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Sr and Nd isotope data of apatite, calcite and dolomite as indicators of source, and the relationships of phoscorites and carbonatites from the Kovdor massif, Kola peninsula, Russia

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Abstract A detailed Sr-Nd isotopic study of primary apatite, calcite and dolomite from phoscorites and carbonatites of the Kovdor massif (380 Ma), Kola peninsula, Russia, reveals a complicated evolutionary history. At least six types of phoscorites and five types of carbonatite have been identified from Kovdor by previous investigators based on relative ages and their major and accessory minerals. Isotopic data from apatite define at least two distinct groups of phoscorite and carbonatite. Apatite from the earlier phoscorites and carbonatites (group 1) are characterized by relatively low $^{87}\text{Sr}/^{86}\text{Sr}$ (0.70330–0.70349) and $^{143}\text{Nd}/^{144}\text{Nd}$ initial ratios (0.51230–0.51240) with $F = 2.01\text{--}2.23$ wt%, Sr = 2185–2975 ppm, Nd = 275–660 ppm and Sm = 31.7–96.2 ppm. Apatite from the second group has higher $^{87}\text{Sr}/^{86}\text{Sr}$ (0.70350–0.70363) and $^{143}\text{Nd}/^{144}\text{Nd}$ initial ratios (0.51240–0.51247) and higher F (2.63–3.16 wt%), Sr (4790–7500 ppm), Nd (457–1074 ppm) and Sm (68.7–147.6 ppm) contents. This group corresponds to the later phoscorites and carbonatites. One apatite sample from a carbonatite from the earlier group fits into neither of the two groups and is characterized by the highest initial $^{87}\text{Sr}/^{86}\text{Sr}$ (0.70385) and lowest $^{143}\text{Nd}/^{144}\text{Nd}$ (0.51229) of any of the apatites. Within both groups initial $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios show negative correlations. Strontium isotope data from coexisting calcite and dolomite support the findings from the apatite study. The Sr and Nd isotopic similarities between carbonatites and phoscorites indicate a genetic relationship between the two rock types. Wide variations in Sr and Nd isotopic composition within some of the earlier carbonatites indicate several

distinct intrusive phases. Oxygen isotopic data from calcite and dolomite ($\delta^{18}\text{O} = +7.2$ to $+7.7\text{‰}$ SMOW) indicate the absence of any low-temperature secondary processes in phoscorites and carbonatites, and are consistent with a mantle origin for their parental melts. Apatite data from both groups of phoscorite plot in the depleted quadrant of an ϵ_{Nd} versus ϵ_{Sr} diagram. Data for the earlier group lie along the Kola Carbonatite Line (KCL) as defined by Kramm (1993) and data from the later group plot above the KCL. The evolution of the phoscorites and carbonatites cannot be explained by simple magmatic differentiation assuming closed system conditions. The Sr-Nd data can best be explained by the mixing of three components. Two of these are similar to the end-members that define the Kola Carbonatite Line and these were involved in the genesis of the early phoscorites and carbonatites. An additional component is needed to explain the isotopic characteristics of the later group. Our study shows that apatite from rocks of different mineralogy and age is ideal for placing constraints on mantle sources and for monitoring the Sr-Nd evolution of carbonatites.

Introduction

Carbonatites have been used effectively in monitoring the geochemical evolution of the upper mantle and placing constraints on the nature of mantle inhomogeneities (e.g. Bell et al. 1982; Nelson et al. 1988; Tilton and Kwon 1990). Because of their high Sr and Nd contents, carbonatites are ideal for such studies because the effects of any contamination by continental crust are reduced, and the isotopic composition of the mantle sources are still retained. Detailed studies of carbonatites from Canada, East Africa and the Kola peninsula of Russia have shown that (1) the sub-continental upper mantle is isotopically heterogeneous;

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(2) the Canadian sub-continental mantle has a higher time integrated Sm/Nd ratio than CHUR (chondrite uniform reservoir) and lower Rb/Sr ratio than bulk Earth; (3) the young East African carbonatites indicate the involvement of HIMU and EM I mantle components (Bell and Blenkinsop 1987) with data points falling along the LoNd array of Hart et al. (1986); (4) the carbonatites from Kola also involve two mantle components which are not the same as those associated with the African carbonatites (Kramm 1993). Whether carbonatites are derived from melts generated from the asthenosphere or lithosphere is still the source of debate, although the isotopic similarities between some oceanic island basalts and carbonatites suggest an asthenospheric source.

The Palaeozoic Kola alkaline province is situated between north-eastern Finland and the eastern part of the Kola peninsula, Russia, and constitutes one of the largest alkaline-carbonatite provinces yet known. It covers an area of about 100,000 km². Twenty-four complexes were intruded into the Baltic Shield 380 to 360 Ma ago (Kramm et al. 1993). Their spatial distribution is controlled by rifting, particularly the north-east trending Kontozero graben and north-west trending Yuov-Kandalaksha deep fracture zone. Although there have been some recent regional isotopic studies of Palaeozoic ultrabasic-alkaline carbonatite and nepheline syenite complexes from the Kola peninsula (Kramm 1993; Kramm et al. 1993), there are no detailed isotopic studies of individual massifs that attempt to evaluate relationships among the various rock types.

The Kovdor alkaline massif was chosen for study for several different reasons. Kovdor covers an area of 40.5 km² and contains a wide variety of magmatic and metasomatic rocks that range from ultrabasites to car-

bonatites. Kovdor is extremely important economically, and because of this it has been the subject of intense study by Russian researchers during the last fifty years. Relative age relationships of the different rocks are now known, and their petrology, geochemistry and mineralogy have been studied in detail.

The aims of our study are three-fold: (1) to place constraints on the relationships between the phoscorites and carbonatites from Kovdor; (2) to evaluate whether the phoscorites are magmatic products or whether they have been produced by metasomatic activity associated with continental crust; (3) to assess the nature of the mantle source region that generated the Kovdor complex.

Historical review of phoscorites

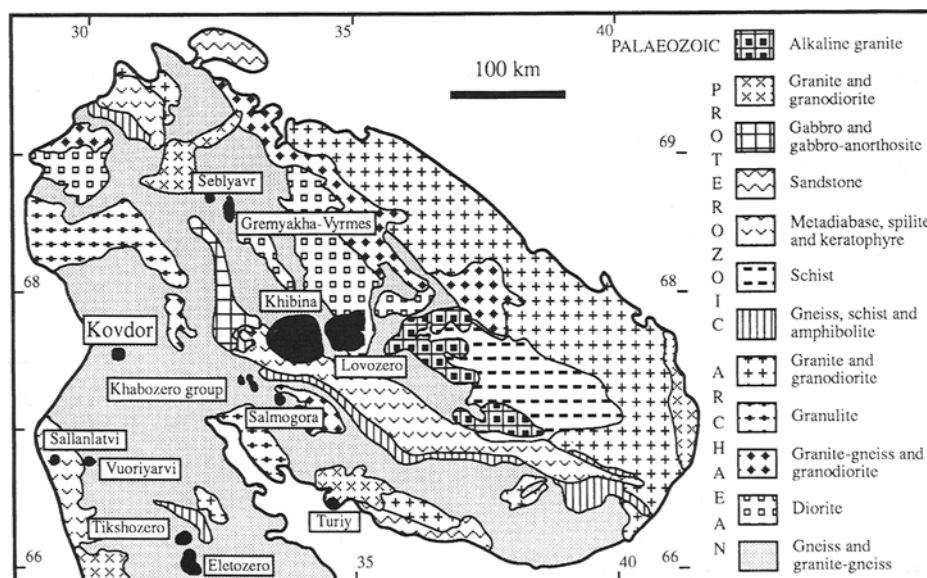
Definition

The term *phoscorite* was first used by Russell et al. (1954) to describe a magnetite-olivine-apatite rock from Phalaborwa, South Africa. In the latest IUGS monograph on igneous rock classification, Le Maitre et al. (1989) define a phoscorite as an igneous "magnetite, olivine, apatite rock usually associated with carbonatites". Similar rock types in the Russian literature have been described as magnetite-forsterite rocks (Kukharev et al. 1965; Kapustin 1980), camaforites (Borodin et al. 1973), the latter a mnemonic device based on calcite, apatite, magnetite and forsterite, and carbonatitoids (Epshteyn and Danilchenko 1988). Egorov's (1993) classification scheme for phoscorites involves nine mineralogical types based on the relative abundances of the three major minerals: olivine, magnetite and apatite.

Occurrences

Phoscorites, although comparatively rare, are relatively abundant in the Kola, Maymecha-Kotuy and Aldan provinces in Russia.

Fig. 1 Simplified geological map of the Kola peninsula, Russia, showing location of main alkaline massifs (black) (after Kukharev et al. 1965). Greymakha-Vyrmes and Eletozero massifs are Proterozoic, all other massifs are Palaeozoic



Detailed descriptions of these phoscorites are given in papers by Kukharenko et al. (1965), Borodin et al. (1973), Kapustin (1980), Kharlamov et al. (1981), Lapin (1979, 1982), Lapin and Vartiainen (1983), Bulakh and Ivanikov (1984), Epshteyn and Danilchenko (1988), Krasnova and Kopylova (1988) and Egorov (1993). Phoscorites and rocks similar to phoscorites have been described from Africa, including Phalaborwa (Russell et al. 1954; Eriksson 1989) and Schiel, South Africa (Verwoerd 1993), Bukusu and Sukulu, Uganda (Davies 1947), Dorowa and Shawa, Zimbabwe (Johnson 1966), Panda Hill (Basu and Mayila 1986) and Ngualla, Tanzania (van Straaten 1989). Apatite-rich rocks have also been described from Oka and Nemeegos, Canada (Philpotts 1967).

General geology and mineralogy

Phoscorites in alkaline massifs normally take the form of stocks and stockworks, as well as ring and linear dykes with sizes from a few centimetres up to several hundred metres. Phoscorites are medium- to coarse-grained, heterogeneous in texture and can have either sharp or gradational contacts with the surrounding country rocks. They can also contain angular or rounded xenoliths of olivinite, pyroxenite or ijolite, and they also occur as xenoliths in carbonatites.

The main minerals in phoscorites are forsterite, magnetite, apatite, calcite and/or dolomite. Other minerals include mica (phlogopite, biotite, tetraferriphlogopite), pyroxene (diopside, aegirine), amphibole (hastingsite, tremolite, richterite) and ilmenite. Common accessory minerals include baddeleyite, calzirtite, pyrochlore, zircon and zirkelite. Serpentine, chlorite, clinohumite occur as secondary minerals after forsterite.

There is considerable debate about which minerals are primary in phoscorites. According to Kapustin (1980), apatite and calcite are formed by metasomatism, while Bulakh and Ivanikov (1984) and Egorov (1993) consider that carbonates in phoscorites are secondary minerals produced by reaction between the phoscorite and carbonatites. Sokolov (1983), however, claims that the carbonates in phoscorites are primary, although secondary carbonates can be present too. The evidence for the primary origin of carbonates includes geological setting of carbonate-bearing phoscorites within alkaline complexes, petrographic observations (textural-structural relationships among carbonates and other minerals) and mineralogical data (chemical composition and physical properties).

In this paper we use the name *phoscorite* as a general term to describe carbonate-forsterite-magnetite-apatite rocks associated with the Kovdor carbonatites. The major minerals making up the phoscorites at Kovdor are indicated by a mineral prefix, e.g. apatite-forsterite phoscorite.

Geological setting of the Kovdor massif

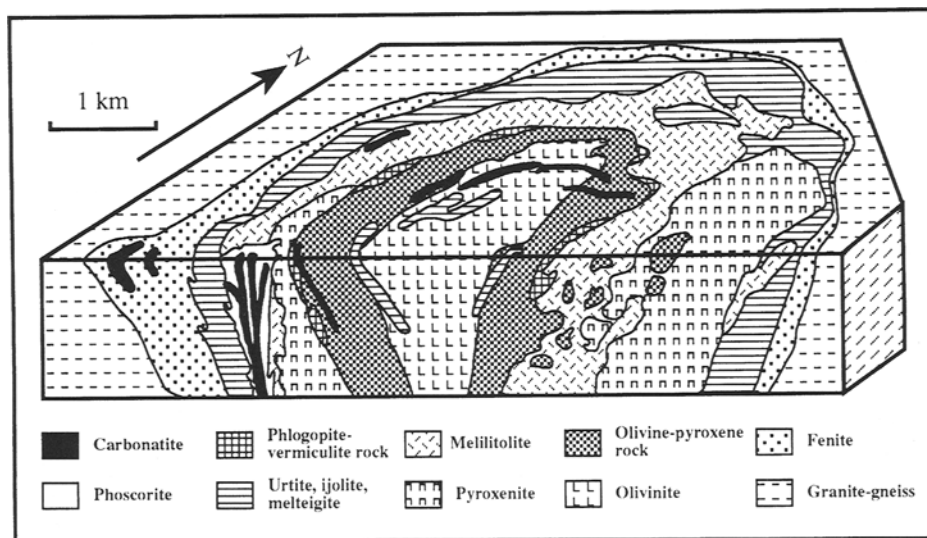
General geology

The Kovdor ultrabasic-alkaline-carbonatite complex is situated in the north-eastern part of the Baltic Shield, Kola peninsula, Russia (67° 35' N and 30° 20' E) (Fig. 1) and is emplaced in granitic gneisses of the Belomorian Group of Archean age. The Kovdor massif, along with the Sokli and Turiy Mys massifs follows the Yuov-Kandalaksha deep fracture zone, which during Proterozoic and Phanerozoic time was repeatedly active (Bulakh and Ivanikov 1984). Three Rb/Sr mineral ages of Kovdor olivinite and pyroxenite indicate intrusion ages of 376.1 ± 0.6 Ma, 374.3 ± 0.3 Ma and 373.1 ± 4.6 Ma (U. Kramm, personal communication 1994). A Rb/Sr age for the carbonatites of 370 Ma was assumed by Kramm (1993) in his study of alkaline massifs of the Kola province. The U-Pb data from baddeleyite from tetraferriphlogopite-bearing phoscorite and carbonatite suggest an age of 380 ± 4 Ma (Bayanova et al. 1991). This age is very close to the Rb/Sr ages of Kramm, and we have used a figure of 380 Ma as the best approximation to the emplacement of the Kovdor phoscorites and carbonatites.

The general geology of the Kovdor massif has been described by Kukharenko et al. (1965) and Kapustin (1980). The concentrically zoned composite plutonic body (Fig. 2) has a tear-drop shape and contains ultrabasic rocks (olivinite, pyroxenite and olivine-pyroxene rocks) in the core. Alkaline rocks (ijolite, melteigite and jacupirangite) form marginal ring dykes. Turjaite, melilitolite, phlogopite-diopside-olivine and garnet-amphibole-monticellite rocks lie between the ultrabasic and the alkaline rocks. Various phoscorites and carbonatites occur at the contact of the pyroxenite and the ijolite in the south-western part of the massif (Fig. 2). The massif is surrounded by a well-defined zone of fenites.

There are three types of economic deposits at Kovdor: (1) magnetite-apatite-baddeleyite in the phoscorites and carbonatites;

Fig. 2 Block diagram of the Kovdor massif, showing position of the phoscorites ("Main ore body") and carbonatites (after Kukharenko et al. 1965)



(2) phlogopite in the phlogopite-olivine-diopside rocks; (3) vermiculite in the surface-weathered phlogopite-bearing rocks.

Phoscorites and carbonatites

The various aspects of the geology, petrography and mineralogy of the phoscorites and carbonatites from the Kovdor have been described in numerous publications, e.g. Kukharensko et al. (1965), Kapustin (1980), Kharlamov et al. (1981), Epshteyn and Danilchenko (1988) and Krasnova and Kopylova (1988).

The phoscorite rocks occur as steeply dipping veins, and pipe-like, almost vertical bodies, which have been traced in drill holes to a depth of almost 1.5 km. Petrographically the bodies are concentrically zoned. Their assemblages gradually change from forsterite and apatite-forsterite phoscorites at the rim, to apatite-forsterite-magnetite and phlogopite-calcite-forsterite-magnetite in the middle part and finally to tetraferriphlogopite-calcite-forsterite-magnetite and dolomite-magnetite phoscorites in the core. All of these varieties form the so-called "Main ore body" (Fig. 2). Ilmenite-dolomite-tremolite-magnetite and ilmenite-tremolite-calcite-magnetite phoscorites, are found in the eastern part of the deposit and form a separate "Satellite" ore body.

Both dolomite and calcite carbonatites are found at Kovdor, and two major types have been recognised. The early type includes calcite carbonatite with forsterite, phlogopite and calcite carbonatite with tetraferriphlogopite; the late type contains dolomite carbonatite, calcite-dolomite carbonatite with ilmenite and dolomite-calcite carbonatite with labuntsovite. Carbonatites as well as phoscorites form steeply-dipping stockwork veins.

Table 1 shows the various stages of phoscorite and carbonatite formation at Kovdor. Krasnova and Kopylova (1988) considered the ilmenite-dolomite-tremolite-magnetite phoscorite and the calcite-dolomite carbonatite with ilmenite as the latest of these rocks, and this has been confirmed (by A.N.Z.) by geological mapping and drill core investigation in the eastern open pit. The genetic relationships, however, between the phoscorites and the carbonatites from the "Main ore body" and the "Satellite" body are still poorly understood.

The samples used in this study are representative of the different types of phoscorites (Ph 1-5) and carbonatites (C 1-3) from the "Main ore body". They include: apatite-forsterite phoscorite (Ph 1)—sample KV 2; apatite-forsterite-magnetite phoscorite (Ph 2)—KV 4; calcite-forsterite-magnetite phoscorite with phlogopite (Ph 3)—KV 5, KV 8, KV 9; calcite-forsterite-magnetite phoscorite with tetraferriphlogopite (Ph 4)—KV 10; dolomite-magnetite phos-

corite (Ph 5)—KV 15; calcite carbonatite with forsterite and phlogopite (C 1)—KV 18, KV 23, KV 24, KV 25; calcite carbonatite with tetraferriphlogopite (C 2)—KV 21; dolomite carbonatite (C 3)—KV 22. These samples were collected from drill core and from the open pit and are virtually free of alteration.

Analytical methods

Microprobe analyses

Wavelength-dispersion analyses of apatite, calcite and dolomite were made on polished thin sections using a Cambridge Microscan (MK) 5 electron microprobe (Carleton University). An acceleration voltage of 15 kV and a beam current of 20 nA were used for analyses. The following standards were used: natural dolomite (Ca, Mg), siderite (Fe) and rhodochrosite (Mn) for calcite and dolomite, and Durango apatite (Ca, P, F), celestite (Sr) and synthetic phases for La and Ce for apatite. The CO₂ was calculated by stoichiometry based on 1 C for calcite and 2 C for dolomite per formula unit.

Isotope analyses

One hundred milligrams of apatite and dolomite samples were hand-picked and then dissolved in HCl-HNO₃ mixture; calcite samples were dissolved in HCl solution. Separation of the Rb and Sr was carried out by conventional ion-exchange methods and separation of the Nd and Sm, by reverse-phase chromatography (Richard et al. 1976). Concentrations of Rb, Sr, Nd and Sm were determined by isotope dilution using single ⁸⁷Rb, ⁸⁴Sr, ¹⁴⁹Sm and ¹⁴⁵Nd spikes. Neodymium and Sr were analysed using a double Re-filament technique. All isotope ratios were measured at Carleton University on a Finnigan-MAT 261 multicollector solid-source mass spectrometer, operated in the static mode. Reproducibility of the ¹⁴³Nd/¹⁴⁴Nd and ⁸⁷Sr/⁸⁶Sr quoted is about 0.004% (2 sigma level) and reproducibility of concentrations is about 0.5% of the quoted values. Isotope results are normalized to ¹⁴⁶Nd/¹⁴⁴Nd = 0.7219 and ⁸⁶Sr/⁸⁸Sr = 0.1194. The ⁸⁷Sr/⁸⁶Sr for the NBS 987 Sr standard measured at Carleton is 0.71025 ± 0.00003 and the ¹⁴³Nd/¹⁴⁴Nd for the La Jolla Nd standard is 0.51186 ± 0.00002. The initial Sr and Nd isotope ratios were calculated using the decay constants for ⁸⁷Rb and ¹⁴⁷Sm of 1.42 × 10⁻¹¹ a⁻¹ and 6.54 × 10⁻¹² a⁻¹, respectively, and an age of 380 Ma.

Table 1 Successive stages of formation of phoscorites and carbonatites of the Kovdor ore deposit, based on the data of Krasnova and Kopylova (1988) and the authors

Stage	Phoscorite	Carbonatites
Ia	Forsterite, apatite-forsterite (Ph 1)	
Ib	Apatite-forsterite-magnetite (Ph 2)	
IIa	Calcite-forsterite-magnetite with phlogopite (Ph 3)	Calcite with forsterite and phlogopite (C 1)
IIb	Calcite-forsterite-magnetite with tetraferriphlogopite (Ph 4)	Calcite with tetraferriphlogopite (C 2)
IIIa	Dolomite-forsterite-magnetite, dolomite-magnetite (Ph 5)	Dolomite (C 3)
IIIb	Dolomite-tremolite-magnetite, tremolite-calcite-magnetite with ilmenite (Ph 6)	Calcite-dolomite with ilmenite (C 4) Dolomite-calcite with labuntsovite (C 5)

Carbon dioxide was analysed on a VG-ISOGAS SIRA-12 fully automated, fixed triple-collector mass spectrometer at University of Ottawa. Precision of the $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ is $\pm 0.1\text{‰}$. The $\delta^{18}\text{O}$ values were calculated using α values of O between CO_2 -carbonate: 1.01025 for calcite (25°C) and 1.01065 for dolomite (50°C).

Results

Mineral chemistry of apatite, calcite and dolomite.

Apatite occurs as fine (0.1–0.5 mm) rounded, elliptical grains in the apatite-forsterite, apatite-forsterite-magnetite and tetraferriphlogopite-calcite-forsterite-magnetite phoscorites, and as short (1–5 mm) and long (up to 15 mm) prismatic grains in the phlogopite-calcite-forsterite-magnetite and dolomite-magnetite phoscorites. In the calcite carbonatites, apatite commonly occurs as elliptical grains (2–5 mm in size) or less commonly as prismatic (up to 20 mm) grains. In the dolomite carbonatite, apatite occurs as fine (0.1–0.5 mm) "sugar-like" grains. Many apatite grains contain primary fluid inclusions, which are elongated parallel to the c-axis of the apatite.

The apatite of the Kovdor phoscorites and carbonatites is fluorapatite (Table 2). The fluorine content increases progressively from early apatite-forsterite-

magnetite phoscorites to late dolomite carbonatites. Although this was originally documented by Rimskaya-Korsakova et al. (1979), our data show much higher F concentrations of between 2.01–3.16 wt% unlike the values of 0.87–2.08 wt% given by Rimskaya-Korsakova et al. (1979). The Sr abundances in apatite determined by both microprobe and isotope analyses are in good agreement, and there is a progressive increase from the older to the younger rock types (Tables 2 and 5), a feature that appears to hold for most carbonatites (Hogarth 1989). Fluorine shows a positive correlation with Sr, but less so with Nd (Fig. 3). The F-Sr-Nd data from the apatites can be divided into at least two groups. The first is characterized by relatively low F-Sr-Nd apatite from the earlier rock types (Ph 1, 2 and 3 phoscorites and C 1 carbonatite) and the second is marked by relatively high F-Sr-Nd apatites from the later rock types (Ph 4 and 5 phoscorites and C 2 and 3 carbonatites). The La and Ce abundances in these apatites also indicate two groups of apatite, one a relatively low La-Ce apatite (Ph 2 and 3 phoscorites and C 1 carbonatite) and the second a relatively high La-Ce apatite (Ph 4 and 5 phoscorites and C 2 and 3 carbonatites).

A progressive increase in F and REE contents in apatite from the early to late stage carbonatites is found

Table 2 Chemical composition of apatite from the Kovdor phoscorites and carbonatites. Uncertainties: major oxides, 2% of the quoted value; minor oxides < 5 wt% (5%) < 1 wt% (10%).

Rock index Sample no.	Ph 2 KV-4	Ph 3 KV-9	C 1 KV-25	Ph 4 KV-10	C 2 KV-21	Ph 5 KV-15	C 3 KV-22
Wt%							
CaO	54.58	54.90	54.87	53.06	54.52	54.30	51.30
SrO	0.29	0.27	0.31	0.71	0.84	0.88	0.87
La ₂ O ₃	0.02	0.05	0.02	0.13	0.15	0.16	0.11
Ce ₂ O ₃	0.09	0.05	0.12	0.22	0.27	0.25	0.25
P ₂ O ₅	41.98	41.32	40.91	41.79	42.12	41.03	41.81
F	2.01	2.23	2.18	2.75	2.63	2.81	3.16
·O=F	0.84	0.94	0.92	1.16	1.10	1.18	1.33
Total	98.13	97.88	97.49	97.50	99.43	98.25	96.17

Fig. 3 Plot of F versus Sr and F versus Nd for apatite from phoscorites and carbonatites. (Filled symbols, phoscorites; open symbols carbonatites)

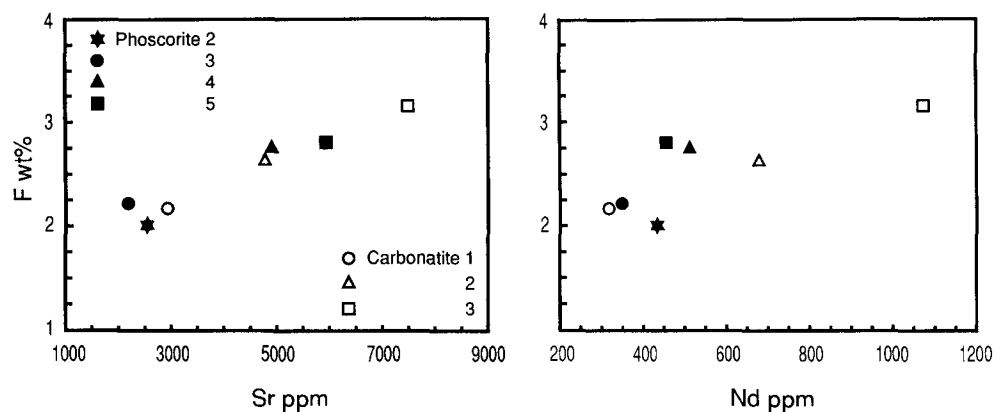


Table 3 Chemical composition of calcite and dolomite from the Kovdor carbonatites

Rock index Sample no.	C 1 KV-25	C 2 KV-21	C 3 KV-22
Wt%			
CaO	54.04	54.74	29.68
MgO	1.24	0.65	20.98
FeO* ^a	0.14	0.14	0.94
MnO	0.06	0.00	0.24
CO ₂	43.89	43.76	46.88
Total	99.37	99.29	98.72

^aFeO* total Fe as FeO

in several complexes, and is attributed by Egorov (1984) to magma differentiation during the formation of the phoscorites and carbonatites.

Calcite in the phoscorites and carbonatites at Kovdor massif occurs as subhedral or anhedral grains 1–5 mm in size, but can reach up to 15 mm in size. All of the calcites we have studied have nearly straight crystal boundaries and almost 120° triple junctions, and there is no evidence of any replacement textures. We assume, therefore, that the mineral is a primary phase. The calcite from sample KV 24 (C 1 carbonatite) forms tabular crystals, up to 5 cm length, flatted perpendicular to the c-axis, a morphology that is considered to be a primary igneous texture by Zhabin (1971). Calcite in the carbonatites with forsterite and phlogopite (C 1 carbonatite) contains dolomite microcrystals, which were interpreted as the products of exsolution from magnesian calcite solid-solution and the temperature of exsolution was estimated as 665–700°C (Zaitsev and Polezhaeva 1994).

The chemical composition of calcite from the Kovdor carbonatites is given in Table 3 and the Sr content of the mineral, determined by isotope dilution, is given in Table 5. These data show that calcite of carbonatite phases 1 and 2 is a low-magnesian variety with a small amount of Fe and Mn, which is fairly common in minerals from many carbonatite complexes (Sokolov 1985). Relatively high Sr content in calcite (average 6035 ppm) characterizes many early stage carbonatites (Pouliot 1970; Sokolov 1985; Platt and Woolley 1990; Clarke et al. 1994).

Dolomite in the late C 3 carbonatite occurs as white subhedral grains (1–10 mm) and its chemical composition (Table 3) is similar to that found in most carbonatites.

Stable isotope data

Carbon and oxygen isotope data from six calcite and one dolomite samples are shown in Table 4. The data are plotted on Fig. 4 together with fields for primary igneous carbonatites and mantle derived oceanic basalts (Keller and Hoefs 1995). The data of two calcite samples from C 1 carbonatites plot in the field of primary igneous carbonatites, while five samples have much higher $\delta^{13}\text{C}$ values. In contrast to the $\delta^{13}\text{C}$ the $\delta^{18}\text{O}$ values are extremely uniform and fall between +7.2 and +7.7‰ SMOW. This is strong evidence for the lack of any interaction between the carbonatites and meteoric water.

The variations in $\delta^{13}\text{C}$ at Kovdor cannot be attributed to fractionation processes that occurred during the emplacement and crystallization of the carbonatite magma, because removal of calcite from the

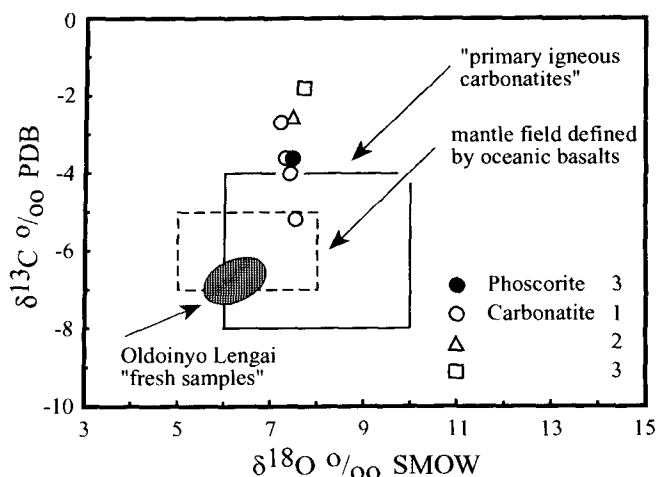


Fig. 4 Plot of $\delta^{13}\text{C}$ versus $\delta^{18}\text{O}$ for calcite and dolomite from phoscorite and carbonatites. Diagram after Keller and Hoefs (1995). Primary igneous carbonatite field from Taylor et al. (1967) and Hoefs (1987). Basalt field from Kyser (1986) and Deines (1989). (*Open symbols carbonatites*)

Table 4 C–O isotope analytical data. Errors are given as 2σ values

Sample	Rock	Mineral	$\delta^{13}\text{C}_{\text{‰/PDB}}$	$\delta^{18}\text{O}_{\text{‰/SMOW}}$
KV-8	Phoscorite 3	Calcite	– 3.6	+ 7.4
KV-18	Carbonatite 1	Calcite	– 3.6	+ 7.3
KV-23	Carbonatite 1	Calcite	– 5.2	+ 7.5
KV-24	Carbonatite 1	Calcite	– 2.7	+ 7.2
KV-25	Carbonatite 1	Calcite	– 4.0	+ 7.4
KV-21	Carbonatite 2	Calcite	– 2.6	+ 7.4
KV-22	Carbonatite 3	Dolomite	– 1.8	+ 7.7

magma would result in $\delta^{13}\text{C}$ enrichments, and this would be accompanied by changes in the $\delta^{18}\text{O}$ values, which do not occur here. We feel that the simplest explanation would be to attribute the variations in the carbon isotopic composition to isotopic heterogeneities in the upper mantle.

Isotopic variations from East African intrusive carbonatites which form a north-south trending zone are $\delta^{13}\text{C} = -2.8$ to -3.9‰ PDB (Peedee belemnite), while carbonate lavas and tuffs as well as intrusive carbonatites from the eastern and western rift areas have values that are significantly lower $\delta^{13}\text{C} = -6$ to

-7‰ PDB. These have been attributed by Deines and Gold (1973) to mantle heterogeneities, a feature that also been deduced from the study of diamonds (Deines et al. 1987).

Radiogenic isotopes

The Rb-Sr and Sm-Nd isotopic data are given in Tables 5 and 6.

The measured $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in the apatite and carbonate minerals can be considered initial because of

Table 5 Rb-Sr isotope analytical data

Sample	Rock	Mineral	Rb ppm	Sr ppm	$^{87}\text{Rb}/^{86}\text{Sr}$	$^{87}\text{Sr}/^{86}\text{Sr}$ measured = initial	$\epsilon_{\text{Sr}}(\text{T})$
KV-2	Phoscorite 1	Apatite	0.06	2560	0.00007	0.70344	- 8.7
KV-4	Phoscorite 2	Apatite		2550		0.70343	- 8.8
KV-5	Phoscorite 3	Apatite		2755		0.70349	- 8.0
KV-8	Phoscorite 3	Apatite	0.04	2975	0.00004	0.70330	- 10.7
KV-8	Phoscorite 3	Calcite		4850		0.70323	- 11.7
KV-9	Phoscorite 3	Apatite		2185		0.70338	- 9.6
KV-18	Carbonatite 1	Apatite		2715		0.70341	- 9.1
KV-18	Carbonatite 1	Calcite		5435		0.70335	- 10.0
KV-23	Carbonatite 1	Calcite		7995		0.70354	- 7.3
KV-24	Carbonatite 1	Calcite		2835		0.70379	- 3.7
KV-24 ^a	Carbonatite 1	Calcite				0.70378	
KV-25	Carbonatite 1	Apatite		2930		0.70385	- 2.9
KV-25	Carbonatite 1	Calcite	0.05	5200	0.00003	0.70391	- 2.0
KV-25 ^a	Carbonatite 1	Calcite				0.70395	
KV-10	Phoscorite 4	Apatite	0.03	4915	0.00002	0.70359	- 6.6
KV-21	Carbonatite 2	Apatite		4790		0.70350	- 7.8
KV-21	Carbonatite 2	Calcite		9895		0.70352	- 7.6
KV-15	Phoscorite 5	Apatite		5940		0.70357	- 6.8
KV-22	Carbonatite 3	Apatite		7500		0.70363	- 6.0
KV-22	Carbonatite 3	Dolomite	0.46	3270	0.00041	0.70357	- 6.8

^a Replicate analysis, errors are given as 2σ values

Table 6 Sm-Nd isotope analytical data. Errors are given as 2σ values

Sample	Rock	Mineral	Sm ppm	Nd ppm	$^{147}\text{Sm}/^{144}\text{Nd}$	$^{143}\text{Nd}/^{144}\text{Nd}$ measured	$^{143}\text{Nd}/^{144}\text{Nd}$ initial	$\epsilon_{\text{Nd}}(\text{T})$
KV-2	Phoscorite 1	Apatite	63.2	410	0.0931	0.51255	0.51232	+ 3.4
KV-4	Phoscorite 2	Apatite	67.1	434	0.0935	0.51259	0.51236	+ 4.1
KV-5	Phoscorite 3	Apatite	62.5	396	0.0954	0.51254	0.51230	+ 3.0
KV-8	Phoscorite 3	Apatite	31.7	275	0.0698	0.51257	0.51240	+ 5.0
KV-9	Phoscorite 3	Apatite	67.7	349	0.1174	0.51265	0.51236	+ 4.1
KV-18	Carbonatite 1	Apatite	96.2	660	0.0881	0.51262	0.51240	+ 5.0
KV-25	Carbonatite 1	Apatite	43.4	316	0.0965	0.51253	0.51229	+ 2.8
KV-10	Phoscorite 4	Apatite	71.0	513	0.0837	0.51263	0.51242	+ 5.4
KV-21	Carbonatite 2	Apatite	93.0	679	0.0828	0.51268	0.51247	+ 6.4
KV-15	Phoscorite 5	Apatite	68.7	457	0.0908	0.51264	0.51241	+ 5.2
KV-22	Carbonatite 3	Apatite	147.6	1074	0.0831	0.51261	0.51240	+ 5.0

the low Rb/Sr ratios. The Rb contents determined for selected samples of apatite, calcite and dolomite range from 0.03 to 0.46 ppm and Sr values were all greater than 2000 ppm.

The variation in Sr isotopic composition for all of the mineral phases is quite pronounced and initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios for all samples range from 0.70323 to 0.70393. All initial ratios were calculated assuming an age of 380 Ma. The apatite from the phoscorites has $^{87}\text{Sr}/^{86}\text{Sr}$ initial ratios between 0.70330 to 0.70359, and those from the carbonatites range from 0.70341 to 0.70385. Calcite from carbonatites has values between 0.70335 and 0.70393, a dolomite sample has an initial Sr ratio of 0.70357, and a calcite sample from a phoscorite yielded a value of 0.70323, the lowest obtained from any of our samples. In Fig. 5a we plot $^{87}\text{Sr}/^{86}\text{Sr}$ versus Sr content for the apatite from both phoscorites and carbonatites. The data define a positive trend and can be divided into two groups, one characterized by relatively low $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.70330–0.70349) and low Sr contents (2185–2975 ppm) corresponding to the earlier phoscorites (Ph 1, 2 and 3) and carbonatites (C 1), and a second group with higher $^{87}\text{Sr}/^{86}\text{Sr}$ (0.70350–0.70363) and Sr abundances (4790–7500 ppm), which corresponds to the later group of phoscorites (Ph 4 and 5) and carbonatites (C 2 and 3). Apatite sample KV 25, from a C 1 carbonatite, fits into neither of these two groupings and is characterized by the highest initial $^{87}\text{Sr}/^{86}\text{Sr}$ found in any of the apatites (0.70385).

Of special significance is the large variation in $^{87}\text{Sr}/^{86}\text{Sr}$ ratios (0.70335–0.70391) shown by the calcite and apatite from C 1 carbonatite (Fig. 5b). The variation in isotope composition was verified by running two duplicate analyses on calcite samples with the highest values. The reproducibility of about 3 parts in 70000 shows that the samples have different Sr isotope composition. This suggests that the early carbonatites either involve different sub-phases of carbonatite activity that have not been recognised in the field, or less likely that the carbonatite was intruded in a plastic state such that any isotopic heterogeneities were preserved.

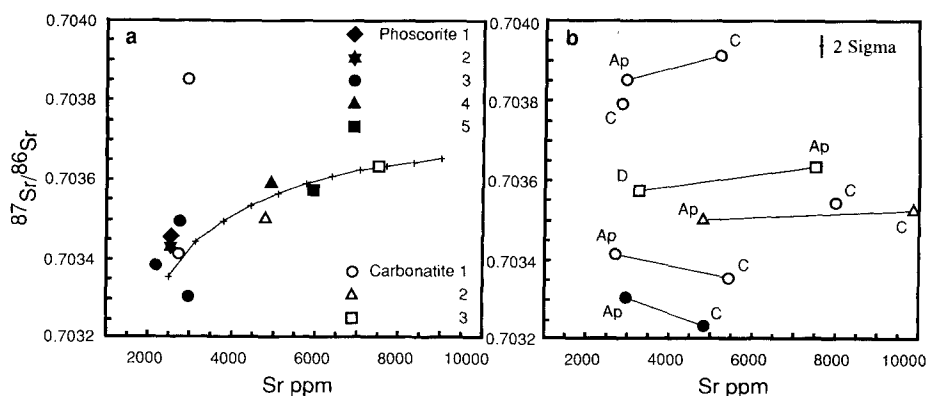
Tie lines have been drawn in Fig. 5b between calcite and apatite from the same sample, and from this it can be seen that although the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios are very similar for all pairs (differences in the isotopic composition between coexisting apatite and calcite or dolomite range from 0.00002 to 0.00007), the Sr content is always much higher in the calcite. The K_D of $\text{Sr}_{\text{calcite}}/\text{Sr}_{\text{apatite}}$ varies from 1.6 to 2.1, whereas the dolomite-apatite pair yields a value of 0.4.

The lowest $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70323 is found in calcite from Ph 3 phoscorite, and this value is substantially lower than the values found in the calcites from C 1 and 2 carbonatites. Although there is ongoing debate about whether the calcite in the phoscorites is primary or secondary, the relatively low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio for sample KV 8 suggests a primary rather than a secondary source. This is also supported by the low $^{87}\text{Sr}/^{86}\text{Sr}$ ratio from the measurements made of apatite from the same sample. In general, the overall grouping of the data with those from the other samples suggests that calcite is part of the same magmatic process.

The Nd initial ratios from the apatites are also variable, but both apatite from phoscorites (0.51230–0.51242) and carbonatites (0.51229–0.51247) show similar ranges. The same is true for most of the Nd abundances (phoscorites: 275–513 ppm; carbonatites: 316–678 ppm), excluding the one dolomite sample with a Nd content of 1074 ppm. The plot of $^{143}\text{Nd}/^{144}\text{Nd}$ initial ratio against total Nd for apatites is shown in Fig. 6 and the later phoscorites and carbonatites have the highest $^{143}\text{Nd}/^{144}\text{Nd}$ ratios and Nd contents.

Like most other carbonatites, the Nd and Sr isotope data from the Kovdor apatites lie well within the depleted quadrant of the Sr-Nd correlation plot, and broadly form two groups corresponding to their relative ages. All of the new Nd and Sr isotope data from Kovdor are plotted in Fig. 7 and shown in this diagram is the Kola Carbonatite Line (KCL), based on initial ratios from carbonatites from Finland and Russia (Kramm 1993). The earlier group from Kovdor clearly lie along this line, and cluster towards that part of the line with low $^{143}\text{Nd}/^{144}\text{Nd}$ and low $^{87}\text{Sr}/^{86}\text{Sr}$ values.

Fig. 5a, b Plot of $^{87}\text{Sr}/^{86}\text{Sr}$ versus Sr: **a** for apatite from phoscorites and carbonatites; **b** for calcite (C), dolomite (D) and apatite (Ap) from phoscorite and carbonatites. Dashed curve represents mixing line between two end-members, one with 2500 ppm Sr and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70335 and second with 9000 ppm Sr and $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70365. Tie-lines join apatite-calcite data from same rock. (Filled symbols phoscorites, open symbols carbonatites)



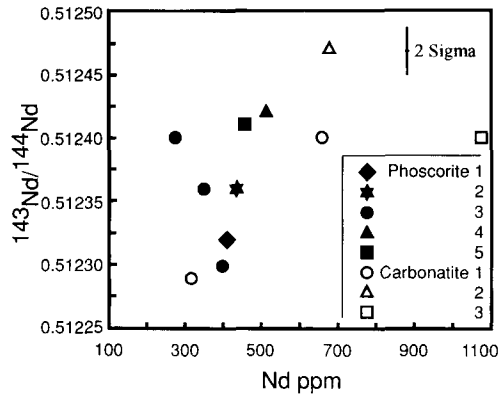


Fig. 6 Plot of $^{143}\text{Nd}/^{144}\text{Nd}$ versus Nd for apatite from phoscorites and carbonatites. (Filled symbols phoscorites; open symbols carbonatites)

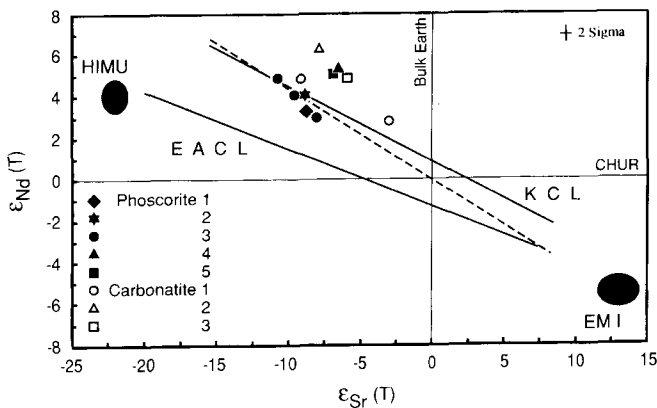


Fig. 7 Plot of $\epsilon_{\text{Nd}}(T)$ versus $\epsilon_{\text{Sr}}(T)$ for the Kovdor data. Assumed age of the complex is 380 Ma. Shown for comparison are the Kola Carbonatite Line (Kramm 1993), the East Africa Carbonatite Line (Bell and Blenkinsop 1987; Bell and Peterson 1991; Simonetti and Bell 1994) and HIMU and EM I components (Hart et al. 1986). Dotted line represents the Kola Carbonatite Line without the sample from the Vuorijarvi massif. Note that the EACL is based on data from young carbonatites (mostly < 40 Ma) and only the slope can be used for comparative purposes. The parameters used for Nd are CHUR values, and those for bulk Earth Sr are: present day $^{87}\text{Sr}/^{86}\text{Sr} = 0.7045$, initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio = 0.69898, $^{87}\text{Rb}/^{86}\text{Sr} = 0.0827$, and the age of the Earth = 4.55 Ga. (Filled symbols phoscorites; open symbols carbonatites)

Discussion

For phoscorites associated with carbonatites in alkaline complexes two models of origin have been proposed, one involves hydrothermal-metasomatic processes and the other magmatic.

A hydrothermal-metasomatic origin for phoscorites was discussed by Kukharenko et al. (1965), Kapustin (1980) and Epshteyn and Danilchenko (1988). These authors argued for a process of infiltration metasomatism, under the influence of $\text{CO}_2\text{-H}_2\text{O}$ fluids related to carbonatites. Such a model was based on the

zoned distribution of phoscorites, with decrease in the number of minerals from the outer to inner zone of bodies, the presence of relicts of the host rocks in the marginal parts of phoscorites, the dependence of the mineral composition of phoscorites on those of the host rocks, and the presence in all varieties of phoscorites of replacement textures. Epshteyn and Danilchenko (1988) believe that the formation of magnetite in phoscorites is the result of replacement of ijolite by carbonatite—"theoretically enough iron could be removed from four volumes of ijolite to form one volume of an iron deposit containing about 25% iron".

In contrast, other workers have proposed that phoscorites can be of magmatic origin (Kharlamov et al. 1981; Lapin 1982; Lapin and Vartiainen 1983; Eriksson 1989). Evidence for the magmatic origin of phoscorites (Egorov 1993) includes the morphology of phoscorite bodies, textural and structural features, mineralogical composition, geothermometry and experimental data. Spherulitic and orbicular structures in phoscorites of the Sokli and Vuorijarvi massifs were attributed by Lapin (1982) and Lapin and Vartiainen (1983) to liquid immiscibility. The spherulites and orbicules contain mainly forsterite and magnetite with subordinate apatite, phlogopite or tetraferriphlogopite and calcite, that occur within a calcite matrix. Lapin and Vartiainen (1983) concluded that "liquid immiscibility is favoured as a mechanism ..." of phoscorite separation from a carbonatitic melt enriched in phosphorus, iron and silicate components, while crystallization differentiation probably plays an important role in carbonatitic melt depleted in those components. Experimental studies in the system magnetite-apatite-diorite have shown immiscibility between magnetite-apatite melt and silicate melt (Philpotts 1967). Geothermometric data from melt and fluid inclusions in forsterite, apatite and calcite from phoscorites of the Kovdor massif indicate a formation temperature of 660–940°C (Kharlamov et al. 1981). The existence of lavas made up mainly of magnetite, from the Laco area, Chile (Park 1961; Henriquez and Martin 1978), shows that magnetite-rich rocks can be produced by magmatic process.

Our geological and petrographical observations suggest that the Kovdor phoscorites are magmatic rocks and Sr-Nd isotope data of apatite from the phoscorites indicate their derivation from a mantle source that has a higher time integrated Sm/Nd ratio than CHUR and a lower Rb/Sr ratio than bulk Earth. However, some phoscorites were also metasomatized by a fluid that accompanied igneous activity. The evidence for this includes changes of mineral assemblages of phoscorites. Secondary phlogopite or tetraferriphlogopite and tremolite are present at the contacts of phoscorites and carbonatites.

The most significant feature to emerge from the data is the large variation shown by both initial Nd and initial Sr ratios for the complex as a whole. This finding

rules out any model that invokes a closed-system behaviour of a single parental magma. These isotopic variations may be attributed to contamination of the melt, but the high Sr and Nd concentrations and low $\delta^{18}\text{O}$ values of carbonatites in general rule out any significant contamination by continental crust. The fact that those samples with the highest Sr and Nd contents are also the most radiogenic suggests that the isotopic variations result from mixing between two mantle-end members rich in both elements. If sample KV 25 from the C I carbonatites is excluded, simple binary mixing between two end-members, one with 2500 ppm Sr and a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70335 and a second with 9000 ppm Sr and a $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.70365 could produce the ranges seen in the Sr isotopic data for apatite from Kovdor phoscorites and carbonatites (Fig. 5a).

Mantle mixing has been invoked to explain many of the isotopic variations that have been observed in carbonatites, particularly those from East Africa (Bell and Blenkinsop 1987) and Kola (Kramm 1993). The East African data fall close to the LoNd array of Hart et al. (1986), suggesting that HIMU and EM I mantle components were involved. This has now been substantiated still further by Pb isotope data (K. Bell and G.R. Tilton, unpublished data). The slope of the Kola Carbonatite Line (-0.365 ± 0.022) is much steeper than the one from East Africa carbonatites (EACL = -0.285 ± 0.019), and Kramm correctly concludes that the end-members cannot be the same. If the isotopic data ($^{87}\text{Sr}/^{86}\text{Sr}_{\text{initial}} = 0.70466$ and $^{143}\text{Nd}/^{144}\text{Nd}_{\text{initial}} = 0.51206$) from the Vuorijarvi carbonatite (sample VJ K 10B, Table 1 in Kramm (1993), is omitted from the calculation then the calculated slope of the KCL is even steeper -0.443 ± 0.050 . Although we do not have good grounds for rejecting this data point, $^{87}\text{Sr}/^{86}\text{Sr}$ initial ratios from other carbonatites and phoscorites of that complex are substantially lower, with initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios in apatite ranging from 0.7028 to 0.7038 (Dudkin et al. 1994), which suggests open-system behaviour for this one sample and the involvement of a more radiogenic source.

The data from the early intruded phoscorites and carbonatites of the Kovdor complex plot along KCL, supporting Kramm's contention that most of the carbonatite complexes from the Kola alkaline province involve mixing of two mantle reservoirs. The fact that the Kovdor data do not all plot along the line, suggests that different magma batches were involved in generating the phoscorites and carbonatites. Although such pronounced variations have rarely been observed in carbonatites from the same complex, similar conclusions have been made about the silicate rocks from Oldoinyo Lengai (Bell and Dawson 1995), the only active carbonatite volcano. Their data show a marked variation and cover almost two thirds the length of the EACL, which implies that not only was each magma batch independent but that each involved the mixing together of different proportions of the same two end-

members with similar Nd/Sr ratios. The same is probably true for the melts responsible for the Kovdor carbonatites and phoscorites.

It is not clear why the data from the later group of carbonatites and phoscorites do not lie on the KCL, but there are two possible explanations for this. The first, involves interaction between carbonatite and phoscorite and meteoric water, the net result of which would be to leave the Nd isotopic composition relatively unchanged, and increase the $^{87}\text{Sr}/^{86}\text{Sr}$ ratios. This is unlikely for following reasons. Firstly, the calcites have relatively primitive $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ values, and secondly both apatite and calcite are rich in Sr. A second model is simply to invoke a third mantle component that has a higher Sr content and higher $^{87}\text{Sr}/^{86}\text{Sr}$ ratio than those two end-members that generated the earlier suites.

Conclusions

The Sr and Nd isotope data of the Kovdor carbonatites and phoscorites reveal a complex history, involving the mixing of magmas derived from isotopically distinct sources. The most important findings in this study are:

1. The evolutionary history of the phoscorites and carbonatites from Kovdor cannot be explained by simple magmatic differentiation assuming closed chemical conditions.
2. The isotopic data fall within the depleted quadrant of the Nd-Sr correlation plot and are consistent with findings from carbonatites from other continents that indicate derivation from a mantle source that has a higher time integrated Sm/Nd ratio than CHUR and a lower Rb/Sr ratio than bulk Earth.
3. The isotopic similarities in terms of Sr and Nd between the Kovdor carbonatites and phoscorites suggest that the two are genetically related.
4. Incursions of carbonatitic melts with quite different isotopic signatures invaded the Kovdor massif during the emplacement of the C I carbonatites.
5. At least three mantle components are required to produce the isotopic variations that have been documented from Kovdor.

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