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ARTICLE

Oxygen isotope systematics of gem corundum deposits in Madagascar: relevance for their geological origin

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Abstract The oxygen isotopic composition of gem corundum was measured from 22 deposits and occurrences in Madagascar to provide a gemstone geological identification and characterization. Primary corundum deposits in Madagascar are hosted in magmatic (syenite and alkali basalt) and metamorphic rocks (gneiss, cordieritite, mafic and ultramafic rocks, marble, and calc-silicate rocks). In both domains the circulation of fluids, especially along shear zones for metamorphic deposits, provoked in situ transformation of the corundum host rocks with the formation of metasomatites such as phlogopite, sakenite, and corundumite. Secondary deposits (placers) are the most important economically and are contained in detrital basins and karsts. The oxygen isotopic ratios ($^{18}O/^{16}O$) of ruby and sapphire from primary deposits are a good indicator of their geological origin and reveal a wide range of $\delta^{18}O$ (Vienna Standard Mean Ocean Water) between 1.3 and 15.6‰. Metamorphic rubies are defined by two groups of $\delta^{18}O$ values in the range of 1.7 to 2.9‰ (cordieritite) and 3.8 to 6.1‰ (amphibolite). "Magmatic" rubies from pyroxenitic xenoliths contained in the alkali basalt of Soamiakatra have $\delta^{18}O$ values ranging between 1.3 and 4.7‰. Sapphires are classified into two main groups with $\delta^{18}O$ in the range of 4.7 to 9.0‰ (pyroxenite and feldspathic gneiss) and 10.7 to 15.6‰ (skarn in marble from Andranondambo). The $\delta^{18}O$ values for gem corundum from secondary deposits have a wide spread between −0.3 and 16.5‰. The ruby and sapphire found in placers linked to alkali basalt environments in the northern and central regions of Madagascar have consistent $\delta^{18}O$ values between 3.5 and 6.9‰. Ruby from the placers of Vatomandry and Andilamena has $\delta^{18}O$ values of 5.9‰, and between 0.5 and 4.0‰, respectively. The placers of the Ilakaka area are characterized by a huge variety of colored sapphires and rubies, with $\delta^{18}O$ values between −0.3 and

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16.5‰, and their origin is debated. A comparison with oxygen isotope data obtained on gem corundum from Eastern Africa, India, and Sri Lanka is presented. Giant placer deposits from Sri Lanka, Madagascar, and Tanzania have a large variety of colored sapphires and rubies with a large variation in $\delta^{18}O$ due to mingling of corundum of different origin: mafic and ultramafic rocks for ruby, desilicated pegmatites for blue sapphire, syenite for yellow, green, and blue sapphire, and skarn in marbles for blue sapphire.

Keywords Madagascar · Gondwana · Oxygen isotopes · Corundum deposits · Ruby · Sapphire

Introduction

For over 10 years Madagascar was a major gem-producing country (extraLapis 2001). Many of the minerals and gemstones that have made Madagascar famous came from pegmatites with deep-blue colored beryl and polychrome tourmaline, rhodizite, morganite, orthoclase (Lacroix 1922a), londonite (extraLapis 2001; Laurs et al. 2002), pezzotaite (Laurs et al. 2003), and emerald from schist-type deposits (Schwarz 1994; Moine et al. 2004).

Corundum occurrences in Madagascar were reported from different parts of the island (Lacroix 1922a) and were exploited as refractory material (Besairie 1966). Before 1993, only a few gem corundum deposits were known in Madagascar in comparison with the main world producers of East Africa, India, Sri Lanka, Myanmar, Thailand, and Australia. Since 1993, large amounts of gem-quality sapphires were recovered from the Andranondambo metamorphic skarn-type deposit in the southern part of Madagascar (Rakotondrazafy 1995) and from alluvial deposits linked to basaltic rocks in the northern region (Schwarz et al. 2000). Most of the economic ruby arrived on the market at the end of 2000 from the secondary deposits of Andilamena and Vatomandry in the eastern part of the island (Schwarz and Schmetzer 2001). The discovery in late 1998 of the first giant alluvial sapphire and ruby deposits in the Ilakaka area assured a long future for the recovery of large quantities of fine gemstones in Madagascar. These new discoveries attracted a large number of dealers from Southeast Asia and Africa, purchasing gems at low prices from hand miners and exporting Malagasy rough to Sri Lanka and Thailand. The opportunity to establish the authentic source of good quality untreated ruby and sapphire was lost due to the low prices of the Malagasy rough material and the early good results obtained on the production by the Thai treatments. Large quantities of gem quality corundum from Madagascar are mixed with others on the international market.

The present study describes the use of oxygen isotopic ratios ($^{18}O/^{16}O$) in the lattice oxygen of ruby and sapphire to provide a gemstone geological identification and categorization. This paper presents an overview of the different types of corundum deposits in Madagascar based on their geologic setting, structural features, ore paragenesis, and on stable oxygen isotopes of corundum. This study is a first step in the identification of the locality of origin of Malagasy gem corundum related to primary deposits, and it will help to assess the geologic origin of sapphire and ruby crystals mingled in the same alluvial basin. A comparison with the oxygen isotope data obtained on gem corundum from Eastern Africa, India, and Sri Lanka is presented.

Materials and methods

A total of 67 samples of natural ruby and sapphire were collected from 22 deposits and occurrences in Madagascar (Fig. 1). A first series of 16 samples analyzed within a worldwide context of oxygen isotope characterization were published by Giuliani et al. (2005). The samples used for the present study were collected by the authors in the field. Two types of corundum were analyzed: (1) gems hosted in rocks (primary deposits) and (2) gems from placers (secondary deposits).

Oxygen isotope analyses of corundum were performed using a modification of the laser fluorination technique described by Sharp (1990). The analyzed samples were of gem quality or generally free of inclusions; the analyses were often duplicated or triplicated to check for isotopic heterogeneity, analytical artifacts, and possible contamination by solid inclusions. The method involves complete reaction of ~1 mg of powdered corundum, heated by a CO_2 laser, with ClF_3 as the fluorine reagent. The released oxygen is passed through an inline Hg diffusion pump before conversion to CO_2 on platinized graphite. The yield is measured by a capacitance manometer, and the gas-handling vacuum line is connected to the inlet system of a dedicated VG PRISM 3 dual inlet isotope ratio mass spectrometer. Oxygen yields differing significantly from the theoretical value of 14.07 μmol/mg were taken as likely evidence of analytical artifact. Precision and accuracy on quartz standards are ±0.1‰ (1σ), and duplicate analyses of sapphire samples suggest similar precision and accuracy for this material. Data are reported in the conventional delta notation relative to V-SMOW (Vienna Standard Mean Ocean Water). $\delta^{18}O(‰)=\left[\left(\left(R_{\text{sample}}/R_{\text{standard}}\right)-1\right)\right] \times 10^3$, where R is the isotopic ratio $^{18}O/^{16}O$.

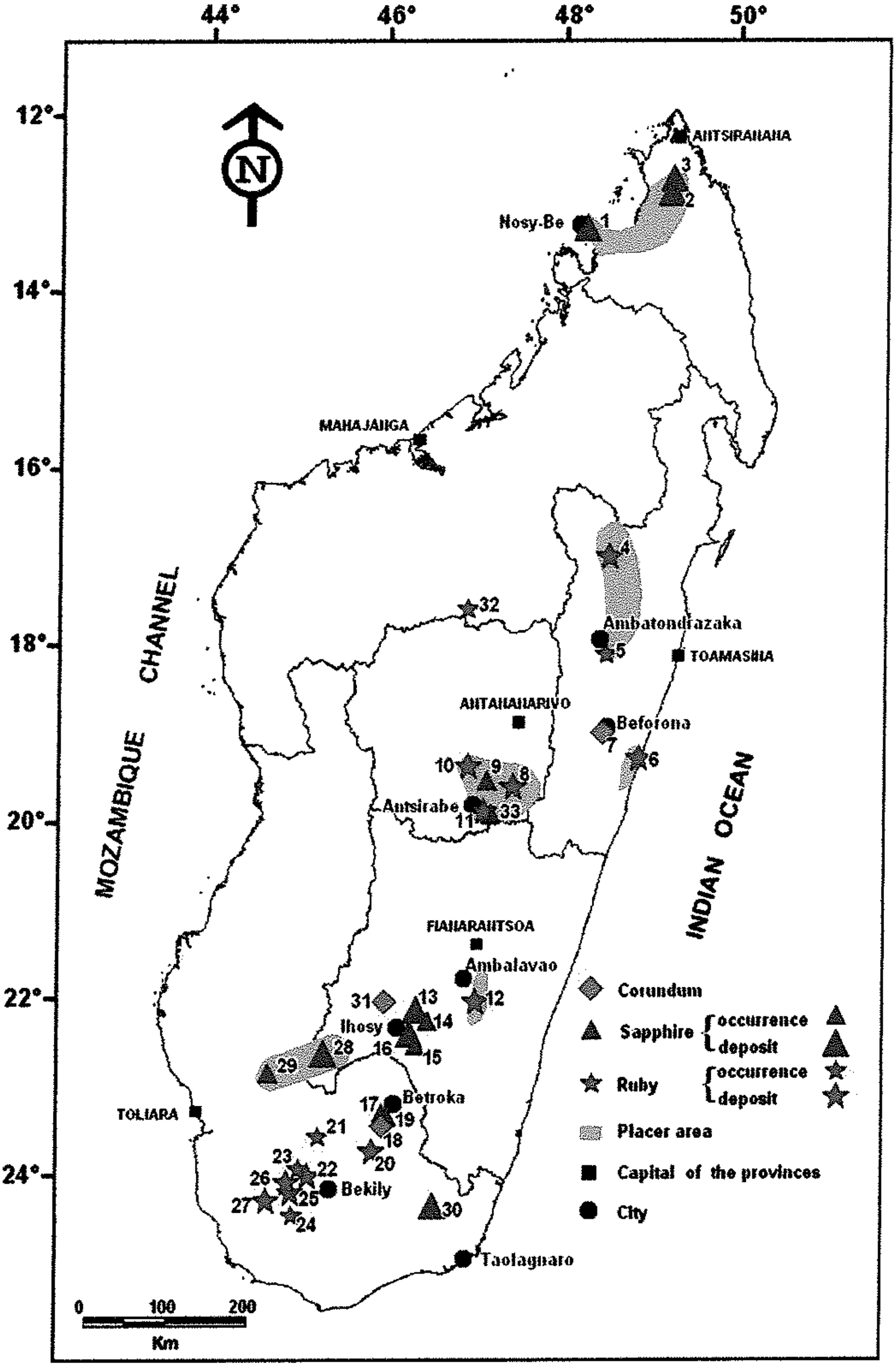


Fig. 1 The sapphire, ruby, and corundum occurrences and deposits of Madagascar. *1* Nosy Be, *2* Ambondromifehy, *3* Anivorano, *4* Andilamena, *5* Didy, *6* Vatomandry, *7* Ambohitranefitra (Beforona), *8* Antsahanandriana, *9* Mandrosohasina, *10* Faratsiho, *11* Soamiakatra, *12* Miarinarivo, *13* Zazafotsy, *14* Sakalalina, *15* Ambinda (Ihosy), *16* Sahambano, *17* Ambinda (Betroka), *18* Vohidava (Voronkafotra), *19* Iankaroka, *20* Ambatomena, *21* Ianapera, *22* Fotadrevo, *23* Anavoha, *24* Maniry, *25* Gogogogo, *26* Vohitany, *27* Ejeda, *28* Ilakaka, *29* Sakaraha, *30* Andranondambo, *31* Sakeny, *32* Andriba, *33* Anjomakely

Geological framework of corundum deposits

Precambrian rocks are exposed in the eastern two thirds of Madagascar whereas the western third is composed of late Paleozoic to recent sedimentary rocks (Fig. 2). The Precambrian basement includes remnants of early crust (de Wit 2003), which were intensely reworked between 950 and 450 Ma, during Pan-African tectonometamorphic events (Kröner 1984). The collisional processes between East and West Gondwana resulted in the formation of Neoproterozoic mobile belts (~650 Ma), mostly metamorphosed at high grade. The metamorphic rocks are high and

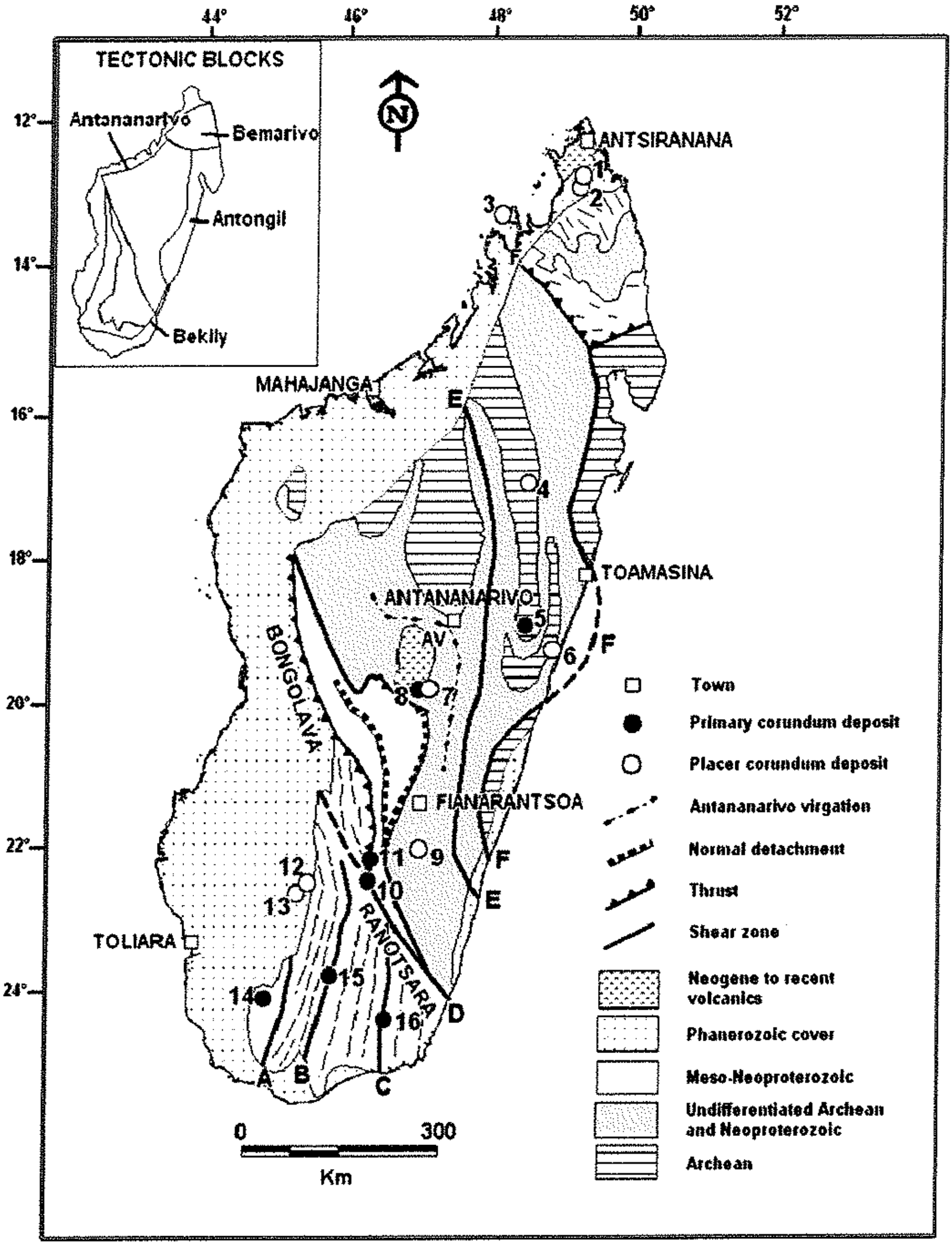


Fig. 2 The Precambrian in Madagascar with its main tectonic structures and main placers and primary gem corundum deposits (modified from de Wit 2003). Shear zones: *A* Ampanihy, *B* Vorokafotra, *C* Tranomaro, *D* Ranotsara-Bongolava, *E* Ifondiana-Angavo, *F* Betsimisaraka. Placer deposits: *1* Anivorano, *2* Ambondromifehy, *3* Nosy Be, *4* Andilamena, *6* Vatomandry, *7* Kianjanakanga-Mandrosohasina, *9* Miarinarivo, *12* Ilakaka, *13* Sakahara. Primary deposits: *5* Ambohitranefitra (Beforona), *8* Soamiakatra, *10* Sahambano, *11* Zazafotsy, *14* Ejeda-Fotadrevo area, *15* Ambatomena, *16* Andranondambo

low pressure granulites, well exposed throughout southeast Madagascar. The granulitic terranes are divided into four major lithostratigraphic groups (Besairie 1967; de Wit 2003) corresponding to the juxtaposition of tectonic blocks of different crustal levels (Martelat et al. 1997, 2000; de Wit et al. 2001). This patchwork is due to the relative movements of major ductile shear zones reflecting a crustal scale strike slip system. Rocks in all blocks suffered metamorphism between 850 and 750°C. The pressure shows an E–W decrease from 11 to 8 kbar in the west to 5 to 3 kbar in the east (Nicollet 1990). Granitoids are abundant in the eastern part whereas anorthosites and metabasites are abundant in the west. Most of the Malagasy primary gem ruby deposits are closely associated with basic ultrabasic complexes in the western area (Mercier et al. 1999a) whereas gem quality blue sapphires from the Andranondambo deposit are linked to veins in skarns formed at the contact between granite and marble in the eastern area (Rakotondrazafy et al. 1996). Recently, colored sapphire crystals of economic interest were found in biotitized gneisses within ductile shear zones of first and second order (Razanatseheno et al. 2005).

From the Upper Carboniferous to Mid-Jurassic (300–180 Ma), Madagascar was adjacent to East Africa with the Seychelles to the northeast (Lawler et al. 1992). This was the period of formation of basins in connection with terrestrial rifts in Africa. Sedimentation covers about one third of Madagascar along its western extensional margin (de Wit 2003), in sequences divided into the Sakoa, Sakamena, and Isalo Groups. The Isalo Group represents the upper part of the rift sequences, 1 to 6 km thick, made up of conglomerates and white sandstones, capped by Lower Triassic red-bed sequences. These detrital sediments marked an active period of rifting and terrestrial sedimentation with the formation of giant gem paleoplacers in the Isalo sandstones. The deposits consist mainly of blue and colored sapphires with few rubies and other semiprecious stones such as alexandrite, topaz, and cat's eye chrysoberyl. These Triassic concentrations were reworked during the Quaternary, and now mineralized gravels are concentrated in the rivers.

During the Upper Cenozoic, flood (dominantly MORB-like) basalts covered a great part of the island (Melluso et al. 2001), and in the Neogene–Quaternary, typical ocean island basalts were widespread in the central and northern part of Madagascar (Melluso and Morra 2000). During the Upper Cenozoic, basalt flows dominated, but basalt–rhyolite associations are common (Androy volcano in the south). The volcanic rocks range from picrites to andesites but their composition varies depending on their original mantle sources and the degree of continental contamination. Neogene–Quaternary alkaline basalts occur mainly in the Ankaratra mountains of central Madagascar and in the Ambohitra igneous province in the north, which includes the Nosy Be archipelago. These large Late Cenozoic alkali basalt provinces are probably the source of the large quantities of blue, green, and yellow sapphires ("BGY sapphires" of Sutherland et al. 1998) recovered from consolidated alluvial deposits derived from eroded basalts and surrounding formations in the Antsiranana province (Ambondromifehy deposit) and Nosy Be (Befotaka deposit). The crystal morphology, internal growth patterns, mineral inclusions, and trace element contents of these sapphire crystals are typical of "basaltic-magmatic" sapphires found worldwide (Schwarz et al. 2000), but the nature of their parent host rocks is still in debate. The corundum deposits in the Ankaratra area, close to Antsirabe City, are also alluvial except for the deposit of Soamiakatra, where ruby is found in pyroxenitic xenoliths included in alkali basalts (Rakotosamizanany 2003).

The corundum deposits of Madagascar

Primary corundum deposits

These are hosted in magmatic and metamorphic rocks (Table 1). In both domains, circulation of fluids provoked in situ transformation of the host rocks with the formation of metasomatic rocks as sakenite (plagioclasite) and corundumite (Lacroix 1922a, 1941). Magmatic deposits are hosted in syenite and alkali basalts in the Beforona and Antsirabe areas, respectively (Fig. 1). Metamorphic-hosted deposits are located in the granulitic domain of southern Madagascar (Fig. 3). Corundum deposits are strongly associated with major and minor shear zones. These structures acted as preferential fluid pathways that resulted in metasomatism of the host rocks. The nature of the parental host rock varies from feldspathic gneisses (Zazafotsy, Sahambano, and Ambinda Sud), cordieritites (Iankaroka and Ambatomena), amphibolites and anorthosites (Ejeda, Fotadrevo, Vohitany, and Gogogogo) to impure marbles (Tranomaro). The sakenites described by Lacroix (1941) are found in paragneiss with intercalations of amphibolites, pyroxenites, and impure marbles (Sakeny, Ejeda, Vohidava, and Tranomaro occurrences) and consist of plagioclasite veins or segregations with ± spinel, ± corundum, ± hibonite, and ± phlogopite.

Placer deposits

These are exploited in volcanic and sedimentary environments:

(1) In basaltic provinces: The sapphire placer deposits in the Antsiranana province are located about 70 km south of Antsiranana City, in the Anivorano and Ambondromifehy area and on Nosy Be Island (Fig. 1). The region is mostly covered by a 3,500-km^2 area of volcanic rocks that date from the early Tertiary period to the Quaternary. The 35-km-wide volcanic flows are formed by successive eruption of basalts, tuffs, pozzolanas, and pyroclastites, which contain enclaves mainly of peridotite and lherzolite. Recent prospecting and fieldwork failed to locate any sapphire-bearing basalt flows in the Montagne d'Ambre volcanics. Nevertheless, Lacroix (1922a) noted the presence of a crystal of sapphire, zircon, and spinel associated with hornblende and syenitic enclaves in basaltic scoria at Lake Mahery from the Montagne d'Ambre, and one crystal at Nosy Mitsio Island, which is constituted of basanite, phonolite, and trachyte. Sapphire-bearing alluvial materials were deposited in voids and cracks of a karst developed on Jurassic Ankaratra limestone and arenites that lie south of the volcanic massif of the Montagne d'Ambre (Schwarz et al. 2000). All gem-bearing sediments forming paleoplacers are cemented by secondary carbonates (deposits of Ambohangimamy, Maromikotra, Sanaderikely contained in limestones) or silica (deposit of Maventibao contained in arenites).

Table 1 Synthesis of the main geologic and isotopic features of the different types of primary corundum deposits in Madagascar

	Deposit							
	Sahambano (S) Zazafotsy (Z)	Iankaroka	Ambatomena	Vohitany (V) Ejeda (E) - Gogogogo (G)	Andranondambo	Beforona	Sakeny (Sk) Vohidava (Vo)	Soamiakatra
References	1, 2, 3, 4	1, 5	1, 3	1, 6, 7, 8, 9	1, 10, 11, 12	13, 14	13, 15	16
Genetic model	MM	M-MM	MM	M-MM	MG-HM	MG or MG-HM?	M-MM?	M
Tectonic unit	S: Southern Madagascar Z: Itremo sheet	Southern Madagascar	Southern Madagascar	Southern Madagascar	Southern Madagascar	Antananarivo block	Southern Madagascar	Antananarivo block
Formation and/or series	S: Tranomaro group	Androyan series	Androyan series	Vohibory series	Tranomaro group	Contact Manampotsy and Beforona groups	Androyan series	Ambatolampy series
	Z: Vohimena series				Anosyan granite			Ankaratra volcanism
Host rock	Metamorphic feldspathic gneiss intercalated with leptynite	Metamorphic cordieritite intercalated with charnockite	Metamorphic cordieritite (and pegmatite) in charnockites	Metamorphic	Skarn	Magmatic syenite	Metamorphic sakenite vein	Volcanic pyroxenite enclave in alkali basalt
				a. Amphibolite and pyroxenite within M-UM (E-G-V)^{a} b. Anorthosite layers (E-G)^{a} c. Metasomatized pegmatite in M-UM (V)^{a}	Fissural skarn			
Wall rocks	Biotitized gneisses	Cordieritite	Metasomatized cordieritite	a. Amphibolite^{a}	Impure marble	Biotite gneiss	Paragneiss	Basalt
	Biotitite	Fissural Mg-biotitite		b. Anorthosite^{a}	Pyroxenite	Micaschist	Amphibolite	
				a. Biotitite and plagioclasite^{a}	Calc-silicate gneisses		Pyroxenite	
Mineralization control	Shear zone Fluid–rock interaction	Shear zone Fluid–rock interaction	Shear zone Fluid–rock interaction	Shear zone Fluid–rock interaction	Veinlet in skarn Fluid–rock interaction	Irregular vein Lens-like	Vein	Pyroxenite

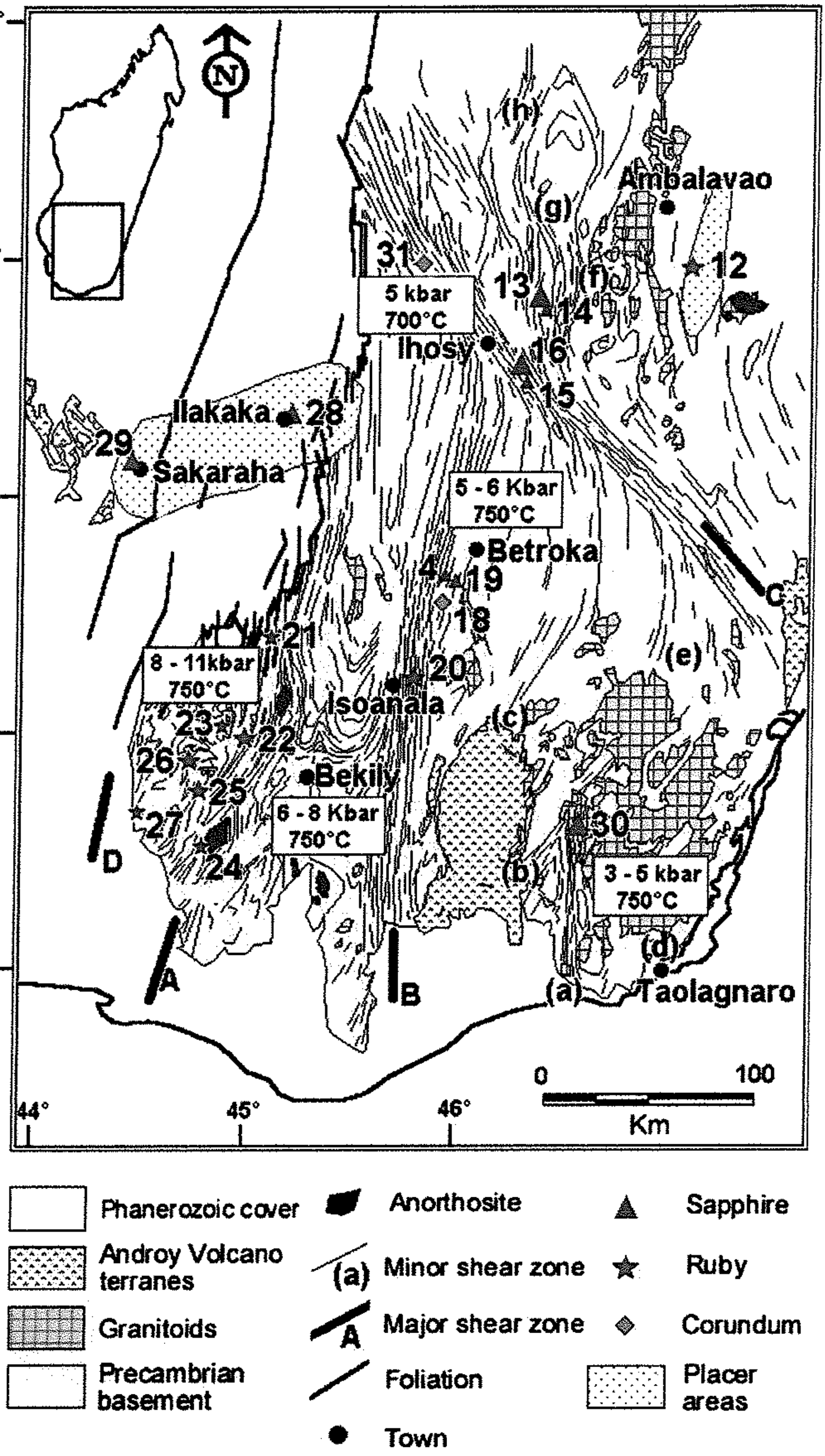
Typical mineral assemblage	Biotite–sapphire–saphirine–plagioclase–K-feldspar–garnet–spinel	Phlogopite–cordierite–sapphire–tourmaline–spinel–saphirine	Cordierite–rutile–phlogopite–saphirine–plagioclase–ruby	(V): hornblende–ruby–plagioclase–spinel–phlogopite	K-feldspar–sapphire–F-apatite–calcite–phlogopite	Biotite–sillimanite–albite–sapphire–microcline	Sapphirine–sapphire–spinel–pyroxene–plagioclase–edenite	Clinopyroxene–ruby–amphibole–anorthite–scapolite–garnet
Metamorphism	Granulite facies	Granulite facies	Granulite facies	Granulite facies	Granulite facies	Granulite facies	Granulite facies	Granulite facies
	T=700°C P=5 kb (17)	T=750°C P=5–6 kb (17)	T=750°C P=5–6 kb (17)	T=730–870°C P=9–11 kb (6, 8)	T_{saphir}=500°C P_{saphir}=2 kb (18)	T=? P=?	T=700°C P=4–5 kb (13, 17)	T=1,100°C P=20 kb (16)
Age of the mineralization	Ar–Ar biotite (19) S: 492±5 Ma[a] Z: 494±5 Ma[a]	Ar–Ar biotite (19) No age (disturbed spectrum)	Ar–Ar biotite (19) 487±4 Ma[a]	Ar–Ar biotite Vohitany (19) No age (disturbed spectrum)	U/Pb zircon (20, 21) 565±15 Ma (20) 516±10 Ma (21)[a] 523±5 Ma (20)[a]	?	?	Basalt (22) Miocene to Quaternary
Corundum	Multicolored sapphire	Polychrome sapphire	Ruby	Ruby	Light to dark blue, pink sapphires	Red to purplish blue to gray sapphire	Gray-white to yellow sapphire	Ruby
$\delta^{18}O$ corundum (‰, V-SMOW)	S: 5.9±0.3 (n=5) Z: 8.9±0.1 (n=2)	2.05±0.5 (n=2)	2.9 (n=2)	V: $5.4<\delta^{18}O<6.1$ (n=2) E: 5.0, 5.9, G: 3.8	$10.1<\delta^{18}O<10.9$ (n=4) $14.0<\delta^{18}O<15.6$ (n=4)	8.1	Vo: 5.8 Sk: 4.9	$1.25<\delta^{18}O<4.7$ (n=2)

M Metamorphic, *MM* Metamorphic metasomatism, *MG* Magmatic, *MG-HM* Magmatic hydrothermal metasomatism, *M-UM* Mafic-ultramafic rocks

References: *1* Razanatseheno et al. (2005), *2* Ralantoarison (2006), *3* Andriamamonjy (2006), *4* Pezzota (2005), *5* Koivula et al. (1992), *6* Nicollet (1986), *7* Nicollet (1990), *8* Mercier et al. (1999a), *9* Pili (1997a,b), *10* Rakotondrazafy et al. (1996), *11* Rakotondrazafy (1995), *12* Schwarz et al. (1996), *13* Lacroix (1922a,b), *14* Rantoanina (1962), *15* Devouard et al. (2002), *16* Rakotosamizanany (2003), *17* Nicollet (1985), *18* Ravololomiandrinarivo et al. (1997), *19* this work, *20* Paquette et al. (1994), *21* Andriamarofahatra and de La Boisse 1986, *22* Besairie and Collignon (1972)

[a] Sapphire mineralization

Fig. 3 Structural and lithological sketch map of southeast Madagascar with the location of the corundum deposits (modified from Martelat et al. 2000). Number of deposits as in legend of Fig. 1. Major shear zones referred as *A* Ampanihy, *B* Beraketa, *C* Ranotsara, *D* shear zone of the Phanerozoic cover. Minor shear zones referred as subpanels **a** to **h**. The Pressure (kbar) and Temperature (°C) of metamorphism are from Moine et al. (1985), Ackermand et al. (1989), and Nicollet (1990)



Two new sapphire deposits were discovered in 2001, i.e., on Nosy Be Island and in the Andovokonko area on the Ambato Peninsula (Ramdohr and Milisenda 2004). The BGY sapphires and zircon are found in alluvial loess in a layer formed of basalt, pebbles located 1 m above the granitic bedrock. At the Andovokonko deposit, sapphire is found on the basalt surface covered by calcrete crust and in tidal flats.

Alluvial corundum deposits in the south-eastern part of the Ankaratra volcanic massif were described by Lacroix (1922a). Gem corundum was extracted at different localities: ruby at Andranomadio, ruby and sapphire at Andriankely, and blue, green, and yellow sapphires at Ampitatafika, Vohimena, Ambatotsipihana, Maroparasy, Sambaina, Ambohimandroso, Iankiana, Vontovorona, Mahanoro, Faratsiho, Vakinakaratra, and Belambo. Nowadays, recent alluvial placers in river and soils are mined by locals. The BGY sapphire deposits of Kianjanakanga–Mandrosohasina and the ruby deposit of Antsabotraka are mined from sedimentary deposits made of basaltic and phonolitic pebbles cemented by lateritic soils.

(2) The giant placer of Ilakaka in the Isalo sedimentary basin: The Ilakaka mining district is located in the Isalo massif, between the cities of Sakaraha and Ilakaka (Figs. 1 and 3). Other districts are found north of Ilakaka and near Bezaha, 120 km southwest of Ilakaka (Garnier et al. 2004). The deposits produce very fine blue, pink, blue-violet, violet, purple, orange, yellow, and translucent sapphires with zircon, alexandrite, topaz, garnet, spinel, andalusite, and tourmaline.

(3) Other placer deposits of unknown origin: New deposits found in 2000 in the area of Vatomandry and Andilamena (deposits 4 and 6 in Fig. 1) drastically changed ruby production in Madagascar. There is no detailed geological information about the two mining areas and the ruby host rock is unknown. The Miarinarivo placer is located 30 km south of Ambalavao City (Fig. 3). The deposit consists of pinkish to brownish corundum in a large volume of alluvium. The crystals are not strongly rounded and their hexagonal habit was preserved during alluvial transport.

The oxygen isotope composition of corundum

Results

$\delta^{18}O$ values for gem corundum from primary deposits range from 1.3 to 15.6‰ (Table 2). Metamorphic rubies are defined by two sets of $\delta^{18}O$ values (Fig. 4). The first group, in the range of 3.8–6.1‰ (n=4), corresponds to ruby hosted by amphibolites for the deposits concentrated in the Vohibory unit. It includes those of Gogogogo, Ejeda, and Vohitany. The second group, between 1.7 and 2.9‰ (n=4), corresponds to polychrome sapphire and ruby contained in the cordieritites of Iankaroka and Ambatomena, respectively.

"Magmatic" ruby samples from xenoliths contained in the alkali basalt of Soamiakatra have $\delta^{18}O$ ranging between 1.3 and 4.7‰ (n=2).

Sapphires are classified into three sets of $\delta^{18}O$ values (Fig. 4): (1) Blue sapphire from Andranondambo, hosted in skarn, show two ranges of $\delta^{18}O$ values, i.e., 10.1 and 10.7‰ (n=3) for stage 1 of skarn metasomatism, and between 14.0 and 15.6‰ (n=4) for stage 3 of vein–skarn. (2) Colored sapphire from Sahambano, Ambinda Sud, and Zazafotsy, contained in biotitites resulting from the metasomatism of feldspathic gneisses, has $\delta^{18}O$ values between 5.6 and 9.0‰ (n=8). Although geologically similar, these two deposits present different sets of $\delta^{18}O$ values: The Zazafotsy sapphire has an oxygen isotopic composition between 8.8–9.0‰ (n=2), whereas at Sahambano, the $\delta^{18}O$ of colored sapphire is between 5.6 and 6.5‰ (n=5), and at Ambinda Sud at 7.6‰ (n=1). The pinkish to reddish sapphire from the syenite of Beforona has a $\delta^{18}O$ of 8.1‰ (n=1); the light to blue-green sapphire originating from the Vohidava sakenite has a $\delta^{18}O$ of 5.8‰ (n=1). (3) Gray to colorless sapphire contained in the biotitized pyroxenite from Ambinda gave a $\delta^{18}O$ value of 4.7‰.

$\delta^{18}O$ values for gem corundum from secondary deposits have a wide spread between −0.3 and 16.5‰ (Table 2). Ruby and sapphire found in placers linked to alkali basalt environments in the northern and central regions of Madagascar have consistent $\delta^{18}O$ values between 3.5 and 6.9‰ (n=11). In the Antsiranana province, the BGY sapphires from Nosy Be Island have a mean $\delta^{18}O$ of 4.5± 0.4‰ (n=2) similar to that of Ambondromifehy, i.e., 4.5± 0.6‰ (n=9). In the Antananarivo province, the BGY sapphires from the Ankaratra region have $\delta^{18}O$ between 4.5 and 6.9‰ (n=3) whereas ruby of the Antsabotraka yielded a $\delta^{18}O$ of 3.5‰.

The ruby from the placers of Vatomandry and Andilamena has $\delta^{18}O$ of 5.9‰ and between 0.5 and 4.0‰ (n=4), respectively. The pinkish to brownish sapphire from Miarinarivo has an oxygen isotopic signature of 5.9‰.

The placers of the Ilakaka area are characterized by a huge variety of colored sapphires and rubies, with $\delta^{18}O$ values between −0.1 and 16.5‰ (Table 2). Ruby and pink to amethyst sapphire have $\delta^{18}O$ in the same range, between 2.6 and 3.8‰ ($\delta^{18}O$=3.4±0.5‰, n=4). Lemon, greenish, and deep-blue sapphires have $\delta^{18}O$ of 6.8, 1.55, and 3.8‰, respectively. Colorless, grayish to transparent sapphires are defined by three sets of $\delta^{18}O$ values (Fig. 4): (1) The first group, in the range of −0.3 to 0.4‰ (n=3), corresponds to the lowest $\delta^{18}O$ values yet reported for natural corundum; (2) The second has a $\delta^{18}O$ of 3.1‰; (3) Sapphire from the third group, between 14.3 and 16.5‰

Table 2 Oxygen isotopic composition of ruby and sapphire from Madagascar: new analyses on corundum originating from Laos, China, and Australia are reported to complete the worldwide oxygen isotopic data base reported by Giuliani et al. (2005)

Country	District or mine	Sample	Nature	Color	Type of deposit	$\delta^{18}O$	
						‰, V-SMOW	References
Primary Deposits							
Madagascar	Andranondambo	SNMAD-1	Sapphire	Colorless to blue	Skarn-stage 3 in marble late K-feldspar veins	14.0	Giuliani et al. (2005)
		SNMAD-2	Sapphire	Deep blue	Skarn-stage 1 marble	10.7±0.1 (n=2)	Giuliani et al. (2005)
		Ambo-1	Sapphire	Colorless to deep blue	Skarn-stage 3 in marble late K-feldspar veins	14.8	This work
		Andro-7 Bazar Be	Sapphire	Deep blue	Skarn-stage 3 in marble late K-feldspar veins	14.7	This work
		Andro-3 Andranoboaka	Sapphire	Deep blue	Skarn-stage 3 in marble late K-feldspar veins	15.6	This work
		Andro-2	Sapphire	Pink	Skarn-stage 1 in marble	10.2	This work
		Andro-6	Sapphire	Blue	Skarn-stage 1 in marble	10.6	This work
		Andro-6h	Hibonite	Black	Skarn-stage 2 in marble	10.9	This work
		Andakato 1	Sapphire	Blue	Skarn-stage 1 in scapolite	10.1	This work
		Andakato 2	Sapphire	Pink	Skarn-stage 1 in pyroxenite	3.9	This work
	Beforona	VG-128	Sapphire	Pinkish	Syenite	8.1	This work
	Vohidava	Vohi-4	Sapphire	Light blue to green	Sakenite in pyroxenite	5.8	This work
	Sakeny	87p	Sapphire	Light brown	Sakenite	4.9	This work
	Ejeda	EJE	Ruby	Red	Amphibolite	5.9	Giuliani et al. (2005)
		199p	Ruby	Red	Sakenite in amphibolite	5.0	This work
	Gogogogo	GO-1	Ruby	Deep pink	Amphibolite	3.8	Giuliani et al. (2005)
	Vohitany	VO-1	Ruby	Deep pink	Amphibolite	6.1	This work
		Vohi-VG	Ruby	Red	Amphibolite	5.4	This work
	Ambinda	Am-1	Sapphire	Grey	Pyroxenite	4.7	This work
	Sahambano	NOI-2	Sapphire	Blue	Biotitised gneiss	5.6	This work
		NOII-1	Ruby	Pink	Biotitite	5.8	This work
		NOII-2	Ruby	Purplish (fuschia)	Biotitite	5.9 (n=2)	This work
		NOII-4	Sapphire	Orange	Biotitite	5.7	This work
		NOII-6	Sapphire	Bluish	Biotitite	6.5	This work
	Ambinda Sud (Sahambano)	Amb-5	Sapphire	Pinkish	Biotitised-sapphirine-bearing gneiss	7.6	This work
	Zazafotsy	Za-1	Sapphire	Blue	Biotitised gneiss	8.8	This work
		Za-2	Sapphire	Pink	Biotitite	9.0	This work
	Iankaroka	131-2a	Sapphire	Light orange	Cordieritite	1.7	This work
		131-2b	Ruby	Deep pink to light red	Cordieritite	2.4±0.3 (n=2)	This work
	Ambatomena	Ambat-1	Ruby	Pink	Biotitised cordieritite	2.9	This work
		Ambat-2	Ruby	Pink	Pegmatite in charnockite	2.9	This work
	Soamiakatra	RNANTA	Ruby	Reddish-purplish	Pyroxenitic enclaves in basalt	4.7	Giuliani et al. (2005)
		SOM-1	Ruby	Deep red	Pyroxenitic enclaves in basalt	1.25	This work
Secondary deposits							
Madagascar	Ilakaka	ILA-1	Sapphire	Colorless	Placer in sandstone	14.3±0.1 (*n*=2)	Giuliani et al. (2005)
		ILA-2	Sapphire	Colorless to pink	Placer in sandstone	3.5	Giuliani et al. (2005)
		ILA-3	Sapphire	Amethyst	Placer in sandstone	3.6	Giuliani et al. (2005)
		ILA-4	Sapphire	Deep pink	Placer in sandstone	2.6±0.3	Giuliani et al. (2005)

Table 2 (continued)

Country	District or mine	Sample	Nature	Color	Type of deposit	$\delta^{18}O$	
						‰, V-SMOW	References
						(n=2)	
		ILA-5	Sapphire	Colorless	Placer in sandstone	16.5	Giuliani et al. (2005)
		ILA-6	Sapphire	Colorless	Placer in sandstone	3.1	Giuliani et al. (2005)
		ILA-7	Sapphire	Light blue	Placer in sandstone	3.8	This work
		ILA-8	Sapphire	Colorless	Placer in sandstone	−0.3	This work
		ILA-9	Sapphire	Transparent	Placer in sandstone	−0.1	This work
		ILA-10	Sapphire	Grayish	Placer in sandstone	0.4±0.3 (n=2)	This work
		ILA-11	Sapphire	Lemon	Placer in sandstone	6.8±0.9 (n=2)	This work
		ILA-12	Ruby	Deep red	Placer in sandstone	3.8	This work
		ILA-13	Sapphire	Greenish	Placer in sandstone	1.55±0.3 (n=2)	This work
	Andilamena	73-30.63A	Ruby	Red	Placer in basalt (?)	3.8±0.1 (n=2)	This work
		AND-1	Ruby	Red	Placer in basalt (?)	2.2±0.1 (n=2)	This work
		AND-2	Ruby	Red	Placer in basalt (?)	0.5	This work
		AND-3	Ruby	Red	Placer in basalt (?)	3.9±0.05 (n=2)	This work
	Ambondromifehy	ANT-1	Sapphire	Yellowish light green	Placer in karst formed in limestone and sandstone	5.1	Giuliani et al. (2005)
		ANT-2	Sapphire	Light blue	Placer in karst formed in limestone and sandstone	5.9	Giuliani et al. (2005)
		ANT-3	Sapphire	Yellow	Placer in karst formed in limestone and sandstone	3.9	This work
		ANT-4	Sapphire	Opaline blue	Placer in karst formed in limestone and sandstone	3.9	This work
		Ambo 12-3	Sapphire	Light blue to lilac	Placer in karst formed in limestone and sandstone	3.75	This work
		ANT-5	Sapphire	Deep blue to black	Placer in karst formed in limestone and sandstone	4.5±0.15 (n=2)	This work
		322-3gy	Sapphire	Yellow to green	Placer in karst formed in limestone and sandstone	4.6	This work
		322-3b	Sapphire	Blue	Placer in karst formed in limestone and sandstone	4.3	This work
		319-2z	Sapphire	Yellow	Placer in karst formed in limestone and sandstone	4.4	This work
	Antanifotsy	SNANTAN1	Sapphire	Bluish	Placer in basalt	6.5	Giuliani et al. (2005)
		SNANTAN2	Sapphire	Greenish to blue	Placer in basalt	6.9	Giuliani et al. (2005)
	Antsabotraka	ANTSA-1	Ruby	Red	Placer in basalt	3.5	This work
	Ambatomainty	AMBATO-1	Sapphire	Light blue to blue	Placer in basalt	4.5±0.1 (n=2)	This work
	Nosy Be	NO-1	Sapphire	Blue	Placer in basalt, sandstone, granite	4.8	This work
		NO-2	Sapphire	Green	Placer in basalt, sandstone, granite	4.2	
	Vatomandry	VAT-1	Sapphire	Red	Placer in basalt (?)	5.9	Giuliani et al. (2005)
	Miarinarivo	MIA-1	Ruby	Pink	Placer in syenite (?)	5.6	This work
Other deposits worldwide							
China	Wenchang, Hainan	W1	Sapphire	Deep blue	Placer in basalt	3.0	This work

Table 2 (continued)

Country	District or mine	Sample	Nature	Color	Type of deposit	$\delta^{18}O$	
						‰, V-SMOW	References
	Changle, Shandong	CHA1	Sapphire	Blue	Placer in basalt	5.65	This work
Laos	Houai-Sai	HO-1	Sapphire	Light green to light yellow	Placer in basalt	5.6	This work
		HO-2	Sapphire	Deep blue	Placer in basalt	4.9	This work
		HO-3	Sapphire	Light green to colorless	Placer in basalt	5.15	This work
Australia	Harts Range	HR1	Ruby	Red	Gneiss	4.35	This work

Data are reported in the conventional delta notation relative to V-SMOW.
n Number of analyses.

($n=2$), has the highest $\delta^{18}O$ values found so far for Malagasy sapphire.

Discussion

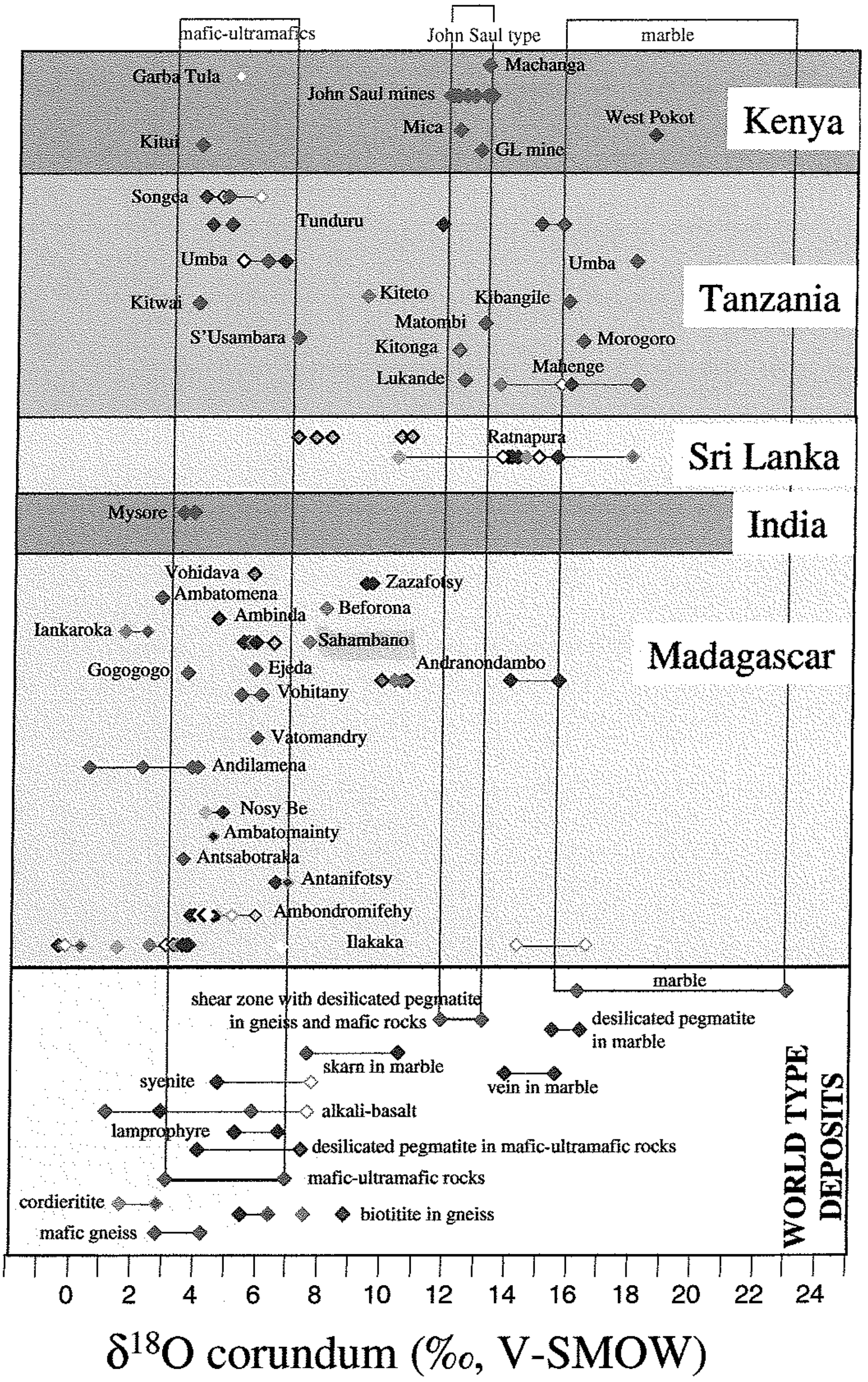
Oxygen isotope systematics of corundum from Madagascar permits us to expand the previous oxygen isotope range defined for the different types of worldwide deposits (Giuliani et al. 2005) and to characterize new types of deposits such as corundum-bearing cordieritites and biotite schists in gneisses. The restricted isotopic range is illustrated by the Malagasy ruby hosted in mafic rocks ($3.8<\delta^{18}O<6.1$‰) falling within the worldwide range defined by Giuliani et al. (2005), i.e., $3.2<\delta^{18}O<6.8$‰. At the scale of a corundum deposit, the variability registered for $\delta^{18}O$ of corundum is low, as shown for the colored sapphires from Sahambano ($\delta^{18}O=5.9\pm0.3$‰, $n=5$). As previously shown by Fallick et al. (1994) and Giuliani et al. (1998) for emerald, and Giuliani et al. (2005) for corundum, the $\delta^{18}O$ of the fluid in equilibrium with corundum was buffered by the local host-rock oxygen isotope composition during metamorphism and fluid rock interaction.

For primary deposits, there are contrasting $\delta^{18}O$ values for magmatic and metamorphic deposits:

(1) Magmatic-hosted corundum from the syenite of Beforona has a $\delta^{18}O$ of 8.1‰ compatible with the worldwide range defined for syenites ($4.8<\delta^{18}O<7.8$‰). Ruby hosted in clinopyroxenite xenoliths in the Soamiakatra alkali basalt has low $\delta^{18}O$ values between 1.3 and 4.7‰, close to the isotopic range defined for ruby in mafic and ultramafic rocks ($3.2<\delta^{18}O<6.8$‰). The corundum–garnet–clinopyroxene assemblage in the clinopyroxenite gave a temperature of formation of ruby around 1,100°C and a pressure of about 20 kbar (Rakotosamizanany et al. 2005). Ruby formed in mafic and ultramafic rocks at the base of the lower crust, and was later intruded and transported up to the surface by the alkali basalts. It was previously suggested that these xenocrysts originally formed in mafic metamorphic rocks under mantle conditions (Sutthirat et al. 2001; Garnier et al. 2005). The oxygen isotope compositions of rubies, including those of Soamiakatra, support this hypothesis (Giuliani et al. 2005; Yui et al. 2006).

(2) Metamorphic-hosted corundum has a wide range of $\delta^{18}O$ values related to the nature of the host rocks. Sapphires from the Andranondambo skarn-type deposit yielded three sets of $\delta^{18}O$ values (Fig. 4): (1) 3.9‰ for a pink sapphire hosted in a pyroxenite from stage 1 of skarn metasomatism. (2) $10.1<\delta^{18}O<10.7$‰ for blue and pink sapphires hosted by scapolitite and marble from stage 1 of skarn metasomatism. The important isotopic variation for sapphires of stage 1 is evidently due to the variability in chemistry of the premetamorphic host rock. During stage 2, hibonite crystallized at the expense of corundum and its $\delta^{18}O$ of 10.6‰ falls within the isotopic range defined for corundum of stage 1. It confirms that in a closed system and at high temperature ($T\sim800$°C), the oxygen isotopic composition of a protolith or a mineral phase controls the final $\delta^{18}O$ of corundum. (3) $14.0<\delta^{18}O<15.6$ (mean $\delta^{18}O=14.8\pm0.6$‰, $n=4$) for blue gem sapphires hosted in K-feldspar veins in marble from stage 3 and formed under retrograde granulite metamorphism. The isotopic range is similar to the range defined worldwide for sapphire hosted in desilicated pegmatites within marbles ($15.5<\delta^{18}O<16.4$‰), where the oxygen isotopic composition of sapphire is buffered by the host rock. The important isotopic variation for sapphires between stages 1 and 3 might be related to the large isotopic variation found in $\delta^{18}O$ of marbles between 6.5 and 19‰ (Boulvais et al. 1998). The lowest $\delta^{18}O$

Fig. 4 Oxygen isotope values of corundum from the different types of primary and secondary deposits of Madagascar, Kenya, Tanzania, Sri Lanka, and India (data from Giuliani et al. 2005; this work). The data reported in the conventional delta notation relative to V-SMOW are compared with the oxygen isotopic ranges defined for corundum types deposit worldwide from the data base published by Giuliani et al. (2005; this work; see Table 2). *Color in diamonds* represents color for ruby (in *red* and *pink*) and colored sapphire (others). White diamonds represent colorless sapphires



values are found in dolomitic marbles whereas calcic ones have higher $\delta^{18}O$ between 13.5 and 19‰.

The sapphire from the sakenites of Vohidava, Sakeny, and Ejeda has a very restricted isotopic range ($4.9<\delta^{18}O<$

5.8‰; Table 2), which reflects the buffering of the isotopic composition of corundum by mafic protoliths (pyroxenite and amphibolite).

Two new types of metamorphic gem corundum deposits are described for the first time in Madagascar: sapphire in biotitites developed in gneisses (Sahambano and Zazafotsy deposits), and ruby and polychrome sapphire in cordieritites intercalated within charnockites (Ambatomena and Iankaroka deposits). Both types of deposit are within shear zones, and corundum records fluid circulation through centimeter- to decimeter-wide channels. Gem corundum in cordieritites has the lowest isotopic range found worldwide for primary deposits ($1.7<\delta^{18}O<2.9‰$). Such ruby and sapphire associated with gem cordierite and sapphire hosted by biotite schists are also described in the Karur–Kangayam gemstone areas belt in the Madurai granulitic block from southern India (Santosh and Collins 2003). Unfortunately, no oxygen isotope data are available for comparison with those of Madagascar.

For placer deposits, the $\delta^{18}O$ values elucidate the geological origin of the most important gem placer deposits of Madagascar and offer clues for the exploration of primary corundum-hosting rocks (Fig. 4).

The majority of gem corundum found today on the world market comes from placers derived from basaltic environments in southern Asia, Australia, and Madagascar. Australia used to be the world's largest seller of dark blue sapphires, but the Ambondromifehy deposit in the Antsiranana province now is one of the most productive in the world (Schwarz et al. 2000). The $\delta^{18}O$ values for 75 BGY sapphires originating from 11 countries show a $\delta^{18}O$ range between 3.0 and 7.7‰ (Giuliani et al. 2005; this work; Table 2). $\delta^{18}O$ values for Malagasy BGY sapphires span 3.8 to 6.9‰, within the worldwide range (Fig. 4). Sapphire from Nosy Be and Ambondromifehy have similar isotopic signatures with a mean $\delta^{18}O$ value of 4.5±0.5‰ ($n=11$). Sapphires from the Ankaratra district have slightly higher $\delta^{18}O$ between 4.5 and 6.9‰ (mean $\delta^{18}O=5.9\pm1.3‰$, $n=3$). Ruby in the Ankaratra district has $\delta^{18}O$ of 1.3 and 4.7‰ for the Soamiakatra and Antsabotraka mines, respectively. This range is lower than the worldwide range previously defined for ruby recovered in alkali basalts ($4.7<\delta^{18}O<7.0‰$; Giuliani et al. 2005).

The BGY sapphires are xenocrysts, which were included in the basalts during their ascent from the mantle to the surface. Different models were proposed for their formation (Irving 1986; Sutthirat et al. 2001; Sutherland and Coenraads 1996; Garnier et al. 2005; Yui et al. 2006): (1) crystallization of corundum from fractionated syenitic melts, (2) crystallization from a mafic melt, and (3) metamorphic crystallization in the lower and middle crust. The $\delta^{18}O$ range of "BGY" Malagasy sapphire ($3.8<\delta^{18}O<6.9‰$) overlaps the range for sapphire hosted in syenites ($4.8<\delta^{18}O<7.8‰$), desilicated pegmatites in mafic rocks ($4.2<\delta^{18}O<6.7‰$), and biotitite in granulitic gneisses ($5.6<\delta^{18}O<9.0‰$). The mineral inclusions identified in Ambondromifehy sapphire (feldspar, zircon, baddeleyite, pyroxene, columbite, hercynite, uraninite, and U-pyrochlore) are typical of magmatic sapphire (Schwarz et al. 2000) and environments rich in incompatible elements and volatiles found in alkali magmas (Upton et al. 1999). In the desilicated pegmatites in mafic rocks from Umba and Kalalani in Tanzania, solid inclusions in sapphire are limited to hematite, rutile, and zircon (Seifert and Hyrsl 1999) whereas sapphire in gneiss-hosted biotitites contain barite, cordierite, zircon, sillimanite, K-feldspar, phlogopite, monazite, and plagioclase (Offant 2005). Given the consistency of the oxygen isotope data and the association with geochemically extreme U/Nb-bearing minerals, the evidence renders unlikely a metamorphic origin for the sapphire. It would rather suggest a crystallization in evolved melts resulting from the fractionation of alkali basalts contaminated by lower crust.

Vatomandry and Andilamena are the recent Malagasy sources of the better quality ruby found on the international market. The quality and color of the Vatomandry ruby show pronounced variation, some being comparable to the Mogok ruby hosted in marbles (Schwarz and Schmetzer 2001). The oxygen isotopic composition of ruby from Andilamena ($0.5<\delta^{18}O<4.0‰$) and Vatomandry (one datum at 5.9‰) suggests three possible origins: (1) mafic rocks ($3.2<\delta^{18}O<6.8‰$), (2) desilicated pegmatites in mafic rocks ($4.2<\delta^{18}O<6.7‰$), and (3) cordieritites ($1.7<\delta^{18}O<2.9‰$). The overlap of the isotopic composition of different lithologies and our insufficient geological knowledge of the productive zone do not permit precise identification of the nature of the primary deposits.

The huge Miarinarivo alluvial placer contains only pinkish to brownish corundum. The $\delta^{18}O$ value of 5.6‰ obtained for one pink sapphire indicates a possible mafic-ultramafic source and syenitic or desilicated pegmatites in mafic rocks. The size of the crystals found in the placer (up to 10 cm) likely precludes a "basaltic" source considering that BGY sapphire crystals elsewhere are always less than 5 cm long (Schwarz et al. 2000).

The placers of Ilakaka represent an extreme mingling of ruby and sapphire from different origins (Fig. 4). The microscopic examination of several crystals of ruby and blue sapphire showed that all rubies have the same gemmological features, i.e., growth, twinning, color bands, and solid inclusions, but blue sapphires defined two different mineralogical groups (Dunaigre 2005). Ruby as well as pink to violet and blue sapphire are within the same $\delta^{18}O$ range, between 2.6 and 3.8‰ ($n=5$). For ruby, the isotopic range corresponds to that defined for rubies from mafic and ultramafic rocks. The Phanerozoic sedimentary sequences and the Isalo Group cover the northern part of the Vohibory unit where intercalations or complexes of mafic and ultramafic rocks hosting ruby are common in the amphibolitic and aluminous gneisses. The erosion of the Vohibory unit during the Upper Carboniferous to Mid-Jurassic necessarily contributed to the concentration of ruby in the basin. A proximal source for ruby in Ilakaka from the Vohibory region is likely.

Blue sapphire, representing more than 95‰ of the production of gemstones recovered in the Ilakaka placers, has a $\delta^{18}O$ value of 3.8‰, which pinpoints two possible origins: (1) BGY basaltic sapphire ($3.0<\delta^{18}O<7.7‰$) and (2) desilicated pegmatites in mafic and ultramafic rocks ($4.2<\delta^{18}O<7.5‰$). Gemmological observations of the sapphire demonstrated solid inclusions of apatite, monazite, feldspar, zircon, calcite, graphite, phlogopite, quartz, biotite, and rutile (Dunaigre 2005). The noticeable lack of columbite, spinel, and uraninite encourages us to prefer the second hypothesis and to propose as an exploration guide to look for colored sapphires in desilicated pegmatites within the Vohibory unit.

The origin of the protolith of yellow and green sapphire crystals is debatable on the basis of the natural occurrences described worldwide. Green and yellow sapphires are reported in the BGY basaltic sapphires (Sutherland et al. 1998), the Garba Tula syenitic dyke (Simonet et al. 2004), desilicated pegmatites from the Eastern African Neoproterozoic belt (Solesbury 1967; Seifert and Hyrsl 1999), and in amphibolitized gabbro with iolite at Mallapaty in southern India (Santosh and Collins 2003). The low $\delta^{18}O$ value of 1.5‰ obtained for the green sapphire has no isotopic equivalent yet reported worldwide, but this value is close to the lowest values found for polychrome sapphires in cordieritites ($\delta^{18}O=1.7‰$). The $\delta^{18}O$ value of the yellow sapphire ($\delta^{18}O=6.8‰$) is within the range of syenites, BGY magmatic sapphire, and desilicated pegmatite in mafic rocks (Umba Valley), but we cannot exclude the hypothesis of a new type of host rock for sapphire.

Colorless, grayish to transparent sapphires define three sets of $\delta^{18}O$ values, between −0.3 and 0.4‰, at 3.1‰, and between 14.3 and 16.5‰, respectively. What is their mineralogical significance? Colorless sapphire can be found in every type of deposit. Crystals are not always homogenous in color, and transparent, colorless to grayish zones are found in the core and/or the outer parts of the sapphires. Sometimes recrystallization of colorless to cloudy crystals leads to the formation of colored sapphire with patchy colorless zones, but the $\delta^{18}O$ values of the different colored zones are the same (Giuliani et al. 2005). After erosion of the primary deposits, fragmentation of the crystals results in the separation of the different colored zones and mingling in gravels.

The $\delta^{18}O$ values between −0.3 and 0.4‰ are outside the range identified for primary natural corundum and, consequently, the origin of these sapphires remains unknown. The $\delta^{18}O$ value of 3.1‰ falls in the range of corundum hosted in mafic-ultramafic rocks or cordieritites. The values between 14.3 and 16.5‰ are the highest measured for Malagasy corundum, but fit with the isotopic range defined for blue to colorless gem sapphires associated with K-feldspar veins in Andranondambo ($14.0<\delta^{18}O<15.6‰$). It overlaps slightly the isotopic range defined for sapphire from desilicated pegmatite in marbles ($15.5<\delta^{18}O<16.4‰$), or ruby and colorless to pinkish sapphire hosted in marbles ($16.3<\delta^{18}O<23.0‰$). These isotopic results open a debate upon the presence of such marble-type deposits in the metamorphic zones of the southern part of Madagascar, similar to those known in Tanzania and Southeast Asia (Giuliani et al. 2005).

Low values of $\delta^{18}O$ are reported in corundum from the Ilakaka placers ($-0.3<\delta^{18}O<1.55‰$) and the Iankaroka and Ambatomena cordieritites ($1.7<\delta^{18}O<2.9‰$). Isotopically rather light values are also found in sapphires from the feldspathic gneisses (metashale protolith) of Sahambano and Zazafotsy ($5.6<\delta^{18}O<9‰$), but no anomalous $\delta^{18}O$ values were observed for ruby in mafic and ultramafic rocks ($3.8<\delta^{18}O<6.1‰$). Other worldwide corundum occurrences from granulite metamorphic conditions also show low $\delta^{18}O$, including ruby in mafic granulites from Mysore (India), in gneisses from Karelia (Russia) and Harts Range (Australia), and sapphire in calcic skarn formed in gneiss from Sri Lanka (Table 2; Fig. 5). Unusually low values of $\delta^{18}O$ were reported in granulitic rocks and metasediments in the Sahara (Fourcade and Javoy 1973), Precambrian granulites from the Strangways Range and the Quairading Region in Australia (Wilson 1978; Wilson and Baksi 1983), and also for Adirondack anorthosites and related wollastonite skarn (Valley and O'Neil 1982, 1984). Wilson and Baksi (1983) proposed a variety of processes of pre-, syn- or postgranulite facies metamorphism: (1) pregranulite reaction between heated seawater and hot basic intrusives, or an initial protolith such as a fossil soil for the sapphirine–spinel–(cordierite) assemblages; (2) syn-granulite depletion in ^{18}O related to dehydration during granulite metamorphism and removal of the resultant products of partial melting with a depletion in ^{18}O by up to 2 or 3‰ for the restite; (3) postgranulite facies metamorphism with recrystallization under the effect of biotite and/or amphibole-metasomatism with depletion in ^{18}O up to 4‰. The last hypothesis initiated a debate on the role of metasomatism in the Strangways Range (Allen 1981), with finally the suggestion that a combination of two or three of these processes might have occurred (Wilson 1981; Wilson and Baksi 1983).

Discussion here will concentrate on the oxygen isotopic ranges defined for corundum in Madagascar and similar isotopic ranges found for host rocks in the Strangways Range and in other localities in the world (Fig. 5): (1) $\delta^{18}O$ values of Malagasy corundum in cordieritites overlap the $\delta^{18}O$ range for magnesio-alumi-

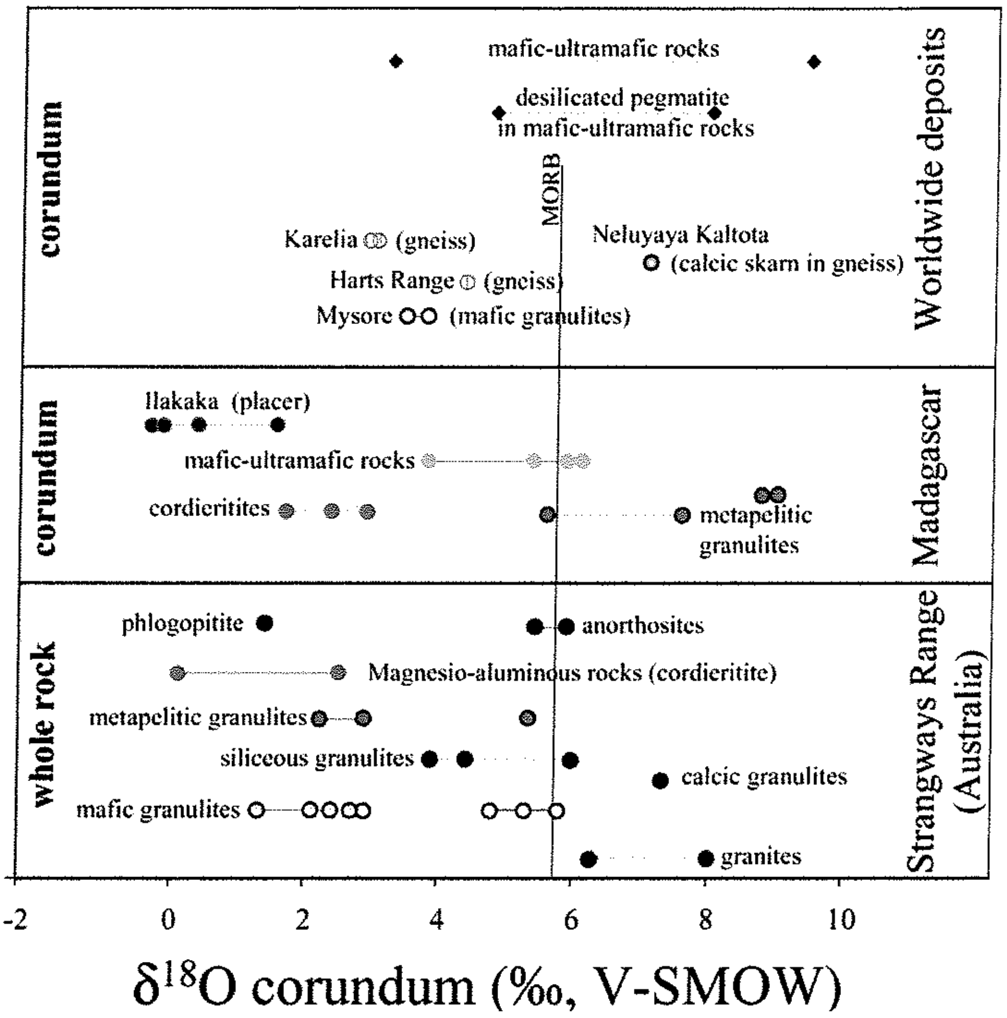


Fig. 5 $\delta^{18}O$ values of some low $^{18}O/^{16}O$ corundum deposits from Madagascar and worldwide, compared with various groups of granulites from the Strangways Range from Australia (Wilson and Baksi 1983; Giuliani et al. 2005; this work)

nous rocks (cordieritites) from Australia; (2) $\delta^{18}O$ of ruby in mafic granulites from Mysore, as well as those in granulitic gneisses from Karelia and Harts Range, are in the isotopic range defined for such rocks by Wilson and Baksi (1983); (3) corundum in metapelitic granulites in the Strangways Range is highly depleted in ^{18}O ($2.2<\delta^{18}O<5.3$‰, $n=3$) as is, to a lesser extent, the Malagasy corundum in metapelitic granulites from the Sahambano and Zazafotsy deposits ($5.6<\delta^{18}O<9.0$‰). These whole-rock $\delta^{18}O$ values are much lower than those found for Archean clastic sedimentary rocks ($8.0<\delta^{18}O<13$‰; Wilson and Baksi 1983) and many younger metamorphosed pelitic sediments ($15<\delta^{18}O<20$‰); (4) $\delta^{18}O$ values of ruby in mafic and ultramafic rocks fall within the normal $\delta^{18}O$ range determined for such host rocks (Giuliani et al. 2005), even those deposits in southern Madagascar that are linked to shear zones and alkaline metasomatism; and (5) $\delta^{18}O$ values in sapphire from the Ilakaka placers are low in the region.

The buffering of oxygen isotopic composition of corundum by its host rock (Giuliani et al. 2005) and the absence of low $\delta^{18}O$ corundum in mafic rocks strengthen the hypothesis of the development of anomalous $\delta^{18}O$ values in pregranulite rocks such as cordieritites, mafic and metapelitic granulites. All these protoliths belong to the Androyan series from southern Madagascar and future investigations of whole rocks and mineral separates from the corundum host rocks and alkaline metasomatites will help us understand this ^{18}O depletion.

The metamorphic ruby and sapphire deposits from Madagascar present numerous geological similarities with those of East Africa (Henn and Milisenda 1997; Mercier et al. 1999b), Sri Lanka (Rupasinghe and Dissanayake 1985), and South India (Santosh and Collins 2003). Based on the correlation of gem and graphite fields, Menon and Santosh (1995) and Dissanayake and Chandrajith (1999) proposed the existence of a Neoproterozoic–early Cambrian gemstone province developed under granulitic conditions in East Gondwana. The common presence of ruby and pink sapphire in Kerala and southern Madagascar led Santosh and Collins (2003) to support the view that the Achankovil shear zone in southern India extends into the Ranotsara shear zone in Madagascar (Fig. 6). However, few studies were devoted to the petrology, mineralogy, and geochro-

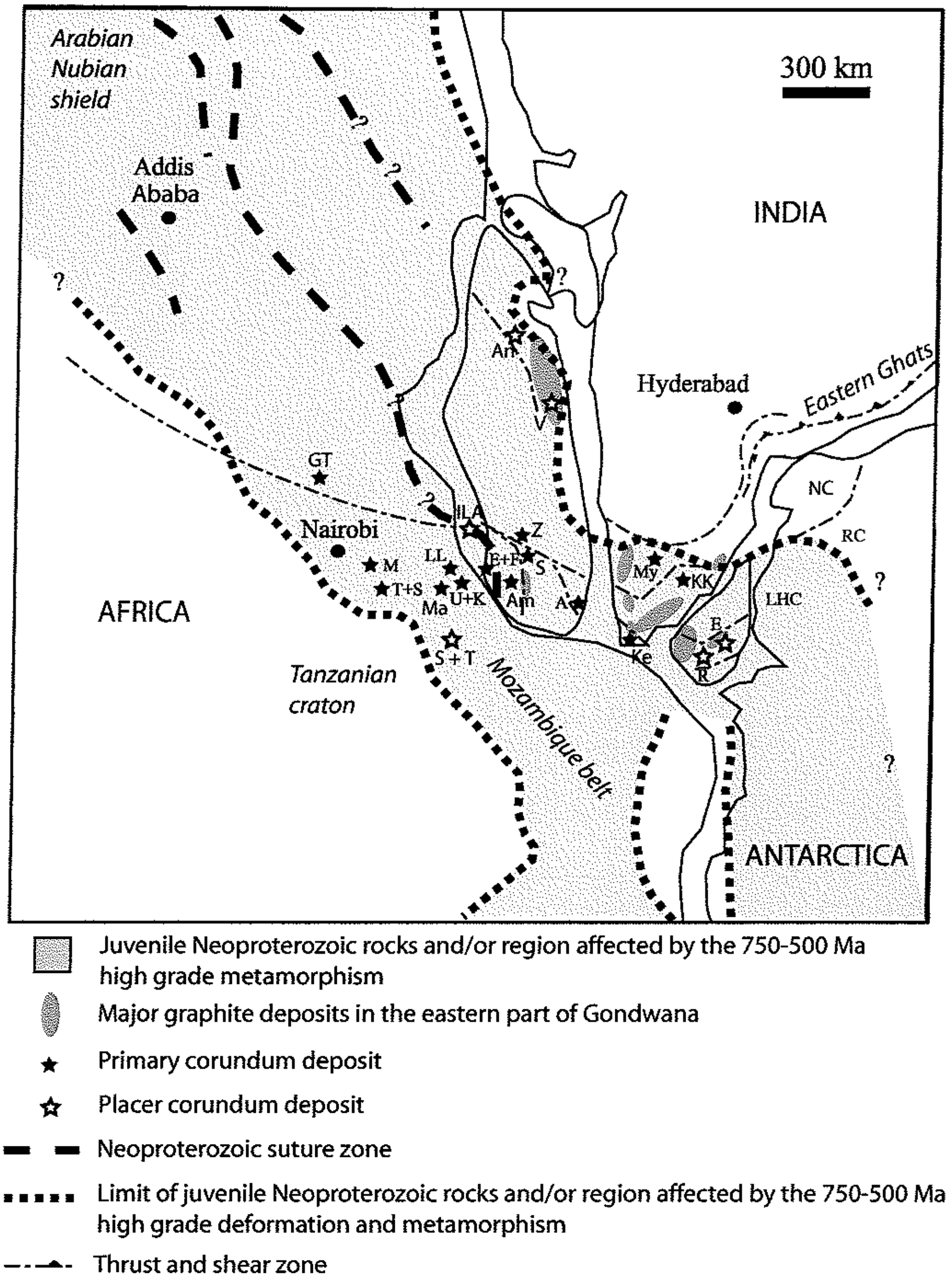


Fig. 6 Juxtaposition of Sri Lanka, India, Madagascar, and Eastern Africa in a tight fit reconstruction of Gondwana with the location of the main gem corundum deposits (modified from Lawler et al. 1998; Collins and Windley 2002; Santosh and Collins 2003). Eastern Africa: *GT* Garba Tula, *M* Mangari, *T+S* Twiga and Si Ndoto, *LL* Longido and Lonsogonoi, *Ma* Mahenge, *U+K* Umba and Kalalani, *S+T* Songea and Tunduru. Madagascar: *ILA* Ilakaka, *Z* Zazafotsy, *S* Sahambano, *E+F* Ejeda and Fotadrevo, *Am* Ambatomena, *A* Andranondambo. South India: *My* Mysore, *KK* Karur–Kangayam corundum belt, *Ke* southern Kerala. Sri Lanka: *R* Ratnapura, *E* Elahera. Antarctica are reported only the metamorphic complexes with *NC* Napier complex, *RC* Rayner complex, *LHC* Lützow–Holm complex

nology of the extensive mineralization associated with the formation of Gondwana.

Figure 4 compares the oxygen isotope data of gem corundum from Madagascar with those from Kenya, Tanzania, Sri Lanka, and India on the basis of data published by Giuliani et al. (2005) and this work. For primary deposits, ruby and pink sapphire group in three sets of $\delta^{18}O$ values: (1) The first group corresponds to ruby hosted in mafic-ultramafic rocks and/or associated desilicated pegmatites found in Kenya, Tanzania, India and Madagascar; (2) The second group is defined for ruby in Kenya and Tanzania hosted in shear zones cross-cutting ultramafic lenses and pegmatites within gneisses (John Saul type deposit; Mercier et al. 1999b); and (3) The third $\delta^{18}O$ group corresponds to ruby hosted in marbles from Kenya and Tanzania. For sapphire, in Madagascar a variety of mineralization types occur, but a key feature for Sri Lanka is the presence of skarn-type deposits, and in Tanzania and Kenya sapphire is related to mafic-ultramafic rocks and syenite, respectively.

For secondary deposits, the $\delta^{18}O$ values obtained for gem corundum from the giant placers of Tanzania (Songea and Tunduru deposits), Madagascar (Ilakaka), and Sri

Lanka (Ratnapura) show mingling of sapphires from several sources. Ruby and pink sapphire from Tanzania and Madagascar indicate principally a mafic and ultramafic source, while pink sapphire in Sri Lanka is probably related to skarn. In Tunduru, two $\delta^{18}O$ values of 14.9 and 15.6‰ for a ruby and a purple sapphire point to a probable origin of pegmatite hosted in marble ($15.5<\delta^{18}O<16.4$‰). The $\delta^{18}O$ values of sapphires from Songea, Tunduru, and Ratnapura have a wide spread in the range ($4.3<\delta^{18}O<17.1$‰) as already seen for Ilakaka (Fig. 4). The $\delta^{18}O$ values for sapphires in the Ratnapura placers fall in a range that indicates a skarn origin in a marble source ($10.3<\delta^{18}O<17.1$‰), while for Songea the isotopic range ($4.3<\delta^{18}O<5.9$‰) suggests an origin in a mafic rock and/or association with desilicated pegmatites. The $\delta^{18}O$ values found for Sri Lankan sapphires, especially blue ones ($13.9<\delta^{18}O<15.4$‰), are within the range defined for those from Andranondambo ($14.0<\delta^{18}O<15.6$‰).

Conclusions

Despite the relative paucity of oxygen isotope data for Indian corundum, the $\delta^{18}O$ systematics of gem corundum in Madagascar and other countries from Gondwana have partly unraveled their geological origin.

The distribution of $\delta^{18}O$ values shows that ruby from Madagascar, as in Eastern Africa, is closely related to mafic-ultramafic protoliths, but great differences exist between these two granulitic blocks of the Gondwana supercontinent:

(1) In Kenya and Tanzania, the ruby deposits called "John Saul mine type" are characterized by shear zones developed in mafic and ultramafic lenses and pegmatites contained in gneiss; these are unknown in Madagascar, Sri Lanka, and India. The Morogoro and Mahenge ruby deposits hosted in Tanzanian marbles (Hänni and Schmetzer 1991), similar to those found in Southeast Asia, have also not been described from Madagascar, Sri Lanka, or India. For this type of deposit, ruby mineralization is restricted to peculiar impure marble horizons, which contained evaporitic layers (Garnier 2003). The presence of evaporites is the key to understanding the formation of such ruby, and future study of the chemistry of marble-hosted metasedimentary series from southern Madagascar will be of interest in the search for new ruby deposits.

(2) In Madagascar, as in Sri Lanka, sapphire hosted in skarn is one of the most important primary deposit types. The isotopic composition of the different types of sapphire from Andranondambo is controlled by the initial premetamorphic isotopic composition of marble. The higher $\delta^{18}O$ found for sapphire in the Ratnapura placers may be related to skarns developed in marble (Silva and Siriwardena 1988), as shown for the blue gem sapphires associated with K-feldspar veins from Andranondambo. Also, a new type of ruby and polychrome sapphire described from southern Madagascar, with typically very low $\delta^{18}O$ values ($1.7<\delta^{18}O<2.9$‰), is so far unique worldwide.

(3) Sapphire deposits linked to syenite are limited to the Garba Tula deposit in Kenya and Beforona in Madagascar. The prospection for such deposits in the southeastern coastal part of Madagascar containing syenitic bodies is a new opportunity for gem exploration.

(4) Giant placer deposits from Sri Lanka, Madagascar, and Tanzania have a large variety of colored sapphire and ruby with a large variation in $\delta^{18}O$ due to mingling of corundum of different origin: mafic and ultramafic protoliths for ruby and blue sapphire, and skarn in marbles for blue sapphire are the main sources for these placers.

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