	Sr	-92.867	34.450	Gulf Coastal Plain	M. Howard	Baksi (1997)
CH-T2	carbonatite	ap	Oka	109	Sr	-74.000	45.500	St. Lawrence Platform	K. Bell	Wen et al. (1987)
J-14	carbonatite	ap, bd	Jacupiranga	131	ap Sr; bd Lu-Hf (ID)	-48.133	-24.700	Parana Basin	R.O.M.	Roden et al. (1985)
J-4	carbonatite	bd	Jacupiranga	131	Lu-Hf (ID)	-48.133	-24.700	Parana Basin	R.O.M.	Roden et al. (1985)
L4-242	carbonatite	ap, zr (4)	Vergil	352	ap Sr; zr Lu-Hf (ID, 3 LA)	-124.417	55.717	Canadian Cordillera	J. Pell	Pell (1994)
4-470	carbonatite	zr (4)	Verity	350	Lu-Hf (ID, 3 LA)	-119.150	52.400	Canadian Cordillera	J. Pell	Rukhlov and Bell (2010)
BI-101	carbonatite	ap	Burritt Island	568	Sr	-79.750	46.250	Canadian Shield	K. Bell	Rukhlov and Bell (2010)
C-6	carbonatite	zr (2)	Calder Island	568	Lu-Hf (ID, Hf)	-79.583	46.250	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
CI-100	carbonatite	ap	Calder Island	568	Sr	-79.583	46.250	Canadian Shield	K. Bell	Rukhlov and Bell (2010)
II-102	carbonatite	ap (2)	Iron Island	568	Sr	-79.750	46.250	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
NI-PYR	carbonatite	zr (2)	Newman Island	568	Lu-Hf (ID, Hf)	-79.583	46.250	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
R-4DYKE	carbonatite	zr	Rankin Island	568	Lu-Hf (ID)	-79.583	46.250	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
STH-104	carbonatite	ap, zr (4)	St. Honoré	571	ap Sr; zr Lu-Hf (LA)	-71.067	48.550	Canadian Shield	K. Bell	McCausland et al. (2009)
STH-105	carbonatite	zr	St. Honoré	571	Lu-Hf (ID)	-71.067	48.550	Canadian Shield	K. Bell	McCausland et al. (2009)
STH-18	carbonatite	zr	St. Honoré	571	Lu-Hf (ID)	-71.067	48.550	Canadian Shield	K. Bell	McCausland et al. (2009)

Table 1. Continued.

Sample	Rock	Fraction	Complex	Age (Ma)	Method	Longitude	Latitude	Location	Provided by	Age reference
SR-18-10	carbonatite	bd	Schryburt Lake	1083	Lu-Hf (ID)	-89.600	52.617	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
SR-18-9	carbonatite	bd	Schryburt Lake	1083	Lu-Hf (ID)	-89.600	52.617	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
LAC5	carbonatite	ap	Lackner Lake	1101	Sr	-83.167	47.750	Canadian Shield	K. Bell	Heaman and Machado (1992)
BB-35	silicocarbonatite	ap, bd	Big Beaver House	1093	ap Sr; bd Lu-Hf (ID)	-89.917	52.917	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
NL203A	carbonatite	ap, zr (2)	Nemegosenda Lake	1105	ap Sr; zr Lu-Hf (LA)	-83.083	48.000	Canadian Shield	K. Bell	Heaman and Machado (1992)
V22/2269.0	carbonatite	bd (7)	Valentine Township	1106	Lu-Hf (ID) - bomb versus hot-plate dissolution experiments	-81.500	50.000	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
V23/1410.3	carbonatite	bd	Valentine Township	1106	Lu-Hf (ID)	-81.500	50.000	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
H8/298.0	silicocarbonatite	cc, kz	Firesand River	1143	cc Sr; kz Lu-Hf (ID)	-84.667	48.000	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
DP-26F	silicocarbonatite	zr (2)	Prairie Lake	1164	Lu-Hf (ID, Hf)	-86.717	49.033	Canadian Shield	K. Bell	Rukhlov and Bell (2010)
P21A/64.8	phoscorite	ap, bd (2)	Prairie Lake	1164	ap Sr; bd Lu-Hf (ID, Hf)	-86.717	49.033	Canadian Shield	K. Bell	Rukhlov and Bell (2010)
A8/889.0	carbonatite	zr	Argor	1769	Lu-Hf (ID)	-81.017	50.750	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
AB3/792.3	carbonatite	ap, zr	Argor	1769	ap Sr; zr Lu-Hf (ID)	-81.017	50.750	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
3-53-82B	carbonatite	zr (2)	"Carb" Lake	1865	Lu-Hf (ID, Hf)	-92.000	54.800	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
H2/72.5	carbonatite	zr (2)	"Carb" Lake	1865	Lu-Hf (ID, Hf)	-92.000	54.800	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
H2-61	carbonatite	ap	"Carb" Lake	1865	Sr	-92.000	54.800	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
TP109-108/1720.8	carbonatite	bd (2)	Spanish River	1881	Lu-Hf (ID, Hf)	46.583	81.717	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
BO-200	carbonatite	zr	Borden	1882	Lu-Hf (LA)	-83.183	47.917	Canadian Shield	K. Bell	Rukhlov and Bell (2010)
BO-203	carbonatite	ap	Borden	1882	Sr	-83.183	47.917	Canadian Shield	K. Bell	Rukhlov and Bell (2010)

Table 1. Continued.

Sample	Rock	Fraction	Complex	Age (Ma)	Method	Longitude	Latitude	Location	Provided by	Age reference
BO-205	carbonatite	zr (3)	Borden	1882	Lu-Hf (ID, 2 LA)	-83.183	47.917	Canadian Shield	K. Bell	Rukhlov and Bell (2010)
G1A/590.0	carbonatite	zr	Goldray	1886	Lu-Hf (ID)	-81.167	50.217	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
G2/1201.4	carbonatite	ap	Goldray	1886	Sr	-81.167	50.217	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
CCM-26/195.85	carbonatite	ap, bd	Cargill Township	1897	ap Sr; bd Lu-Hf (ID)	-82.817	49.317	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
CCM-6/140.1	silicocarbonatite	zr	Cargill Township	1897	Lu-Hf (ID)	-82.817	49.317	Canadian Shield	R.O.M.	Rukhlov and Bell (2010)
DOD-77	carbonatite	ap, zr (5)	Dolodau Dike	2680	ap Sr; Lu-Hf (ID, 4 LA)	-75.000	49.767	Canadian Shield	K. Bell	Tilton and Bell (1994)
DOD-91	silicocarbonatite	zr (2)	Dolodau Dike	2680	Lu-Hf (ID, LA)	-75.000	49.767	Canadian Shield	K. Bell	Tilton and Bell (1994)
LSC65	carbonatite	ap, zr (2)	Lac Shortt	2691	ap Sr; zr Lu-Hf (ID, LA)	-75.883	49.583	Canadian Shield	K. Bell	Dion et al. (1995)
LSC7910	carbonatite	zr (2)	Lac Shortt	2691	Lu-Hf (ID, LA)	-75.883	49.583	Canadian Shield	K. Bell	Dion et al. (1995)
LSC7912	carbonatite	ap, zr (3)	Lac Shortt	2691	ap Sr; zr Lu-Hf (ID, 2 LA)	-75.883	49.583	Canadian Shield	K. Bell	Dion et al. (1995)
S-10	carbonatite	wr (2)	Sarfartoq	591	Rb-Sr	-51.250	66.500	Greenland	A. Simonetti	Bizzarro et al. (2002)
T-1	carbonatite	wr	Tupertalik	3007	Rb-Sr	-51.750	65.417	Greenland	A. Simonetti	Bizzarro et al. (2002)

Fraction abbreviations: ap - apatite, am - hornblende phenocryst, ank - ankerite, bd - baddeleyite, cc - calcite, dol - dolomite, kz - kimzeyite, sid - siderite, zr - zircon, wr - whole rock. Number in parentheses refers to number of analyzed fractions.

Methods: Rb-Sr - Rb/Sr by isotope dilution and unpsiked Sr isotopic composition by TIMS, Sr - unspiked Sr isotopic composition by TIMS, Sm-Nd - Sm/Nd by isotope dilution and unspiked Nd isotopic composition by TIMS, Lu-Hf ID - Lu/Hf by isotope dilution and Hf isotopic composition by solution MC-ICP-MS and Lu by TIMS, LA - Lu/Hf and Hf isotopic composition by laser ablation MC-ICP-MS, Hf - unspiked Hf isotopic composition by solution MC-ICP-MS, U-Th-Pb - U/Pb and Th/Pb by isotope dilution and Pb isotopic composition by TIMS.

R.O.M. - Royal Ontario Museum.

Table 2. Isotopic compositions of mantle components (after Hart et al., 1992; Stracke et al., 2005; Stracke, 2012).

Component	$^{87}Sr/^{86}Sr$	$^{143}Nd/^{144}Nd$	$^{176}Hf/^{177}Hf$	$^{206}Pb/^{204}Pb$	$^{207}Pb/^{204}Pb$	$^{208}Pb/^{204}Pb$	$\varepsilon_{Sr}(0)$	$\varepsilon_{Nd}(0)$	$\varepsilon_{Hf}(0)$	$\gamma_{Pb}(0)$
DMM	0.7022	0.51335	0.28350	17.5	15.35	36.8	-32.6	13.9	25.0	-4.6
EM1	0.7053	0.51234	0.28263	17.4	15.47	39.0	11.4	-5.8	-5.8	-5.1
EM2	0.7078	0.51258	0.28288	19.3	15.64	39.8	46.8	-1.1	3.1	5.2
FOZO minimum	0.7030	0.51288	0.28295	19.4	15.57	39.2	-21.3	4.7	5.6	5.8
FOZO maximum	0.7033	0.51305	0.28315	20.5	15.70	39.8	-17.0	8.0	12.6	11.8
HIMU	0.7028	0.51290	0.28296	21.8	15.85	40.8	-24.1	5.1	5.9	18.9

$\varepsilon_{Sr}(0) = [(^{87}Sr/^{86}Sr_{sample}/^{87}Sr/^{86}Sr_{BE}) - 1]*10^4$, where $^{87}Sr/^{86}Sr_{sample}$ is the present-day ratio in the sample and $^{87}Sr/^{86}Sr_{BE}$ is the present-day ratio in the bulk Earth (after DePaolo and Wasserburg, 1976).
$\varepsilon_{Nd}(0) = [(^{143}Nd/^{144}Nd_{sample}/^{143}Nd/^{144}Nd_{CHUR}) - 1]*10^4$, where $^{143}Nd/^{144}Nd_{sample}$ is the present-day ratio in the sample and $^{143}Nd/^{144}Nd_{CHUR}$ is the present-day ratio in the chondritic uniform reservoir (CHUR; after Jacobsen and Wasserburg, 1980; Hamilton et al., 1983).
$\varepsilon_{Hf}(0) = [(^{176}Hf/^{177}Hf_{sample}/^{176}Hf/^{177}Hf_{CHUR}) - 1]*10^4$, where $^{176}Hf/^{177}Hf_{sample}$ is the present-day ratio in the sample and $^{176}Hf/^{177}Hf_{CHUR}$ is the present-day ratio in CHUR (after Iizuka et al., 2015).
$\gamma_{Pb}(0) = [(^{206}Pb/^{204}Pb_{sample}/^{206}Pb/^{204}Pb_{BSE}) - 1]*10^2$, where $^{206}Pb/^{204}Pb_{sample}$ is the present-day ratio in the sample and $^{206}Pb/^{204}Pb_{BSE}$ is the present-day ratio in the bulk silicate Earth (BSE; after Allègre and Lewin, 1989).
DMM = depleted MORB mantle, EM1 = enriched mantle 1, EM2 = enriched mantle 2, FOZO = 'FOcus ZOne', HIMU = high-$^{238}U/^{204}Pb$ or μ.

that indicate a depleted mantle source, and using the bulk Earth reservoir (DePaolo and Wasserburg, 1976), determined a model age of ~3 Ga for depletion. This model age corresponds to the age of a depleted, closed-system reservoir formed by either the extraction of a significant volume of continental crust in the Neoarchean or a major change in the mode of heat transfer in the upper mantle at that time (Bell et al., 1982; Bell and Blenkinsop, 1987a). Close agreement between the data from the Baltic and Canadian shields suggests similar differentiation histories for mantle sources below both of these regions (e.g., Tilton and Kwon, 1990; Kramm, 1993; Tilton and Bell, 1994; Rukhlov et al., 2001; Bell and Rukhlov, 2004). Nelson et al. (1988) also noted similar evolution for mantle sources of global carbonatites. An alternative model proposed by Tichomirowa et al. (2006) involves a continuous decrease in Rb/Sr for the depleted mantle reservoir beneath the Baltic Shield.

Figure 2 shows initial $^{87}Sr/^{86}Sr$ ratios for global carbonatites as a function of age. Two important features emerge from Figure 2. First, the spread of the isotopic data suggests involvement of at least two distinct mantle sources, one depleted and one enriched, with intermittent mixing between the two over a period of 3 Ga. This is particularly well seen in the range of values from the Kola carbonatites (ca. 380 Ma; e.g., Kramm, 1993; Zaitsev and Bell, 1995; Beard et al., 1996; Arzamastsev et al., 1998; Rukhlov, 1999; Verhulst et al., 2000; Dunworth and Bell, 2001; Zaitsev et al., 2002; Bell and Rukhlov, 2004; Sindern et al., 2004; Lee et al., 2006; Balaganskaya et al., 2007; Kogarko et al., 2010; Mitchell et al., 2011; Wu et al., 2013; Zartman and Kogarko, 2014) and young carbonatites from elsewhere (e.g., Bell and Blenkinsop, 1987b; Gerlach et al., 1988; Nelson et al., 1988; Kogarko, 1993; Simonetti and Bell, 1994a, b; Bell and Dawson, 1995; Simonetti et al., 1995, 1998; Bell and Simonetti, 1996; Bell and Tilton, 2001; Hoernle et al., 2002). Second, the lowest $^{87}Sr/^{86}Sr$ values fall along a single development line that corresponds to a Rb/Sr ratio of 0.02, which is quite different from that of BSE (0.03) and consistent with the findings of Bell et al. (1982). This widespread depleted source appears to have behaved as a relatively closed system that retained its Rb/Sr ratio over long time periods, either by preservation in the lithosphere or at deeper levels in the mantle, isolated from mantle convection. Lithospheric residence seems unlikely because it implies synchronous metasomatism of the lithosphere on a global scale by an unknown mantle process. A continuous transport-depletion model (e.g., Hart and Brooks, 1970; Ben Othman et al., 1984) based on the data from MORB, ophiolites, komatiites, and meteorites implies much lower Rb/Sr ratio than that of the depleted carbonatite source. This depleted reservoir, indicated by the lowest $^{87}Sr/^{86}Sr$ values in carbonatites, has a present-day value that is quite different in its isotopic composition to that of DMM (see Bell and Tilton, 2001, 2002) and similar to FOZO, an end-member identified in OIBs, oceanic plateaus, flood basalts, and several young (<0.2 Ga) carbonatites. FOZO is considered to be relatively primitive and of deep mantle origin (Hart et al., 1992; Hauri et al., 1994; Bell and Tilton, 2002; Campbell and O'Neill, 2012) and appears to be a common source component for the 380 Ma, plume-related Kola carbonatites based on the Sr-Pb-Nd isotopic evidence (Fig. 3; e.g., Kramm, 1993; Dunworth and Bell, 2001; Bell and Rukhlov, 2004; this study).

The initial isotopic ratios from Kola cannot be directly compared with the present-day mantle components because the isotopic compositions of the latter 380 Ma ago are unknown. However, because the Sr isotopic composition of the depleted source appears to have changed little over the last 380 Ma (Fig. 2), we assume that the Kola and young carbonatites share the same reference reservoir in Sr-Pb-Nd isotope space. In addition, overlapping of the mixing patterns of the Kola and young carbonatites in $\varepsilon_{Sr}(T)$ vs. $\varepsilon_{Nd}(T)$, $\varepsilon_{Sr}(T)$ vs. $\gamma_{Pb}(T)$, and $\gamma_{Pb}(T)$ vs. $\varepsilon_{Nd}(T)$ diagrams (Fig. 3) indicates involvement of the same mantle components, one depleted (FOZO) and the other enriched (EM1). The Sr-Pb-Nd isotopic compositions of some young (<200 Ma) carbonatites also indicate involvement of HIMU and EM1 (Fig. 3; e.g., Simonetti and Bell, 1994a; Bell and Dawson, 1995; Bell and Simonetti, 1996; Bell and Tilton, 2001). In Figure 3, DMM is spatially isolated from the mixing patterns in all of the diagrams and thus played little, if any, role in generating the carbonated melts (Hoernle and Tilton, 1991; Bell and Simonetti, 1996; Bell and Tilton, 2001, 2002; Ignacio et al., 2006). An enriched end-member, 'ITEM' (Italian enriched mantle), marks the most radiogenic compositions of Cenozoic carbonatites from Italy ($^{87}Sr/^{86}Sr$ up to 0.71077; Bell et al., 2013) and, on the basis of our data, seems to have also been tapped intermittently over a period of 3 Ga. It could represent either old recycled crust and/or sediment entrained in the upwelling FOZO mantle, or C–H–O–K-rich fluids released from the core-mantle boundary during plume activity, triggered by core-mantle perturbations (Vidale and Hedlin, 1998; Bell et al., 2013; Herzberg et al., 2013). Very radiogenic $^{87}Sr/^{86}Sr$ ratios (>0.708) in separated carbonate fractions, coupled with much lower $^{87}Sr/^{86}Sr$ ratios (<0.705) in whole rock fractions from some late-stage carbonatites at Kandaguba complex, Kola Alkaline Province (Fig. 3), indicate isotopic disequilibrium resulting from complex petrogenetic processes (Bulakh et al., 2000; Pilipiuk et al., 2001).

4.2. Pb isotopic evolution

Tilton (1983) reported a major Neoarchean depletion event for both the Canadian and Baltic shields based on Pb isotopic data from granitic rocks, sulphide ores, komatiites, and carbonatites. Subsequent work by Kwon et al. (1989) and Tilton and Kwon (1990) determined a model age of ~3 Ga relative to BSE using initial Pb isotopic ratios from carbonatites and alkaline-silicate rocks with ages between 2.7 and 0.1 Ga from the Canadian Shield. These ratios converge with BSE at ~3 Ga, supporting the widespread Neoarchean differentiation event recorded by the Sr data (Tilton and Kwon, 1990).

Figure 4 shows initial $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios for carbonatites worldwide and some 2.7 Ga syenites from the Canadian Shield plotted as a function of time. Evolution curves show the different BSE models after Galer and Goldstein

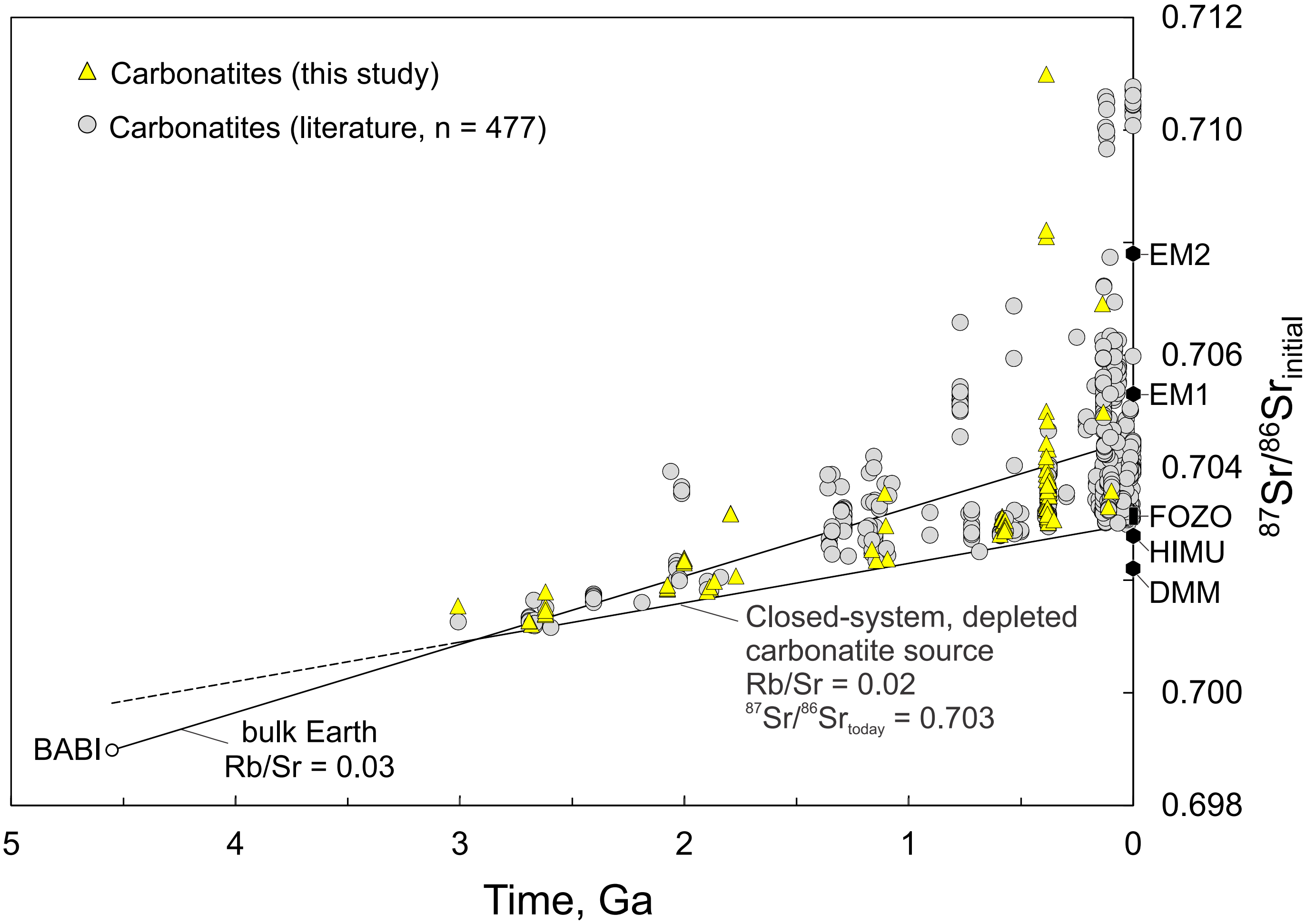

Fig. 2. Initial $^{87}Sr/^{86}Sr$ versus time for carbonatites worldwide and late Archean syenitic complexes from the Canadian Shield. Evolution curve for bulk Earth (after DePaolo and Wasserburg, 1976) assuming primordial $^{87}Sr/^{86}Sr = 0.6990$ of basaltic achondrite best initial (BABI). Depleted MORB mantle (DMM), enriched mantle 1 and 2 (EM1 and EM2), 'FOcus ZOne' (FOZO), and high-$^{238}U/^{204}Pb$ or μ (HIMU) mantle components after Hart et al. (1992), Stracke et al. (2005), and Stracke (2012). The development line indicates the presence of an ancient, depleted mantle reservoir at least 3.0 Ga old. Data from Alberti et al. (1999), Andersen (1987, 1997), Baksi (1997), Balaganskaya et al. (2000), Barreiro and Cooper (1987), Beard et al. (1996), Bell and Blenkinsop (1987a, b), Bell and Peterson (1991), Bell and Tilton (2001), Bell et al. (1987, 2013), Bernard-Griffiths et al. (1991), Bizimis et al. (2003), Bizzarro et al. (2001, 2002, 2003), Blaxland et al. (1978), Castorina et al. (1996), Cavell and Baadsgaard (1986), Comin-Chiaramonti et al. (2002), Conticelli et al. (1995, 2002), Cooper and Mellish (2001), Cooper and Reid (1998), Corfu and Noble (1992), Coulson et al. (2003), Dawson et al. (1995), Dunai et al. (1989), Dunworth and Bell (2001), Eby et al. (1995), Eriksson (1989), Grüenenfelder et al. (1986), Hansen (1981), Harmer (1985, 1999), Harmer et al. (1998), Hoernle and Tilton (1991), Hoernle et al. (2002), Huang et al. (1995), Kalt et al. (1997), Kampunzu et al. (1998), Keller and Krafft (1990), Kramm (1993), Kramm and Kogarko (1994), Kramm et al. (1997), Kumar et al. (1998), Kwon et al. (1989), Lancelot and Allègre (1974), le Roex and Lanyon (1998), Liegeois et al. (1991), Melluso et al. (2004), Middlemost (1990), Mitchell et al. (1994), Morisset (1992), Mourtada et al. (1997), Natarajan et al. (1994), Nelson et al. (1988), Nielsen and Buchardt (1985), Pandit et al. (2002), Paslick et al. (1995), Pearce and Leng (1996), Pollock (1987), Ray et al. (2000), Reischmann (1995), Ruberti et al. (1997, 2002), Savatenkov et al. (1999), Schleicher et al. (1990, 1991, 1998), Schultz et al. (2004), Silva et al. (1988), Simonetti and Bell (1994a, b), Simonetti et al. (1995, 1998), Smithies and Marsh (1998), Srivastava et al. (2005), Sun et al. (1986), Taubald et al. (2004), Thompson et al. (2002), Tilton and Bell (1994), Tilton et al. (1987, 1998), Toyoda et al. (1994), Veena et al. (1998), Verhulst et al. (2000), Verwoerd et al. (1993), Villeneuve and Relf (1998), Wagner et al. (2003), Waight et al. (2002), Walter et al. (1995), Wen et al. (1987), Ying et al. (2004), Zaitsev and Bell (1995), Zaitsev et al. (2002), Ziegler (1992), and references in Table 1.

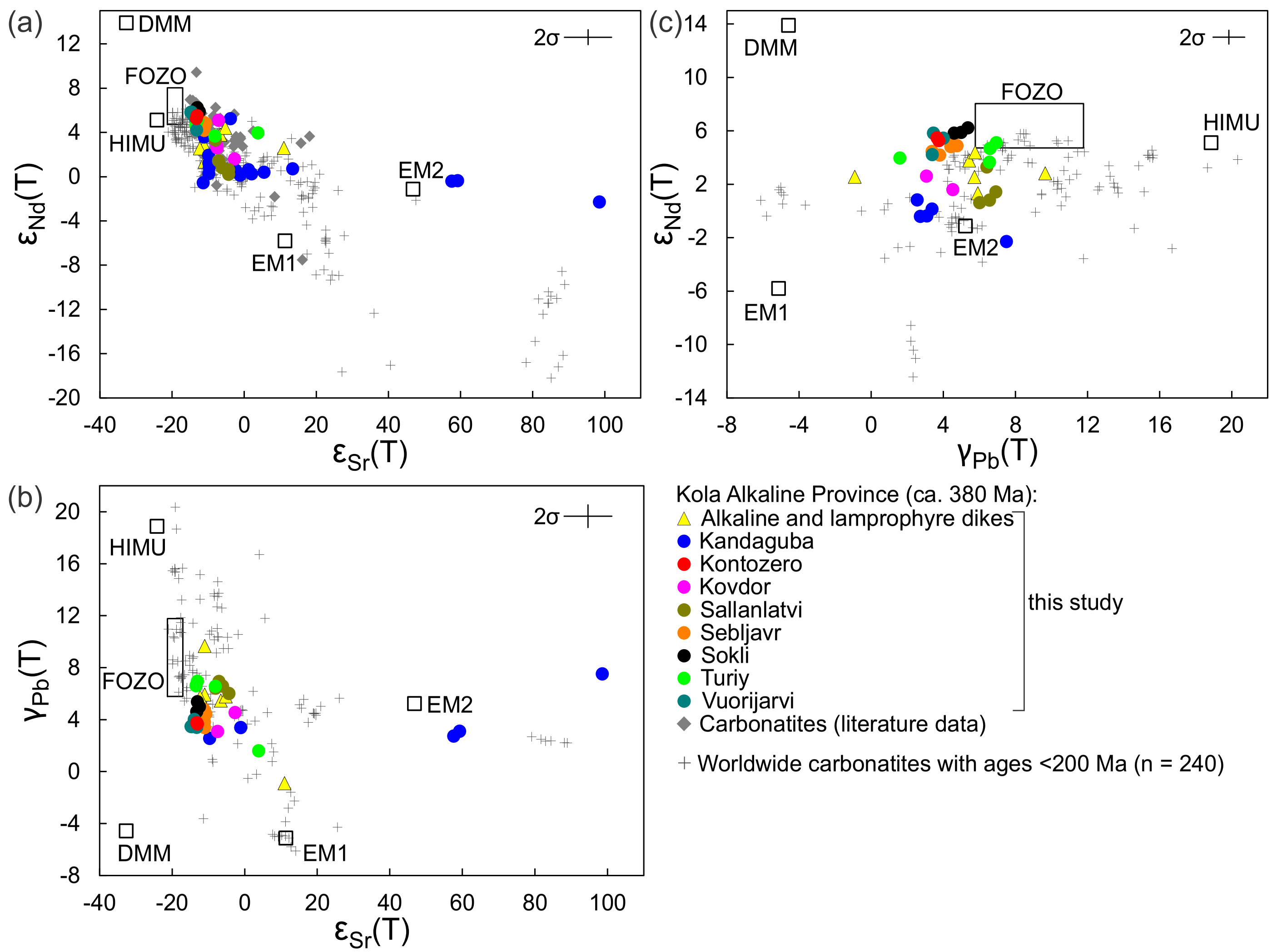

Fig. 3. Sr-Pb-Nd isotope correlation diagrams for carbonatite and alkaline complexes of the Kola alkaline province (380 Ma) in the Baltic Shield. **a)** $\varepsilon_{Sr}(T)$ versus $\varepsilon_{Nd}(T)$. **b)** $\varepsilon_{Sr}(T)$ versus $\gamma_{Pb}(T)$. **c)** $\gamma_{Pb}(T)$ versus $\varepsilon_{Nd}(T)$. $\varepsilon_{Sr}(T) = [(^{87}Sr/^{86}Sr_{sample}/^{87}Sr/^{86}Sr_{BE}) - 1]*10^4$, where $^{87}Sr/^{86}Sr_{sample}$ is the initial ratio in the sample and $^{87}Sr/^{86}Sr_{BE}$ is the ratio in the bulk Earth (after DePaolo and Wasserburg, 1976) at that time; $\gamma_{Pb}(T) = [(^{206}Pb/^{204}Pb_{sample}/^{206}Pb/^{204}Pb_{BSE}) - 1]*10^2$, where $^{206}Pb/^{204}Pb_{sample}$ is the initial ratio in the sample and $^{206}Pb/^{204}Pb_{BSE}$ is the ratio in the bulk silicate Earth (BSE; after Allègre and Lewin, 1989) at that time; $\varepsilon_{Nd}(T) = [(^{143}Nd/^{144}Nd_{sample}/^{143}Nd/^{144}Nd_{CHUR}) - 1]*10^4$, where $^{143}Nd/^{144}Nd_{sample}$ is the initial ratio in the sample and $^{143}Nd/^{144}Nd_{CHUR}$ is the ratio in the chondritic uniform reservoir (CHUR; after Jacobsen and Wasserburg, 1980; Hamilton et al., 1983) at that time. Depleted MORB mantle (DMM), enriched mantle 1 and 2 (EM1 and EM2), 'FOcus ZOne' (FOZO), and high-$^{238}U/^{204}Pb$ or μ (HIMU) mantle components after Hart et al. (1992), Stracke et al. (2005), and Stracke (2012). Data from Beard et al. (1996), Dunworth and Bell (2001), Kramm (1993), Rukhlov (1999), Verhulst et al. (2000), Zaitsev and Bell (1995), and Zaitsev et al. (2002). Also shown are data from the <200 Ma carbonatites worldwide (after Alberti et al., 1999; Barreiro and Cooper, 1987; Bell and Blenkinsop, 1987b; Bell and Peterson, 1991; Bell and Tilton, 2001; Bell et al., 2013; Bernard-Griffiths et al., 1991; Bizimis et al., 2003; Bizzarro et al., 2001; Castorina et al., 1996; Comin-Chiaramonti et al., 2002; Conticelli et al., 1995; Cooper and Reid, 1998; Hoernle and Tilton, 1991; Hoernle et al., 2002; Huang et al., 1995; Kalt et al., 1997; Keller and Krafft, 1990; Kwon et al., 1989; le Roex and Lanyon, 1998; Melluso et al., 2004; Mitchell et al., 1994; Morisset, 1992; Nelson et al., 1988; Paslick et al., 1995; Ruberti et al., 1997, 2002; Schleicher et al., 1991; Schultz et al., 2004; Simonetti and Bell, 1994a, b; Simonetti et al., 1995, 1998; Srivastava et al., 2005; Tilton et al., 1987, 1998; Toyoda et al., 1994; Veena et al., 1998; Wagner et al., 2003; Walter et al., 1995; Wen et al., 1987; Ying et al., 2004; Ziegler, 1992).

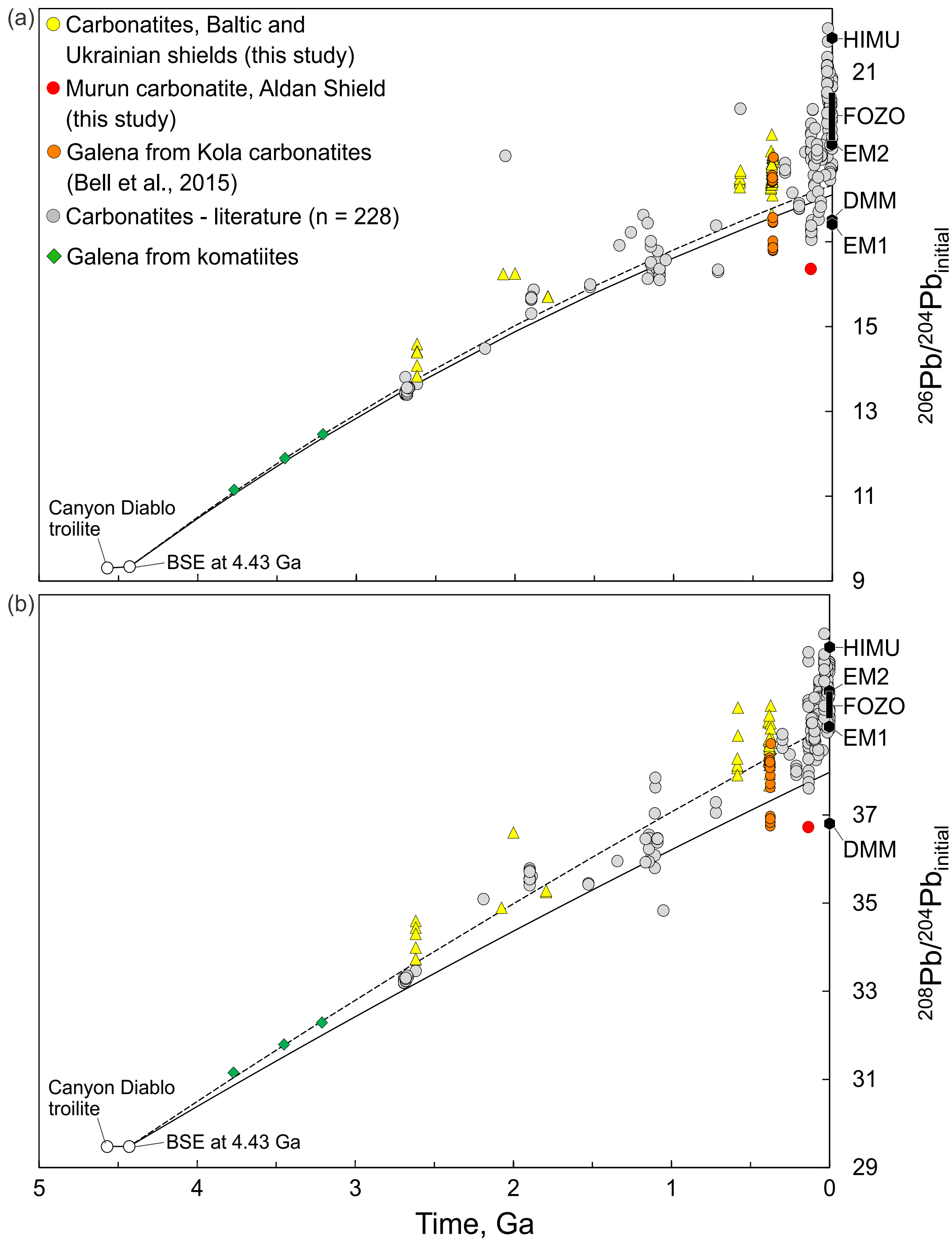

Fig. 4. Pb isotope development diagrams for carbonatites and alkaline complexes worldwide. **a)** Initial $^{206}Pb/^{204}Pb$ versus time. **b)** Initial $^{208}Pb/^{204}Pb$ versus time. BSE = bulk silicate Earth models (solid line after Galer and Goldstein, 1996; dashed line, Allègre and Lewin, 1989) assuming closed-system evolution from 4.43 Ga (Doe and Stacey, 1974; Wood et al., 2008). Depleted MORB mantle (DMM), enriched mantle 1 and 2 (EM1 and EM2), 'FOcus ZOne' (FOZO), and high-$^{238}U/^{204}Pb$ or μ (HIMU) mantle components after Hart et al. (1992), Stracke et al. (2005), and Stracke (2012). Initial Earth $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios taken from Canyon Diablo troilite values (Tatsumoto et al., 1973) and $^{238}U/^{204}Pb$ = 0.7 at 4.57 Ga (after Allègre et al., 1995). Data from Barreiro and Cooper (1987), Bell and Tilton (2001), Bell et al. (1987, 2013), Bizimis et al. (2003), Cavell and Baadsgaard (1986), Corfu and Noble (1992), Dawson et al. (1995), Eby et al. (1995), Eriksson (1989), Grüenenfelder et al. (1986), Harmer et al. (1998), Hoernle and Tilton (1991), Hoernle et al. (2002), Huang et al. (1995), Kalt et al. (1997), Kramm and Koark (1988), Kramm and Kogarko (1994), Kwon et al. (1989), Lancelot and Allègre (1974), le Roex and Lanyon (1998), Nelson et al. (1988), Paslick et al. (1995), Ray et al. (2000), Reischmann (1995), Rukhlov and Bell (2010), Schleicher et al. (1991), Simonetti and Bell (1994a, b), Simonetti et al. (1995, 1998), Stendal et al. (2004), Thompson et al. (2002), Tilton and Bell (1994), Tilton et al. (1987, 1998), Toyoda et al. (1994), Veena et al. (1998), Verwoerd et al. (1993), and references in Table 1. Also shown are galena data from Isua, Pilbara and Barberton komatiites (after Appel et al., 1978; Richards et al., 1981; Stacey and Kramers, 1975).

(1996) and Allègre and Lewin (1989) assuming closed-system evolution from 4.43 Ga to the present (Doe and Stacey, 1974; Wood et al., 2008). Although the interpretation of the Pb isotope data is model-dependent, several important features are apparent. First, because most carbonatite data plot above BSE estimates (Fig. 4), their Pb isotopic data record higher time-integrated U/Pb and Th/Pb ratios of their sources than those of BSE, consistent with the models of increasing μ for the residual in the mantle partial melting processes (Hofmann, 1988; Kwon et al., 1989; Meijer et al., 1990; Collerson et al., 2010). Second, both the findings from U-Pb and Th-Pb in carbonatites worldwide are similar to those from Sr, lacking evidence for depleted mantle before ~3 Ga. Isotopic data for galena from Isua, and Pilbara and Barberton komatiites (Appel et al., 1978; Richards et al., 1981; Stacey and Kramers, 1975), as well as data from 2.7 Ga carbonatites and syenites from the Canadian Shield, plot close to values proposed for BSE (e.g., Allègre and Lewin, 1989; Kwon et al., 1989; Galer and Goldstein, 1996). However, the new calcite data from the Siilinjarvi carbonatite (2.6 Ga) lie about BSE towards slightly more radiogenic $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios (Fig. 3), indicating higher U/Pb and Th/Pb ratios in their source than those in BSE, consistent with the findings of Tichomirowa et al. (2006). Third, the Pb isotope ratios from young (<0.2 Ga) carbonatites (Figs. 3 and 4) cover the range of data from OIBs involving HIMU, EM1, and FOZO (e.g., Simonetti et al., 1995, 1998; Bell and Simonetti, 1996; Bell and Tilton, 2001, 2002). Kwon et al. (1989) noted that the data from Canadian carbonatites younger than 2.7 Ga form negative slopes when plotted in $^{87}Sr/^{86}Sr$ vs. $^{206}Pb/^{204}Pb$ diagrams, similar to the mixing trends found between HIMU and EM1 end-members ('LoNd' array of Hart et al., 1986). Kwon et al. (1989) and Tilton and Kwon (1990) suggested that the carbonatite arrays resemble the slope of the OIB field, implying that both end members might have existed for at least 2.2 Ga. With the addition of new data from the Kola carbonatites (Fig. 3), it also appears that both FOZO and EM1 contributed to the 380 Ma plume.

Lead isotope data for galena from several of the Kola carbonatites show a considerable range of values that form a near-linear Pb-Pb isotope trend, a feature consistent with binary mixing (Bell et al., 2015). The more radiogenic end member, with values similar to FOZO and marked by a cluster of data from Kovdor, Sallanlatvi, Sokli and Vuoriyarvi, is interpreted to represent a closed-system, deep-seated, mantle reservoir (Fig. 4; Bell et al., 2015). The other, less radiogenic end member is represented by data from the Khibiny REE carbonatites and is similar to the very low $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios for calcite from a Sr-Ba carbonatite from the Murun potassic complex (134 Ma) from the Aldan Shield (Mitchell et al., 1994). Both seem to have tapped the same source.

4.3. Nd isotopic evolution

Bell and Blenkinsop (1987a), Kwon et al. (1989), and Tilton and Bell (1994) presented Nd isotopic data for the same Canadian Shield complexes analyzed for Sr and Pb. In contrast to the Sr and Pb isotopic data, most Archean carbonatites and syenitic rocks are characterized by positive ε_{Nd}(T) values relative to CHUR (after Jacobsen and Wasserburg, 1980; Hamilton et al., 1983) indicating that depleted mantle existed long before 3 Ga (Tilton and Kwon, 1990). The widespread nature of this depleted source is shown by the close agreement between Nd isotopic data for Archean carbonatites from the Baltic Shield, Greenland, and Canada (Karhu et al., 2001; Rukhlov et al., 2001; Bizzarro et al., 2002; Bell and Rukhlov, 2004; Tichomirowa et al., 2006, Zozulya et al., 2007).

The new Sm-Nd data shown in Figure 5a (n = 86) are similar to previously published values. Two important features are revealed in a compilation of ε_{Nd}(T) values from worldwide carbonatites and from the oldest basalts, komatiites, and crustal rocks from northwestern Canada, South Africa, and West Greenland (Fig. 5a; Bowring and Housh, 1995; Blichert-Toft and Arndt, 1999; Blichert-Toft et al., 1999; Vervoort and Blichert-Toft, 1999). About 70% of the data have positive ε_{Nd}(T) values marking the involvement of a depleted source. Furthermore, the scatter and range of ε_{Nd}(T) values from carbonatites probably reflects intermittent mixing between enriched and depleted mantle sources over at least 3.0 Ga. With decreasing age, the ε_{Nd}(T) values from carbonatites show much greater scatter and, although this might simply reflect the scarcity of Archean and Proterozoic carbonatites, we consider that it indicates a greater involvement of an enriched source, from either continental crust, or a deep-seated, primitive source.

Significantly, based on our new database, the present-day isotopic composition of the depleted mantle source is best represented by FOZO. The young carbonatites (<0.2 Ga) show a considerable range of ε_{Nd}(T) values (from +6.1 to –23.3), while the Archean carbonatites have much more restricted range of ε_{Nd}(T) values (from +4.4 to –2.6). We interpret these variations as indicating mixing between a depleted, FOZO-like source and a more enriched source such as the one reflected in the Phalaborwa (2.1 Ga), Spitskop (1.3 Ga), Tanil Nadu (0.8 Ga), and Murun and Laiwu-Zibo (0.1 Ga) complexes, and Cenozoic carbonatites in Italy (Eriksson, 1989; Mitchell et al., 1994; Kumar et al., 1998; Schleicher et al., 1998; Harmer, 1999; Pandit et al., 2002; Bizimis et al., 2003; Ying et al., 2004; Bell et al., 2013).

The main finding that emerges from the Nd data is that they point to a differentiation event much older than the 3 Ga depletion indicated by the Sr and Pb data (Tilton and Kwon, 1990; Rukhlov et al., 2001; Bell and Rukhlov, 2004). Either the Rb/Sr, U/Pb and Th/Pb ratios behaved differently than the Sm/Nd ratios during the Neoarchean event, or these apparent differences might be related to the choice of CHUR as the reference reservoir, especially if a non-chondritic model is assumed for BSE (e.g., Campbell and O'Neill, 2012). However, the convergence of terrestrial and extraterrestrial Nd-Hf isotope data (Martian, lunar, and eucrites) suggests that the CHUR estimate for BSE is probably correct (Bouvier et al., 2008). The Nd data from the Archean carbonatites show a spread similar to that from some of the Earth's oldest rocks (Fig. 5a; see Bowring

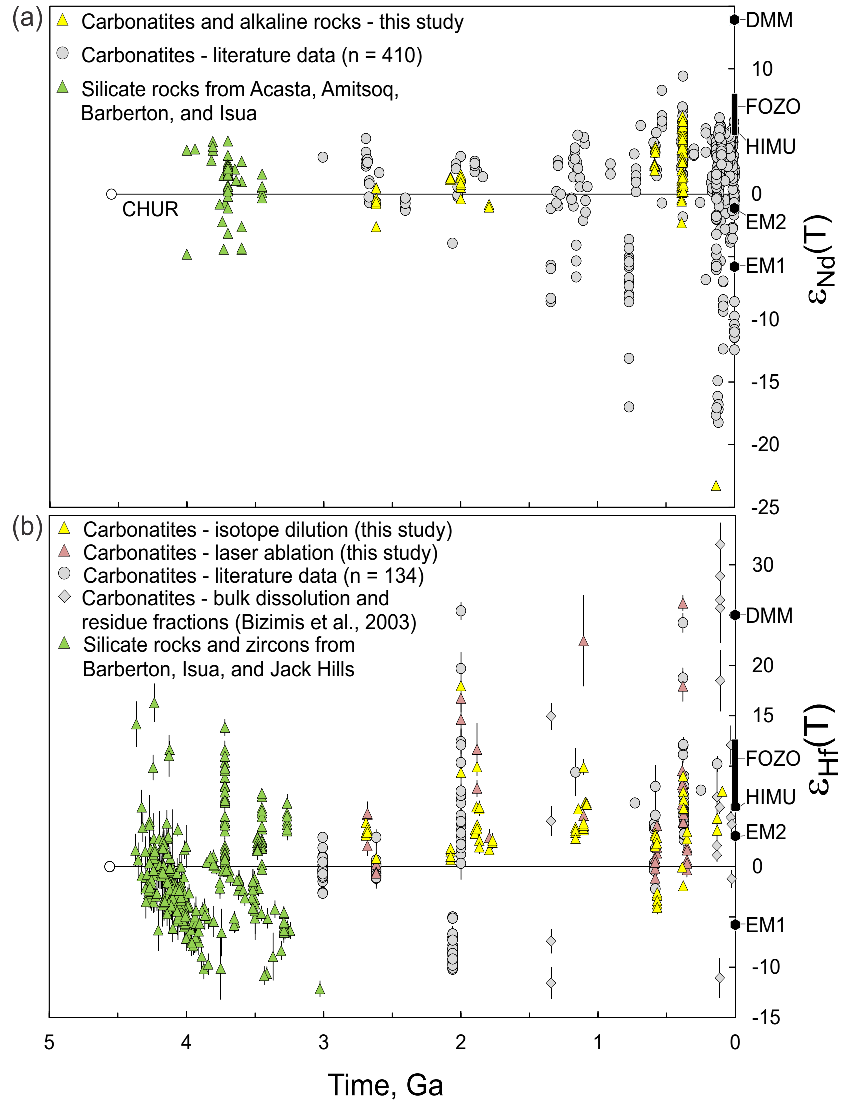

Fig. 5. Nd and Hf evolution diagrams for carbonatites worldwide and late Archean syenitic complexes from the Canadian Shield. Depleted MORB mantle (DMM), enriched mantle 1 and 2 (EM1 and EM2), 'FOcus ZOne' (FOZO), and high-$^{238}U/^{204}Pb$ or μ (HIMU) mantle components after Hart et al. (1992), Stracke et al. (2005), and Stracke (2012). **a)** $\varepsilon_{Nd}(T)$ versus time; $\varepsilon_{Nd}(T) = [(^{143}Nd/^{144}Nd_{sample}/^{143}Nd/^{144}Nd_{CHUR}) - 1]*10^4$, where $^{143}Nd/^{144}Nd_{sample}$ is the initial ratio in the sample and $^{143}Nd/^{144}Nd_{CHUR}$ is the ratio in the chondritic uniform reservoir (CHUR; after Jacobsen and Wasserburg, 1980; Hamilton et al., 1983) at that time. Data from Alberti et al. (1999), Andersen (1987, 1997), Baksi (1997), Balaganskaya et al. (2000), Barreiro and Cooper (1987), Beard et al. (1996), Bell and Blenkinsop (1987a, b), Bell and Peterson (1991), Bell and Tilton (2001), Bell et al. (1987, 2013), Bernard-Griffiths et al. (1991), Bizimis et al. (2003), Bizzarro et al. (2002), Castorina et al. (1996), Comin-Chiaramonti et al. (2002), Conticelli et al. (1995, 2002), Cooper and Mellish (2001), Cooper and Reid (1998), Corfu and Noble (1992), Coulson et al. (2003), Dunworth and Bell (2001), Eby et al. (1995), Eriksson (1989), Graham et al. (2004), Halama et al. (2005); Harmer (1999), Harmer et al. (1998), Hoernle and Tilton (1991), Hoernle et al. (2002), Huang et al. (1995), Kalt et al. (1997), Karhu et al. (2001), Keller and Krafft (1990), Kramm (1993), Kramm and Kogarko (1994), Kramm et al. (1997), Kumar et al. (1998), Kwon et al. (1989), le Roex and Lanyon (1998), Melluso et al. (2004), Middlemost (1990), Mitchell et al. (1994), Morisset (1992), Nelson et al. (1988), Pandit et al. (2002), Paslick et al. (1995), Pearce and Leng (1996), Pollock (1987), Ray et al. (2000), Reischmann (1995), Ruberti et al. (1997, 2002), Savatenkov et al. (1999), Schleicher et al. (1998), Schultz et al. (2004), Simonetti and Bell (1994a, b), Simonetti et al. (1995, 1998), Smithies and Marsh (1998), Srivastava et al. (2005), Sun et al. (1986), Thompson et al. (2002), Tilton and Bell (1994), Tilton et al. (1987, 1998), Toyoda et al. (1994), Veena et al. (1998), Verhulst et al. (2000), Verwoerd et al. (1993), Villeneuve and Relf (1998), Wagner et al. (2003), Walter et al. (1995), Wen et al. (1987), Yang et al. (2003), Ying et al. (2004), Zaitsev and Bell (1995), Zaitsev et al. (2002), Ziegler (1992), and references in Table 1. Also shown are data from the oldest silicate rocks from northwestern Canada, South Africa, and West Greenland (after Bowring and Housh, 1995; Blichert-Toft and Arndt, 1999; Blichert-Toft et al., 1999; Vervoort and Blichert-Toft, 1999). **b)** $\varepsilon_{Hf}(T)$ versus time; $\varepsilon_{Hf}(T) = [(^{176}Hf/^{177}Hf_{sample}/^{176}Hf/^{177}Hf_{CHUR}) - 1]*10^4$, where $^{176}Hf/^{177}Hf_{sample}$ is the initial ratio in the sample and $^{176}Hf/^{177}Hf_{CHUR}$ is the ratio in CHUR (after Iizuka et al., 2015) at that time. Data from Patchet et al. (1981), Bizzaro et al. (2002), Bizimis et al. (2003), Woodhead and Hergt (2005), Wu et al. (2006, 2010, 2011), Tappe et al. (2007, 2008), Kogarko et al. (2010), Ghobadi et al. (2012), Tichomoriwa et al. (2013), and references in Table 1. Also shown are data from the oldest silicate rocks and detrital zircons from South Africa, Western Australia, and West Greenland (Blichert-Toft and Arndt, 1999; Blichert-Toft et al., 2004; Harrison et al., 2005; Hoffmann et al., 2010; Kemp et al., 2010; Amelin et al., 2011; Puchtel et al., 2013). Error bars are 2σ uncertainties that include propagated errors associated with age, measured $^{167}Lu/^{177}Hf$ and $^{176}Hf/^{177}Hf$ ratios, ^{176}Lu decay constant, and CHUR parameters.

and Housh, 1995; Blichert-Toft and Arndt, 1999; Blichert-Toft et al., 1999; Vervoort and Blichert-Toft, 1999; Hoffmann et al., 2010; Caro, 2011), indicating a major differentiation event in the mantle before 3.0 Ga, perhaps within the first few hundred million years of Earth history.

4.4. Hf isotopic evolution

Although relatively scarce compared to Sr and Nd, the available Hf isotopic data from carbonatites worldwide show a huge range of initial $^{176}Hf/^{177}Hf$ ratios (Fig. 5b). Bizimis et al. (2003) reported Hf isotopic compositions for whole-rock, carbonate, and non-carbonate (leached residue) fractions from several carbonatites with different ages, which account for much of the scattering of ε_{Hf}(T) values in Figure 5b (from +32 to –12). Because Hf in carbonatites resides mainly in non-carbonate minerals (see Chakhmouradian, 2006), unlike Nd, other REEs, and Sr, it is difficult to obtain a complete suite of Sr-Nd-Pb-Hf isotopic data from the same mineral or whole-rock sample. Even though carbonatites typically contain <1 ppm Lu, they still have very high Lu/Hf ratios (averaging ~1.2; Woolley and Kempe, 1989; Bizimis et al., 2003) so that their initial $^{176}Hf/^{177}Hf$ ratios cannot be measured directly. Therefore, initial $^{176}Hf/^{177}Hf$ ratios are best measured using primary magmatic minerals with low Lu/Hf ratios such as zircon or baddeleyite (Patchett et al., 1981) both of which are common in many carbonatites. Both of these minerals have very low $^{176}Lu/^{177}Hf$ ratios (<0.0047; e.g., Tichomirowa et al., 2013; this study) and very high Hf content (~1 wt.%).

Lu-Hf data from zircons and baddeleyites from sixteen carbonatite complexes with ages ranging from 2.6 to 0.4 Ga from the Baltic and Canadian shields and the Canadian Cordillera have positive ε_{Hf}(T) values as high as +26, reflecting depleted mantle (Rukhlov and Bell, 2003). However, Bizzarro et al. (2002) did note enriched, negative ε_{Hf}(T) values from the 3.0 Ga Tupertalik carbonatite from Greenland, the oldest known carbonatite, and attributed them to an enriched, sub-chondritic source isolated in the deep mantle for at least 3 Ga. Similar values have also been reported from the Phalaborwa carbonatite (2.1 Ga) in South Africa (Scherer et al., 2001, Wu et al., 2006, 2010, 2011). Carbonatites and kimberlites of different age from Greenland show ε_{Hf}(T) values of between +7.9 to –5.1, indicating both depleted and enriched sources (Bizzarro et al., 2002).

Figure 5b shows ε_{Hf}(T) values for carbonatites worldwide, along with data from the oldest terrestrial rocks and detrital zircons from South Africa, western Australia, and West Greenland (Blichert-Toft and Arndt, 1999; Blichert-Toft et al., 2004; Harrison et al., 2005; Hoffmann et al., 2010; Kemp et al., 2010; Amelin et al., 2011; Puchtel et al., 2013), plotted as a function of time. Our new Lu-Hf data obtained by solution MC-ICPMS with isotope dilution are in close agreement with the data obtained by laser ablation (Rukhlov and Bell, 2003) for Zr-minerals (Table 1) and with our database for carbonatites worldwide (Patchet et al., 1981; Bizzaro et al., 2002; Bizimis et al., 2003; Woodhead and Hergt, 2005; Wu et al., 2006, 2010, 2011; Tappe et al., 2007, 2008; Kogarko et al., 2010; Ghobadi et al., 2012; Tichomirowa et al., 2013).

Most of the carbonatite data shown in Figure 5b reflect depleted mantle. The late Archean carbonatites have ε_{Hf}(T) values between +5.3 and –2.7, whereas the younger carbonatites have values between +26.2 and –10.2. The extremely radiogenic values for some of the young carbonatites are well beyond the range of the ε_{Hf}(T) values from modern oceanic basalts (FOZO, HIMU, EM1 and EM2 shown in Fig. 5b) and such anomalous values are thus unlikely to reflect the isotopic composition of their mantle sources. Insights into the Hf data come from analyses of individual mineral grains.

Large variations of ε_{Hf}(T) values (up to 20 units) within single zircon grains, documented by Tichomirowa et al. (2013) from the Tikseozero carbonatite complex (2.0 Ga) in the Baltic Shield, are accompanied by enrichment in Ca and other 'impurities', variable $\delta^{18}O$ values, and U/Pb disturbances. Tichomirowa et al. (2013) attributed these wide variations of Hf isotope compositions to a dissolution-reprecipitation of zircon and incorporation of radiogenic Hf from co-existing high-Lu/Hf phases such as carbonate and apatite during melt evolution. Because of their very high Lu/Hf ratios, these phases can rapidly develop very high $^{176}Hf/^{177}Hf$ ratios over a time interval of a few million years (Bizimis et al., 2003). Considering the protracted magmatic evolution proposed for the Oka carbonatite complex (0.1 Ga) in Canada of a few Ma (Chen and Simonetti, 2013), and the extremely radiogenic ε_{Hf}(T) values from Oka (Bizimis et al., 2003) and other carbonatite complexes (e.g., Wu et al., 2010; Tichomirowa et al., 2013), the data may thus reflect incorporation of radiogenic Hf during mineral growth. The wide range of ε_{Hf}(T) values for some of our data, especially from the Tiksheozero (2.0 Ga), Nemegosenda Lake (1.1 Ga), and Afrikanda (0.38 Ga) complexes could be consistent with these findings. If we consider some of these highly radiogenic values to be anomalous, then the present-day ε_{Hf}(T) value for one of the end members could still be considered similar to FOZO.

In spite of the wide variation in ε_{Hf}(T) values, the Hf isotopic data from carbonatites, along with those from the older silicate rocks and the Nd isotope data, suggest that the depleted mantle was formed by a major differentiation event before 3.0 Ga, perhaps during the Hadean (>4.0 Ga).

5. Conclusions

The Sr-Pb-Nd-Hf isotopic data from globally distributed carbonatites record the evolution of a primitive mantle source that has behaved as a relatively closed-system, at least during the last 3 Ga of Earth history, with the present-day, isotopic attributes similar to FOZO. Solar-like noble gas (He, Ne, Ar, Kr, and Xe) and N isotopic signatures in carbonatites further suggest that this widespread source may represent the deep, undegassed mantle. The Nd and Hf data from carbonatites and the oldest silicate rocks indicate a major depletion of chondritic Earth >3.0 Ga, perhaps during the Hadean (>4.0 Ga), and possibly a second, much later event at ~3 Ga, marked by a depleted mantle with low Rb/Sr and high U/Pb ratios. The ~3 Ga event could reflect a major change in the Earth's thermal regime marked by the onset of modern-style plate tectonics accompanied by the production of voluminous and more sialic, juvenile, continental crust (Tilton and Kwon, 1990; Dhuime et al., 2015). It appears that over the last 3 Ga, HIMU- and EM-like mantle end-members, and rarely, a more exotic, high-$^{87}Sr/^{86}Sr$ component (Bell et al., 2013) have been mixed in variable proportions with a depleted source (FOZO) accounting for the isotopic heterogeneity of the carbonatitic melt. The interpretation that we favour is one in which these enriched mantle end-members represent complementary partial melt (EM1) and residual (HIMU) of the FOZO protolith (Collerson et al., 2010) or, alternatively, primordial materials, perhaps including recycled Hadean crust, sampled by deep-mantle plumes originating from the D" layer (e.g., Tolstikhin and Hofmann, 2005; Campbell and O'Neill, 2012).

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