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# Chemical and oxygen isotopic compositions, age and origin of gem corundums in Madagascar alkali basalts

Saholy Rakotosamizanany<sup>a,b,c</sup>, Gaston Giuliani<sup>b,c,\*</sup>, Daniel Ohnenstetter<sup>c</sup>,

Amos Fety Michel Rakotondrazafy<sup>a</sup>, Anthony E. Fallick<sup>d</sup>, Jean-Louis Paquette<sup>e</sup>, Massimo Tiepolo<sup>f</sup>

<sup>a</sup> Université d'Antananarivo, Faculté des Sciences, Département des Sciences de La Terre, Ambohitsaina, BP 906, Antananarivo 101, Madagascar

<sup>b</sup> Université Paul Sabatier, GET/IRD, UMR CNRS-IRD-CNES 5563, 14 avenue Edouard Belin, F-31400 Toulouse, France

<sup>c</sup> Université de Lorraine, CRPG UMR 7358 CNRS-UL, 15 rue Notre-Dame-des-Pauvres, F-54501 Vandœuvre-lès-Nancy cedex, France

<sup>d</sup> Scottish Universities Environmental Research Centre, East Kilbride, Glasgow G75 0QF, Scotland, United Kingdom

<sup>e</sup> Clermont Université, Université Blaise Pascal, Laboratoire Magmas et Volcans, BP 10448, F-63000 Clermont-Ferrand, France

<sup>f</sup>CNR, Istituto di Geoscienze e Georisorse, Unità di Pavia, via Ferrata 1, I-27100 Pavia, Italy

Madagascar is a large producer of gem corundum recovered from continental basaltic fields. The main mining areas are sapphire-bearing palaeoplacer deposits such as Ambondromifehy and Nosy Be in Antsiranana Province, northern Madagascar; Soamiakatra-Mandrosohasina in Antananarivo Province, central Madagascar; and Vatomandry district in Toamasina Province, eastern Madagascar. In Antananarivo Province, Soamiakatra is a primary deposit where ruby is found in metagabbro and pyroxenite xenoliths, brought-up to the upper crust by the Ankaratra volcanics. Petrographic studies indicate two different conditions of ruby formation, at the boundary of the eclogite domain ( $T \sim 1100$  °C,  $P \sim 20$  kb) and granulite facies ( $T \sim 1100 \text{ °C}$ , P < 15 kb). In contrast, most of the sapphires in placer and paleoplacer have two origins: (i) differentiation of alkaline magma in chambers at the lower continental crust-mantle boundary (90% of the sapphires). They are related to syenite and anorthoclasite xenoliths in the basalts. These alkali-basalt hosts are linked with asthenosphere upwelling and E-W and N-S lithosphere thinning during Oligocene-Quaternary times. Zircons associated with the sapphires from Mandrosohasina and Ambatomainty sapphire deposits gave U–Pb ages at 7 Ma. The sapphires have low  $\delta^{18}$ O values of 4.1 ± 0.4% (n = 8), within the range of sapphire in syenitic rocks. Chemical composition and mineral inclusions in sapphires, such as columbite-(Fe), tantalite-(Mn), pyrochlore group, samarksite group, uraninite and anorthoclase confirm their syenitic origin. (ii) metamorphic xenocrysts (10%) brought up by the same magma. Oxygen isotopic compositions of rubies from placer deposits,  $\delta^{18}$ O = 3.1 ± 1.1% (n = 6) are typical of ruby in mafic and ultramafic rocks and 'plumasite' in mafic rocks ( $1.25 < \delta^{18}O < 7.5\%$ , n = 35).

In Toamasina Province, the sapphires of Vatomandry are mainly of metamorphic origin (~85%) and their  $\delta^{18}O = 4.1 \pm 0.4\%$  (*n* = 9) are low and overlaps the range defined for metasomatic sapphires linked to 'plumasites' and biotite schists in shear zones; ~15% of the sapphires are magmatic in origin with a low  $\delta^{18}O$ -isotopic range in the sapphire-bearing syenites field. In Antsiranana Province, there are no rubies and sapphires are either magmatic (~40%) or metamorphic (~60%) in origin. The  $\delta^{18}O = 4.5 \pm 0.5\%$  (*n* = 11) values are similar to the mean  $\delta^{18}O$  of sapphires from other two Provinces. Two U/Pb ages on zircons gave two contrasted ages at respectively 40.6 Ma for Ambondromifehy and 0.7 Ma for Nosy Be sapphire deposits. The characteristics of the corundum, their isotopic compositions as well as their ages demonstrate the existence of two distinct sources of corundum associated with alkali-basalts in Madagascar. On one hand, the rubies associated with metagabbros and garnet-bearing pyroxenites are linked to mafic and ultramafic complexes of eclogite facies at the boundary between lower crust and upper mantle, retrograded to granulite facies during the Pan-African event. On the other hand, the sapphires brought up during the Eocene to Quaternary are interpreted to be either magmatic and coeval with a volcanic event involving differentiated alkaline magma, or metamorphic and extracted from the pre-existing Precambrian basement during the extrusion of the magma.

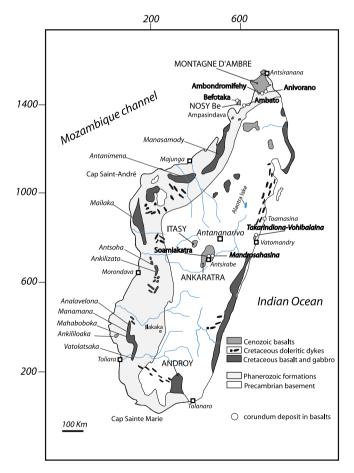
### 1. Introduction

Madagascar is a major gem corundum-exporting country (Yager et al., 2008). In 1997, it was the second largest producer of

<sup>\*</sup> Corresponding author. Address: CRPG/CNRS, 15 rue Notre-Dame-des-Pauvres, BP 20, F-54501 Vandœuvre-lès-Nancy, France. Tel.: +33 3 83 59 42 38; fax: +33 3 83 51 17 98.

E-mail address: giuliani@crpg.cnrs-nancy.fr (G. Giuliani).

sapphires (4248 kg) following the discovery in 1996 of the Ambondromifehy placers, located in the basaltic Province of Antsiranana (Fig. 1; Schwarz et al., 2000). In 1999 after the discovery of giant placer deposits in the Ilakaka and Sakahara region, southern Madagascar (Garnier et al., 2004), the production of sapphires increased from 3810 to 9536 kg in 2000. Since 2004, the Malagasy production decreased to 4700 kg per year (Fig. 2A). The production of ruby wasnot significant before 2000, until the discovery of two deposits in the Province of Toamasina named Andilamena (Rakotondrazafy et al., 2008) and Vatomandry (Schwarz and Schmetzer, 2001). Since 2000, the mean annual production of ruby has remained at around 850 kg per year. In 2005, Madagascar was the third largest ruby-exporting country (Fig. 2B) after Kenya (5100 kg) and Tanzania (2800 kg). The deposits of Ambondromifehy and Vatomandry are placers related to basaltic environments (Giuliani et al., 2007a). These alluvial deposits are characterized by large volumes of gems extracted from detrital sediments, with a low grade of economic sapphires and/or ruby (circa 10-15% of the rough production; Schwarz et al., 2000). These sapphires called 'basaltic' or "BGY" blue-green-yellow sapphires (Sutherland et al., 1998a,b; Schwarz et al., 2000; Graham et al., 2008) are the main type of sapphires exported throughout the world (Fig. 1C and D) with



**Fig. 1.** The volcanic Provinces of Madagascar and localities of the 'basaltic' corundum deposits related to the Cenozoic basalts in the Provinces of Antsiranana, Antananarivo and Toamasina (modified from Rasimanana, 1996). The deposits of gem corundum: north in the Antsiranana Province, the mining districts of Ambondromifehy and Anivorano in the Montagne d'Ambre, Ambato peninsula and Befotaka in Nosy Be island; in central Madagascar in Antananarivo Province, the mines of Soamiakatra, Mandrosohasina–Kianjanakanga (Antsirabe area) in the volcanic Ankaratra massif; and in east Toamasina Province, with the deposits of Vatomandry in the Takarindiona and Vohibalaina basaltic areas. The geographic coordinates are related to the Laborde projection.

the main producing-countries being Australia (5500 kg), China (3000 kg) and Cameroon (1000 kg).

In Madagascar, the main corundum deposits (Fig. 1) form sapphire-bearing palaeoplacers and placers of the Ambondromifehy district in the northern part of the country, Soamiakatra–Mandrosohasina and other deposits from Antsirabe region in the central part, and Vatomandry in the eastern part of the island. This paper aims to review geological and recent metallogenetic studies to classify ruby and sapphires in the different gem Provinces and to discuss the genetic model for their formation. Recent knowledge on these corundum deposits was obtained by the PhD thesis of Rakotosamizanany (2009) and fieldwork developed by the Institute of Research and Development (IRD) with the academics of the University of Antananarivo and the PGRM program from the Ministry of Energy and Mines of Madagascar (BGS-USGS-GLW, 2008).

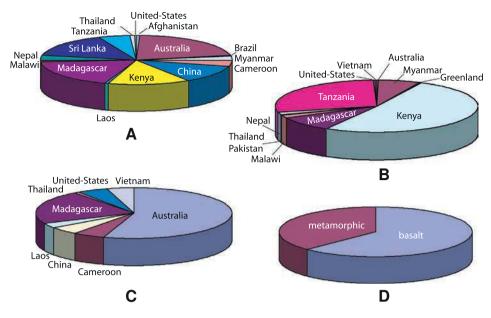
The details of the geology of the deposits, mineralogy and chemistry of the gem-corundum and oxygen isotope data of ruby and sapphires are presented in the Electronic Supplement Material (see ESM text, tables and figures). The analytical procedures used for mineral chemistry, isotopic and radiometric data are compiled in the Appendix.

#### 2. Mesozoic and Cenozoic magmatism in Madagascar

Madagascar experienced several significant magmatic episodes during the break-up of the Gondwana super-continent and continuous continental drift between Africa and Madagascar, India and Sri Lanka, and Antarctica (de Wit, 2003). The first was an Upper Cretaceous episode at the Turonian–Coniacian (87 Ma). The two other episodes are Cenozoic in age, with volcanic flows in the Paleocene and Eocene (65–34 Ma), and in the Upper Miocene (circa 10 Ma), and finally in the Quaternary (Karche, 1973; Melluso et al., 1997; Melluso and Morra, 2000; Mottet, 1981; Tucker et al., 2008; Bardintzeff et al., 2010; Cucciniello et al., 2011).

The Upper Cretaceous magmatic episode produced tholeiitic basaltic flows and doleritic dykes (Fig. 1; Besairie et al., 1957). It formed widespread stratified flows on the sedimentary units in the western and southern coastal regions in the Morondava and Majunga, and Androy areas (Fig. 1). The Androy massif sequence, 1500 m thick, contains tholeiitic basalts associated with alkali-basalts and rhyolites. The Cretaceous flows are also found on the eastern border of Madagascar in the Provinces of Antsiranana, Toamasina and Fianarantsoa (Besairie et al., 1957; Nicollet, 1984).

The Paleogene-Neogene and Quaternary magmatic episodes north of Toliara (Ankililoaka flows), south and north of Antsiranana (Montagne d'Ambre, Nosy Be, Ambato peninsula, Ampasindava), around Antsirabe (Massifs of Itasy and Ankaratra; Besairie, 1971), and close to Vatomandry (Fig. 1), form a sequence of alkali-basalts, ignimbrites, rhyolites, trachytes, phonolites, and basaltic and basanitic tuffs (Karche, 1973). The gem corundum deposits in basaltic areas are only related to these two Cenozoic episodes, in Antsiranana, Antananarivo and Toamasina Provinces (see ESM text). The deposits from Antsiranana Province are located 70 km south of Antsiranana city, in the areas of Anivorano and Ambondromifehy on the southern border of the Montagne d'Ambre (Fig. 1), and in the Ambato peninsula and Nosy Be Island. The sapphires and/or ruby deposits from Antananarivo Province are located within the Ankaratra volcanic massif. Two types of deposits are recognized: (i) a primary ruby deposit associated with a basaltic dyke at Soamiakatra (Antanifotsy region); and (ii) secondary deposits (i.e. alluvial placer and paleoplacer), located to the north and east of Antsirabe city (Mandrosohasina, Kianjanakanga, Ambatomainty, Antsabotraka). The rubies and sapphires of Toamasina



**Fig. 2.** (A) World sapphire production in 2005 (Yager et al., 2008). (B) World ruby production in 2005. (C) World 'basaltic' sapphire production in 1997. The discovery of the Ambondromifehy sapphires in Antsiranana Province (northern of Madagascar) occurred in 1997 with a significant production of 4248 kilos (kg). Before 1997, the production of Madagascar was 115 kg in 1995 and 160 kg in 1996. From 1998 to 1999, the production was respectively 2547 and 3810 kg. The difference in sapphire production between 1996 and 1999 thus shows the economic importance of such alkali basalt-related deposits. In 2000, the giant economic placer of llakaka (Fig. 1) which is not related to alkalibasalts allowed the Malagasy production to reach the official record of 9536 kg. (D) World sapphire production in 1997 in terms of 'basaltic' vs. metamorphic deposits.

Province are located in the area of Vatomandry. They were officially discovered in 2000 and the alluvial and paleoplacer deposits border the Takarindiona and Vohibalaina basaltic massifs. The mining sites are Amfao, Sahanonoka, and Antsidikana near the commune of Amboditavolo and located on the massif of Vohibalaina, and Antanambao, Mahatsara and Ankazombanga on the southern and eastern borders of the Takarindionamassif.

These large Late Cenozoic alkali-basalt Provinces are the source of the large quantities of "BGY" (Blue–Green–Yellow) sapphires recovered from consolidated alluvial deposits derived from eroded basalts. The corundums are xenocrysts and/or crystals in xenoliths carried by these basalts. The crystal morphology, internal growth patterns, mineral inclusions and trace element contents in these sapphires are typical of 'basaltic-magmatic' sapphires found worldwide (Schwarz et al. 2000), but the nature of their parent host rocks is still under debate.

# 3. Origin of corundum gem fields in alkali basalts from Madagascar

# 3.1. Chemical classification of sapphire and ruby: petrogenetic significance

Gem corundum contains more than 98 wt% of Al<sub>2</sub>O<sub>3</sub> and additional elements such as Fe, Cr, V, Ti, Mg and Ga. Sutherland and Schwarz (2001) and Sutherland et al. (1998b, 2003) used the Fe<sub>2</sub>O<sub>3</sub>/TiO<sub>2</sub> vs. Cr<sub>2</sub>O<sub>3</sub>/Ga<sub>2</sub>O<sub>3</sub> diagram for deciphering metamorphic vs. magmatic origin corundum. The compositions of different representative coloured gem corundum from Madagascar are shown in Table 1 (see also ESM Tables 5–7 and 9). The 460 chemical analyses obtained on Malagasy rubies and sapphires are reported in Fig. 3: (i) All of the points related to rubies from the central and eastern Provinces plot in the metamorphic field. They overlap data of rubies from Andilamena located in the north of Vatomandry area, hosted in different types of rocks viz. mafic–ultramafics and plumasite (i.e. a desilicated pegmatite formed by fluid metasomatism of both pegmatite and mafic or ultramafic rocks. The plumasite consists of plagioclase or K-feldspar, corundum, biotite and other accessory minerals; Simonet et al., 2008) and placers (Rakotondrazafy et al., 2008). For the Soamiakatra deposit, primary ruby is hosted by metagabbros and pyroxenites (see the petrographic description in ESM text and ESM Figs. 4 and 5). (ii) The majority of sapphires plot in the magmatic domain. The sapphires from Antsiranana Province are not plotted in these diagrams because they do not contain detectable chromium. (iii) The distribution of sapphires from Vatomandry area is heterogeneous: most are located in the magmatic domain yet others are in the metamorphic. The latter corresponds to colourless, light to dark green and light to purplish blue sapphires. They are characterized by the presence of significant  $Cr_2O_3$  contents (150 ppm <  $Cr_2O_3$  < 2100 ppm).

The chemical classification diagram used for the characterization of gem-corundum (Giuliani et al., 2010; Uher et al., 2012) represents the fields of the different types of primary deposits (Giuliani et al., 2007b). It used the database of 2000 microprobe analyses obtained on corundum hosted by different type of rocks worldwide and analyzed with the same analytical conditions (see ESM Appendix I). The diagram  $FeO - Cr_2O_3 - MgO - V_2O_3$  vs.  $FeO + TiO_2 + Ga_2O_3$  (Fig.4 A) uses: (1) FeO as a significant trace element of corundum. The FeO content discriminates between ironpoor rubies in marbles and iron-rich rubies in mafic-ultramafic rocks (Pham et al., 2004); (2) the second discriminator between ruby and sapphire is the addition to (parameter on the Y-axis) or subtraction from (parameter on the X-axis) FeO of trace elements associated preferentially with sapphire (TiO<sub>2</sub> and Ga<sub>2</sub>O<sub>3</sub>) or ruby (Cr<sub>2</sub>O<sub>3</sub>, V<sub>2</sub>O<sub>3</sub>, MgO). The different gem corundum deposits are: for rubies, (R1) marble; (R2) John Saul mine type; (R3) mafic and ultramafic rocks; (R4) in metasomatites; for sapphires, (S1) syenitic rocks; (S2) metasomatites; (S3) alkali-basalt and lamprophyre.Finally, the chemical diagram permits a clear separation with limited overlap of the main types of deposit found worldwide (Fig. 4A). The chemical diagram provides sufficient discrimination to underpin the different types of corundum deposits. The LA-ICP-MS technique used recently is more precise than EPMA (Abduriyim and Kitawaki, 2006; Peucat et al., 2007; Sutherland and Abduriyim, 2009). This particularly applies for Mg and Ga with

#### Table 1

Representative chemical analyses (wt%) of sapphires and rubies from the Ambondromifehy and Nosy Be mines in the Antsiranana Province, the Kianjanakanga, Ambatomainty, Soamiakatra and Antsabotraka deposits in the Antananarivo Province, and the Vatomandry deposits in the Toamasina Province.

Sample	Colour	$Al_2O_3$ (wt%)	MgO (wt%)	TiO <sub>2</sub> (wt%)	V <sub>2</sub> O <sub>3</sub> (wt%)	Cr <sub>2</sub> O <sub>3</sub> (wt%)	Ga <sub>2</sub> O <sub>3</sub> (wt%)	FeO (wt%)	Total (wt%)
Ambondromifehy									
ambo3-3	Colourless	99.17	0.000	0.028	0.001	0.000	0.052	0.640	99.89
ambo5	Milky	99.21	0.000	0.007	0.002	0.000	0.044	1.169	100.44
ambo6-2	Greenish blue	99.43	0.004	0.059	0.001	0.019	0.050	0.506	100.07
ambo6-2a	Light blue	99.20	0.000	0.129	0.003	0.015	0.049	0.549	99.94
ambo4	Dark blue	99.23	0.000	0.126	0.001	0.000	0.076	0.643	100.07
ambo2	Yellowish	98.42	0.000	0.018	0.000	0.000	0.075	1.121	99.63
ambo6-2b	Pinkish	99.20	0.000	0.129	0.003	0.015	0.049	0.549	99.94
Nosy Be									
NBS9-2-pt1	Colourless	99.30	0.003	0.075	0.006	0.002	0.020	0.799	100.20
NBS10-1-pt3	Dark blue	99.14	0.000	0.091	0.000	0.000	0.044	0.462	99.74
NBS11-3pt6	Greenish blue	98.45	0.008	0.107	0.000	0.094	0.062	0.700	99.42
Kianjanakanga									
K1-1	Blue	98.58	0.006	0.178	0.005	0.000	0.022	0.982	99.78
Ambatomainty									
AMBAT1-3	Dark brown	99.36	0.000	0.010	0.002	0.000	0.010	0.021	100.71
AMBAT1-2a	Green	99.18	0.002	0.168	0.005	0.000	0.010	1.069	100.44
AMBAT1-2b	Blue	98.80	0.005	0.311	0.009	0.001	0.016	1.122	100.26
Soamiakatra									
S13-5	Light purplish	97.55	n.d.	0.022	0.000	0.634	0.011	0.503	98.72
S222-2	Dark purplish to red	97.70	n.d.	0.043	0.063	0.933	0.007	0.526	99.27
s540-64	Brownish to red	99.50	n.d.	0.011	0.009	0.070	0.012	0.360	99.97
S64-3	Light pink	97.75	n.d.	0.012	0.000	0.243	0.009	0.255	98.27
s411-41	Dark pink	98.87	n.d.	0.013	0.011	0.647	0.017	0.451	100.01
Antsabotraka									
ANTS4-2-4	Brownish to dark red	99.48	0.015	0.016	0.010	0.130	0.015	0.388	100.06
ANTS4-3-3	Brownish to red	99.46	0.013	0.024	0.010	0.177	0.013	0.473	100.17
Vatomandry									
V5-1	Purplish-blue	99.19	0.011	0.012	0.008	0.119	0.010	0.381	99.73
V1-2a	Colourless	99.28	0.009	0.045	0.009	0.029	0.003	0.500	99.88
V2"R-2	Light to dark green	98.44	0.012	0.048	0.009	0.088	0.018	0.442	99.06
V2B-2a	Red	98.43	0.020	0.039	0.039	0.761	0.021	0.523	99.83
V2R-1b	Brownish to red	98.52	0.023	0.011	0.025	0.610	0.025	0.669	99.88

fairly high detection limit by EPMA, respectively 34 and 43 ppm (see Appendix), preventing full use of Mg and Ga (Ga/Mg ratio) as a genetic indicator (Peucat et al., 2007). The Ga/Mg ratio is commonly <6 in magmatic sapphires and <3 in metamorphic ones. Also the Fe/Mg ratio is significantly higher for magmatic sapphires (Fe/  $Mg \gg 100$ ) and lower for metamorphic and metasomatic ones (Fe/ Mg < 100; Peucat et al., 2007). The diagram proposed by Giuliani et al. (2010) discriminates mostly all the geological origins of corundum for the following reasons: (i) the database contains more than 2000 EPMA analysis representative of all the types of primary deposits; (ii) the magmatic sapphire domain (S1) is defined by the geology, chemistry and oxygen isotopes of corundums (Giuliani et al., 2005, 2007a,b, 2010; Uher et al., 2012). The sapphires are hosted in syenitic rocks or as megacrysts in syenitic xenoliths carried by alkali basalts. Most of sapphires in placers linked to the erosion of alkali basalts (domain S2) overlap the domain S1. The main superposition of domains is for ruby of metasomatic(-metamorphic) origin that overlaps the domain of ruby associated with mafic and ultramafic rocks (strictly metamorphic); (iii) the diagram uses Ga and Mg despite their high detection limit, and the two elements are opposed: Ga is added to Ti while Mg is added with Cr and V. This choice fits with the fact that Ga and Ti are concentrated in magmatic sapphires (syenitic rocks) while Mg, Cr and V are concentrated in metamorphic rubies hosted in Mg–Cr–(V)-rich protoliths.

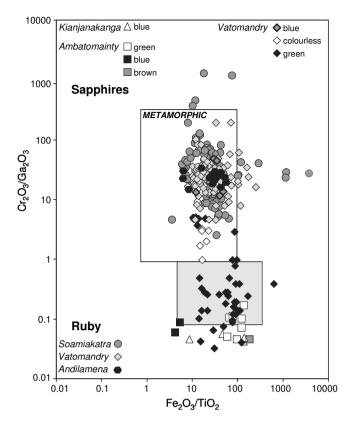
#### 3.1.1. Ruby

All the rubies plot in R3 mafic–ultramafics and R4 metasomatic domains: they lie in an ellipsoidal field with the main axis oriented NE–SW in the diagram. Several points emerge: (i) the deposits of

Soamiakatra, Vatomandry, and Andilamena show a distinctive bimodal chemical distribution (Fig. 4B). The first field in the R3 domain concerns the light purplish, dark purplish to red, mauve to purple violet rubies, and the majority of the dark purplish to red rubies from Soamiakatra. For Vatomandry, the field includes brownish to dark red and red rubies; (ii) the second field plots in the R3 and R4 domains. For Soamiakatra it includes light to dark pink ruby, brownish to dark red, and some dark purplish to red rubies; for the Vatomandy placers, these include the purplish to red and pinkish rubies. The rubies of Andilamena also plot in two distinctive fields; (iii) Antsabotraka and Ambatomainty rubies are brownish to red or dark red and overlap in the R3-R4 domains: they probably have the same origin; (iv) one field of Andilamena rubies is located in R3-R4 domains. Field observations indicate that these rubies are of metasomatic origin (i.e. desilicated rubybearing pegmatite in mafic-rocks). The other field in the R3 domain corresponds to ruby in mafic rocks.

#### 3.1.2. Sapphires

The sapphires from the three Provinces overlap the 'basaltic' chemical domain (S3). They come from placers and their origin is much debated. In the Giuliani et al. (2010) diagram (Fig. 5), the sapphires from all the basaltic Provinces plot either in the magmatic (S1) or metamorphic domains (S2), or both. These results match the conclusion from the Sutherland et al. (1998b, 2003) diagram (Fig. 3). The exact origin of these sapphires could be approached using the limit of separation for the domains S1 and S2. From this we can estimate, for each Province and kind of colour, the proportion of metamorphic sapphires (in % metam.). For the Antsiranana Province, all the grey–purplish blue–pink coloured



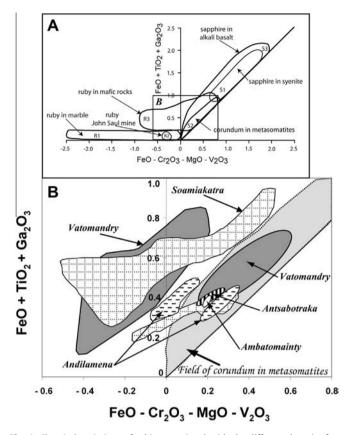
**Fig. 3.** Diagram  $Fe_2O_3/TiO_2$  vs.  $Cr_2O_3/Ga_2O_3$  for the sapphires and rubies from the Antananarivo and Toamasina Provinces. As the sapphires from Antsiranana Province are free of chromium, their chemical ratio cannot be reported in the diagram of Sutherland et al. (1998b, 2003). The metamorphic and magmatic boxes defined for Australian sapphires by Sutherland et al. (1998b, 2003) are drawn. The iron content of sapphire obtained by EPMA as FeO (in wt%) was converted to  $Fe_2O_3$  (in wt%).

sapphires aremetamorphic (100% metam.), the green-colourlesslight blue (75 < % metam. < 85), the yellow (50% metam.), deep blue-milky (20 < % metam. < 50), and orange (0% metam.). For Antananarivo Province, the Ambatomainty blue-green-brown sapphires are magmatic while blue sapphires from Kianjanankanga are 30% metam. For Toamasina Province, corundum is dominantly metamorphic with the blue and colourless sapphires (100% metam.) and green sapphires (63% metam.).

3.2. Oxygen isotopes coupled with mineral inclusions and chemistry of gem corundum: clues for their origin ?

### 3.2.1. Sapphires

3.2.1.1. Vatomandry mining district. Most of the  $\delta^{18}$ O-values of the sapphires  $(3.3 < \delta^{18}O < 4.7\%)$ ; mean  $\delta^{18}O = 4.1 \pm 0.4\%$ , n = 9; Table 2) are typical for sapphires in basaltic environments ( $3.0 < \delta^{18}O < 8.2\%$ ) mean  $\delta^{18}$ O = 5.8 ± 1.2‰; *n* = 150; Giuliani et al., 2005, 2007b; Yui et al., 2003, 2006; Fig. 6). The  $\delta^{18}$ O-values of Vatomandry sapphires overlap the range for sapphires associated with syenites  $(4.4 < \delta^{18}O < 8.3\%)$ , mean  $\delta^{18}O = 6.8 \pm 1.4\%$ , n = 25; Giuliani et al., 2007a), metasomatic sapphires linked to desilicated pegmatites  $(4.2 < \delta^{18}O < 7.5\%)$ , mean  $\delta^{18}O = 5.7 \pm 0.9\%$ ; n = 16), and biotite schists in shear zones ( $4.6 < \delta^{18}O < 9.0\%$ ; *n* = 11, Fig. 6). In the diagram  $FeO - Cr_2O_3 - MgO - V_2O_3$  vs.  $FeO + TiO_2 + Ga_2O_3$  (Fig. 5B) the blue and colourless sapphires plot in the metasomatites field while 40% of green sapphires are in the syenitic field. A magmatic origin for these sapphires is confirmed by the mineral inclusions such as members of the pyrochlore, samarskite and columbite groups (ESM Table 8). These minerals are classically found in sapphires associated with syenite, nepheline syenite or alkaline



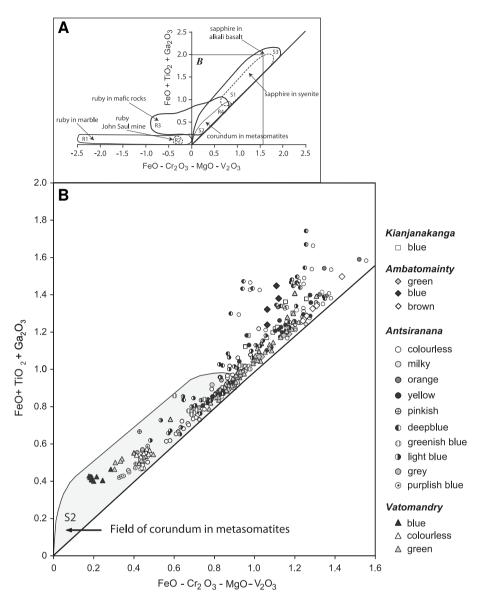
**Fig. 4.** Chemical variations of rubies associated with the different deposits from Antananarivo and Toamasina Provinces in the diagram  $FeO - Cr_2O_3 - MgO - V_2O_3$ vs. FeO + TiO<sub>2</sub> + Ga<sub>2</sub>O<sub>3</sub> (in wt%). (A) The classification diagram based on a database of 2000 EPMA analyses showing the main chemical fields defined for different types of corundum deposits worldwide (Giuliani et al., 2010; Uher et al., 2012). (B) Field of chemical composition of ruby in each deposit: the primary deposit of Soamiakatra, and the placer deposits of Antsabotraka, and Ambatomainty for Antananarivo Province, and Vatomandry for Toamasina Province. The ruby deposits of Andilamena located in Toamasina Province are not related to alkali basalts but they form a huge placer. These deposits probably derived from plumasite in mafic rocks (because rubies plot in the corundum metasomatite field) and mafic rocks (because some of these rubies plot in the corundum mafic rocks field). The iron content of sapphire measured by EPMA is expressed as FeO (in wt%).

magmatic environments enriched in incompatible elements and volatiles (Upton et al., 1999, 2009; Simonet, 2000; Monchoux et al., 2006; Graham et al., 2008; Sutherland et al., 2009; Uher et al., 2012).

Other mineral inclusions such as zircon, spinel, ilmenite, hematite, sillimanite, anorthite associated with spinel found in the green sapphires, or rutile and K-feldspar with sillimanite in colourless sapphires, indicate a metamorphic paragenesis. The  $\delta^{18}$ O-value of 9.5‰ for the colourless sapphire of Ambodirina pinpoints clearly a metamorphic skarn origin. A similar metamorphic origin based on the nature of solid inclusions and oxygen isotopes was proposed for some sapphires from the placers of the French Massif Central (Giuliani et al., 2009), e.g. Chomelix ( $\delta^{18}$ O = 8.6 ± 0.3‰, *n* = 3), Mont Coupet ( $\delta^{18}$ O = 8.7‰ and 10.25‰), and Le Bras ( $\delta^{18}$ O = 13.9‰), and also colourless or pastel coloured sapphires from Barrington in Australia (Giuliani et al., 2005; Khin Zaw et al., 2006).

3.2.1.2. Antsirabe mining districts. These sapphires have two ranges of  $\delta^{18}$ O-values (Table 2; Fig. 6):

(1)  $6.5 < \delta^{18}O < 6.9\%$  for the Antanifotsy sapphires. The isotopic range overlaps those of sapphires associated with syenite, 'plumasite' and biotite schists in shear zones;



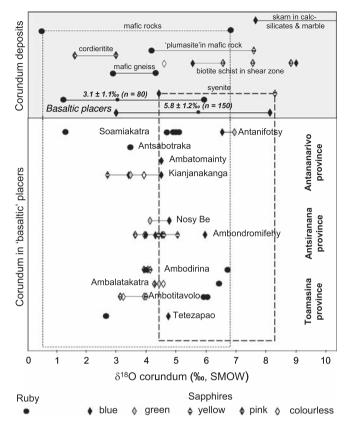
**Fig. 5.** Chemical variations of "BGY" sapphires associated with different deposits from Antsiranana, Antananarivo and Toamasina Provinces in the diagram  $FeO - Cr_2O_3 - MgO - V_2O_3$  vs.  $FeO + TiO_2 + Ga_2O_3$  (in wt%). (A) The classification diagram showing the main chemical fields defined for different types of corundum deposits worldwide (Giuliani et al., 2010; Uher et al., 2012). The iron content of sapphire measured by EPMA is expressed as FeO (in wt%). (B) Field of chemical composition of "BGY" sapphires for each deposit: Ambondromifehy and Nosy Be for Antsiranana Province, Kianjanakanga and Ambatomainty for Antananarivo Province, and Vatomandry for Toamasina Province. The sapphires plot either in the metasomatites field or in the syenitic field. They overlap the general worldwide field defined for sapphires in alkali basalt (S3).

(2)  $2.7 < \delta^{18}O < 4.5\%$  for Ambatomainty and Kianjanakanga mines in the Mandrosohasina area. The  $\delta^{18}O$ -values of the sapphires from Kianjanakanga ( $2.7 < \delta^{18}O < 3.9\%$ , n = 5) overlap the  $\delta^{18}O$ -range of the sapphires from Ambodirina, Ambalatakatra and Ambotitavolo from Vatomandry ( $3.3 < \delta^{18}O < 4.7\%$ , n = 9).

The chemical composition of all the Blue–Green–Brown sapphires from Ambatomainty and 70% of the sapphires from Kianjanakanga indicates a syenitic source (Fig. 5B). Micro-xenoliths of sapphire-bearing anorthoclasite (ESM Fig. 13C and D) at Kianjanakanga, however, indicate corundum growth from a felsitic magma enriched in incompatible and volatile elements. Xenoliths of anorthoclasite carrying sapphires have also been described in the Menet trachyte at the Puy de Menoyre in the French Massif Central (Brousse and Varet, 1966; Giuliani et al., 2010) and in the Cerová Highlands (southern Slovakia) at Gortva in the Western Carpathians (Uher et al., 2012). These xenoliths vary from a few cm to 30 cm and the blue sapphires up to1 cm long are disseminated within a potassic feldspar matrix associated with biotite and maghemite. The  $\delta^{18}$ O-values of the sapphires from Menet and Gortva are respectively between 4.4% and 4.5% (Giuliani et al., 2009) and 5.1%. They are in the range of those of Mandrosohasina district (Table 2). These  $\delta^{18}$ O-values lie in the isotopic range for syenites  $(4.4 < \delta^{18}O < 8.3\%)$ . Brousse and Varet (1966), Gaillou (2003) and Gaillou et al. (2010) have shown that these anorthoclasites are the plutonic equivalents of the trachytes. In addition, the presence of inclusions of trachytic and rhyolitic glasses in the sapphires(Gaillou, 2003) indicates that felsic magmas were parental to these anorthoclasites. Although the Mandrosohasina sapphires are magmatic, 30% of Kianjanakanga sapphires have a metamorphic origin as shown by their chemistry and their mineralogical association with ilmenite and spinel.

District	Mine	Sample	Mineral	Colour	Type of deposit	δ <sup>18</sup> O (%, V-SMOW)	References
Vatomandry	Ambodirina	A1	Ruby	Pinkish	Placer in basaltic environment	6.7	This work
		A2	Sapphire	Green to blue	Placer in basaltic environment	3.9	This work
		A3	Rubv	Light red	Placer in basaltic environment	4.0	This work
		A4	Sannhire	Colourless	Placer in hasaltic environment	9.5	This work
		A5	Sampling	Vollowitch	Discon in baselies and incompati	C 7	This work
						4 T	
		Ab	Sappnire	Greenisn	Placer in basaltic environment	4.0	I his work
	Ambalatakatra	Am1	Sapphire	Light blue to colourless	Placer in basaltic environment	4.6	This work
		Am2	Sapphire	Light green to colourless	Placer in basaltic environment	4.4	This work
		Am3	Ruby	Deep red	Placer in basaltic environment	6.4	This work
		Am4	Rubv	Light pink	Placer in basaltic environment	4.3	This work
	Amhotitavolo	11	Sannhire	Blue	Placer in hasaltic environment	3.3	This work
		1 2	Sanhire	Creen	Dlacar in hasaltic anvironment	0.6	This work
		1 1				0.0	
		Σ1	Kuby	Deep red	Placer in basaltic environment	0.0	I his work
		L4	Sapphire	Yellowish	Placer in basaltic environment	3.9	This work
		L5	Ruby	Pinkish	Placer in basaltic environment	3.2	This work
		VAT-1	Ruby	Deep red	Placer in basaltic environment	5.9	Giuliani et al. (2007)
	Tetezapao	VATO-1	Sapphire	Blue	Placer in basaltic environment	4.7	This work
	ı	VATO-2	Ruby	Red	Placer in basaltic environment	2.7	This work
Antsirahe	Snamiakatra	51	Ruhv	Deen red	Basaltic dvke primarv deposit	5.0	This work
		5 5	Ruhy	Pod Pod	Beseltic dube primary denosit	5 C C	This work
		1 5	Dubu		Deceltie delle eniment decesit	1.0	This work
		22	kuby 	Ked	basaluc dyke primary deposit	4.9	I IIIS WOLK
		RNANTA	Ruby	Red to crimson	Basaltic dyke primary deposit	4.7	Giuliani et al. (2007
		SOM-1	Ruby	Deep red	Basaltic dyke primary deposit	1.25	Giuliani et al. (2007
	Antanifotsy	SNANTAN1	Sapphire	Bluish	Placer in basaltic environment	6.5	Giuliani et al. (2005
		SNANTAN2	Sapphire	Bluish to greenish	Placer in basaltic environment	6.9	Giuliani et al. (2005)
	Ambatomainty	AMBATO-1	Sapphire	Light blue	Placer in basaltic environment	4.5	Giuliani et al. (2007
	Antsabotraka	ANTSA-1	Rubv	Red	Placer in basaltic environment	3.5	Giuliani et al. (2007)
	Mandrosohasina	MAN1	Sannhire	Yellow	Placer in hasaltic environment	7.6	
		CNAM	Sanhire	Blue	Dlacar in basaltic anvironment	45	This work
			Cambiro	Colourloss	Discontin business curriconnent		This work
						ה. יי	
	Klanjanakanga	KIA I	sappnire	Deep blue	Placer in Dasaltic environment	3.4	I NIS WOLK
		Kia2	Sapphire	Opaline	Placer in basaltic environment	3.5	This work
Antsiranana-Nosy Be	Ambondromifehy	ANT-1	Sapphire	Yellow to light green	Placer in a limestone karst	5.1	Giuliani et al. (2005)
		ANT-2	Sapphire	Light blue	Placer in a limestone karst	5.9	Giuliani et al. (2005)
		ANT-3	Sapphire	Yellow	Placer in a limestone karst	3.9	Giuliani et al. (2007)
		ANT-4	Sapphire	Opaline to blue	Placer in a limestone karst	3.9	Giuliani et al. (2007)
		Amho 12-3	Sannhire	l ioht blue to lilac	Placer in a limestone karst	3.8	
		ANT-5	Sapphire	Deen blue	Placer in a limestone karst	4.5	Giuliani et al. (2007)
		377-301	Sannhire	Vellowish to greenish	Placer in a limestone karst	46	
		322-3h	Sannhire	Blue	Placer in a limestone karst	4.3	Giuliani et al. (2007
		210.22	Caphira	Vallour	Directions in the providence of the providence o	~~~~	
	Nocv Re-Refotaka	NO-1	Sanhire	RUD	Alluvial placer on basalt granite and sandstone	4.8	Giuliani et al (2007)
		N0-2	Sannhire	Green	Alluvial placer on basalt granite and sandstone	4.7	Giuliani et al (2007)
		1 02	ounddac		ruturiat pracet our pagart, grannes and ganacoure	117	

**Table 2** Oxygen isotopes compositions (in ‰, V-SMOW) of rubies and sapphires from the different Madagascar deposits associated with alkali basalts.



**Fig. 6.** Oxygen isotope values of corundum from the different types of primary and secondary deposits related to corundum gem fields in alkali basalt from Madagascar (data from Giuliani et al., 2005, 2007a; this work). The data reported in the conventional delta notation relative to V-SMOW (Vienna Standard Mean Ocean Water) are compared with the oxygen isotopic ranges defined for corundum types deposit worldwide from the data base published by Giuliani et al. (2005, 2007a,b, 2009, 2012).

3.2.1.3. Antsiranana mining districts. The sapphires of Ambondromifehy and Nosy Be have  $\delta^{18}$ O-values between 3.9‰ and 5.9‰ (n = 11, Table 2 and Fig. 6). The mean  $\delta^{18}$ O-value of  $4.5 \pm 0.5\%$ (n = 11) is similar to Antsirabe and Vatomandry values respectively  $4.1 \pm 0.4\%$  (*n* = 8) and  $4.5 \pm 1.5\%$  (*n* = 10). The  $\delta^{18}$ O-values from the Antsiranana Province sapphires  $(3.8 < \delta^{18}O < 6.9\%, n = 11)$ overlap the isotopic range for sapphires hosted in syenite, in desilicated pegmatite cross-cuting mafic rocks (plumasite), and biotite schist from shear zones. The colour distribution of the rough sapphires from the ABFG mine in the Anivorano-Sud district indicates that 88% of the production is blue to green sapphire (and 15% blue to violet; Schwarz et al., 2000). In the present study, the blue sapphires are divided into dark blue, light blue and purple blue. The chemical diagram (Fig. 5) shows that dark blue sapphires are mostly magmatic (30% metam. and with no Cr<sub>2</sub>O<sub>3</sub>) and the light blue are largely metamorphic (70% metam. with  $150 < Cr_2O_3 < 190$  ppm); all the purplish blue sapphires are metamorphic (450 ppm <  $Cr_2O_3$  < 620 ppm). The greenish blue sapphires are also metamorphic in origin (85% metam. and with 450 ppm <  $Cr_2O_3$  < 620 ppm).

The mineral inclusions identified in the Ambondromifehy sapphires indicate both their metamorphic and magmatic origins (ESM Table 1). Magmatic paragenesis is characterized by the association columbite-(Fe), tantalite-(Fe), columbite-(Mn), Nb-rich rutile, uraninite, pyrochlore group, samarskite group, (ESM Fig. 8A–D). Metamorphic paragenesis is indicated by the association of hematite, rutile and zircon (ESM Table 1) as described in the sapphires from the desilicated pegmatites of Umba and Kalalani in Tanzania (Seifert and Hyrsl, 1999). The presence of U-rich

thorianite and thorianite in the sapphires from Ambondromifehy could indicate a skarn origin such as those found in the Tranomaro granulites from southern Madagascar (Rakotondrazafy et al., 1996). Further, the paragenesis of the gangue preserved around the sapphires pinpoints a metamorphic origin: K-feldspar, garnet and monazite groups (ESM Fig. 8E) or K-feldspar, biotite group, Ca-rich albite, spinel series, sillimanite, zircon, apatite group and monazite group (ESM Fig. 8F). This paragenesis is similar to that identified in sapphires hosted in biotite schists related to a shear zone in the Sahambano district, near Ihosy (Ralantoarison et al., 2006; Rakotondrazafy et al., 2008).

Given the consistency of the oxygen isotope data, and the paragenesis and chemical features of the sapphires, both metamorphic and magmatic origins are clearly evident. Dark blue sapphires tend to be magmatic while greenish to light blue sapphires are metamorphic.

Around half of the  $\delta^{18}$ O-values of the sapphires (2.7 <  $\delta^{18}$ O < 3.9‰) from the three Provinces are anomalously low for sapphires originating from a basaltic environment  $(3.0 < \delta^{18}O < 8.2\%)$ n = 150; Giuliani et al., 2007a; and this work). They are too low for an original mantle signature normally expected at  $5.6 \pm 0.3$ for clinopyroxene in unaltered peridotite (Mattey et al., 1994). The experimental work of Hong-Sen et al. (2002) demonstrated the coexistence of megacrysts corundum with garnet by the transformation of alkali basalt into eclogite at the pressure of 33 kb for a temperature of 1400 °C. Sutherland et al. (1998b, 2003) proposed that the partial melting of the lithospheric mantle rich in amphibole could permit crystallization of corundum in the residual liquid. Nevertheless, such a mechanism would not explain  $\delta^{18}$ O-values less than 5.5‰. The experimental work of Hirschmann et al. (2003) demonstrated that at  $P \sim 20-25$  kb, the partial melting of garnet pyroxenites would produce alkaline magmas rich in alumina. Such magmas could form corundum during crystal fractionation in the upper mantle. Yui et al. (2006) indicated that these magmas could originate from the melting of garnet pyroxenites within mantle peridotites following a hybrid model of formation. These garnet pyroxenites would originate from subducted oceanic cumulates of gabbroic composition that reacted at high temperature with seawater before subduction and infiltration, by metamorphic fluids (in the case of metamorphic sapphires), or by mantle magmas producing aluminous magmas (in the case of magmatic sapphires). The  $\delta^{18}$ O-values less than 5.5% would be a consequence of the interaction of the pre-oceanic cumulates with seawater (Yui et al., 2006); such a mechanism might also account for the heterogeneous distribution of the low  $\delta^{18}$ O values.

#### 3.2.2. Rubies

The  $\delta^{18}$ O-values of the rubies from the Vatomandry mining district span 2.7–6.7‰ (n = 8) with a mean value of 5.1 ± 1.4‰ (Fig. 6). They fit within the worldwide range defined for ruby in basaltic environments  $(1.3 < \delta^{18}O < 5.9\%)$ , mean  $\delta^{18}O = 3.1 \pm 1.1\%$ , n = 80; Giuliani et al., 2007b) and ruby associated with mafic and ultramafic rocks ( $0.25 < \delta^{18}O < 6.8\%$ , n = 19). Two O-isotopes composition ranges are observed for Vatmomandry (Fig. 6), respectively between 2.7% and 4.3% (n = 4) and 5.9% and 6.7% (n = 4):(i) The first range fits within the oxygen isotopic range of rubies from Soamiakatra and Antsabotraka  $(1.25 < \delta^{18}O < 5.0\%, n = 6)$ . At Soamiakatra, the host-rocks of ruby are metagabbros and pyroxenites (Rakotosamizanany, 2009: Rakotosamizanany et al., in preparation). Rubies with similar O-isotopic range  $(1.3 < \delta^{18}O < 4.2\%)$ are described in the Chanthaburi-Trat alkali-basalt gemfield from Thailand (Yui et al., 2006). These xenocrystic rubies contain garnet and clinopyroxene. They formed in mafic metamorphic rocks under subcontinental upper mantle conditions and their protolith was a garnet pyroxenite (Sutthirat et al., 2001). Such mineral assemblages have been also found in ruby-bearing garnet pyroxenites within orogenic peridotite from Beni Bousera, Morocco (Kornprobst et al., 1990), Val Malenco in Italy (Muntener and Hermann, 1996) and in eclogites from China (Yui et al., 2006).

The  $\delta^{18}$ O-values of ruby between 1.25‰ and 3.2‰ (ruby from Soamiakatra, Antsabotraka, Ambotitavolo and Tetezapao, and from Chanthaburi-Trat) are too low for pristine upper mantle and indicate a previous high-temperature seawater interaction of the parental rocks of the ruby-bearing metagabbros and pyroxenites (Yui et al., 2006). This hypothesis was proposed by Pearson et al. (1993) for the low  $\delta^{18}$ O-values of clinopyroxene from the ruby-bearing pyroxenites from Beni Bousera peridotite complex, and Alt (1995) for ultramafics within ophiolites and modern oceanic crust.(ii) The second oxygen isotopic range fits with that defined for metamorphic ruby in mafic rocks and plumasites (4.2 <  $\delta^{18}$ O < 7.5‰, mean  $\delta^{18}$ O = 5.7 ± 0.9‰; *n* = 16). Both origins are plausible as shown by the chemical composition and the nature of the solid inclusions trapped by these rubies.

# 3.3. U/Pb ages of zircon associated with gem corundum from Antananarivo and Antsiranana Provinces

U–Pb dating of zircons yield indirect ages for zircon crystals associated with sapphire and/or ruby in the paleoplacers. The four U–Pb ages reported are not sufficient to propose a scenario concerning crystallization of corundum and zircon but they help to constrain the temporal relationship between their formation and the age of alkali-basalt volcanism.

### 3.3.1. Zircons from Antananarivo Province: Mandrosohasina and Ambatomainty paleoplacers

Two U–Pb analyses were performed on zircons associated with sapphires in paleoplacers (Table 3). The analyses, done by ICPMS-laser ablation, gave similar weighted mean  $^{206}$ Pb– $^{238}$ U ages respectively, 7.3 ± 0.2 Ma for Mandrosohasina and 7.1 ± 0.2 Ma for Ambatomainty (Fig. 7A and B). In Antananarivo Province, the volcanism of Ankaratra is considered Oligocene to Pleistocene. K–Ar and  $^{40}$ Ar/ $^{39}$ Ar dating on whole rocks of high-titanium phosphorus basalt, ankaratrite, basanite and mugearite yielded ages between 27.9 and 1.44 Ma (Mottet, 1981; Emerick and Duncan, 1982; Tucker et al., 2008; Bardintzeff et al., 2010). The U–Pb ages obtained for two zircon grains are similar, within the range 7.3–7.1 ± 0.2 Ma and support magmatism in this region during the Miocene (Tortonian–Messinian boundary).

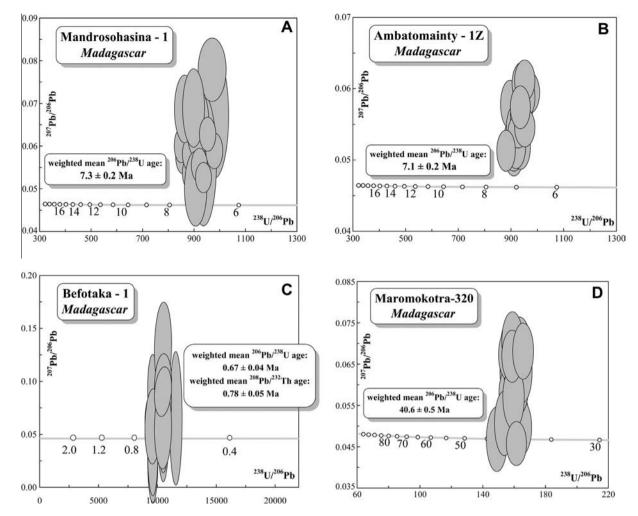
# 3.3.2. Zircons from Antsiranana Province: Nosy Be and Ambondromifehy paleoplacers

Two U–Pb analyses were performed by ICPMS-laser ablation on two zircons Befotaka-1 and Maramokotra-320, respectively from Nosy Be and Ambondromifehy placers (Table 4). Befotaka-1 yielded a mean  $^{206}$ Pb– $^{238}$ U age of 0.67 ± 0.04 Ma (Fig. 7C) comparable to the

#### Table 3

U-Pb ages obtained by excimer laser ablation (ELA)-ICP-MS laser on zircon grains from the Ambatomainty and Mandrohasina sapphire deposits in the Antananarivo Province (Antsirabe region).

Antananarivo Province (Antsirabe)	<sup>206</sup> Pb/ <sup>238</sup> U	Error $2\sigma$	<sup>207</sup> Pb/ <sup>206</sup> Pb	Error $2\sigma$	Rho	$^{206}$ Pb/ $^{238}$ L Ages (2 $\sigma$ )
Ambatomainty-1Z						
AMB1Z-1	0.001120	0.000039	0.0526	0.0030	0.54	$7.2 \pm 0.3$
AMB1Z-2	0.001140	0.000040	0.0578	0.0030	0.59	$7.4 \pm 0.3$
AMB1Z-3	0.001090	0.000038	0.0561	0.0030	0.56	$7.0 \pm 0.2$
AMB1Z-4	0.001120	0.000039	0.0511	0.0029	0.55	$7.2 \pm 0.3$
AMB1Z-5	0.001120	0.000039	0.0513	0.0025	0.61	$7.2 \pm 0.3$ $7.2 \pm 0.3$
AMB1Z-6	0.001090	0.000038	0.0538	0.0025	0.63	$7.0 \pm 0.2$
AMB1Z-7	0.001110	0.000039	0.0525	0.0025	0.58	$7.0 \pm 0.2$ $7.1 \pm 0.2$
AMB1Z-7 AMB1Z-8	0.001090	0.000038	0.0518	0.0026	0.59	$7.0 \pm 0.2$
AMB12-9	0.001050	0.000037	0.0594	0.0026	0.66	$6.8 \pm 0.2$
AMB12-10	0.001080	0.000038	0.0607	0.0027	0.65	$7.0 \pm 0.3$
AMB12-10 AMB1Z-11	0.001110	0.000039	0.0546	0.0029	0.57	$7.0 \pm 0.3$ $7.2 \pm 0.2$
AMB1Z-12	0.001080	0.000038	0.0560	0.0029	0.60	$7.2 \pm 0.2$ $7.0 \pm 0.2$
AMB1Z-12 AMB1Z-13	0.001030	0.000038	0.0589	0.0023	0.64	$6.9 \pm 0.2$
AMB1Z-13 AMB1Z-14	0.001090	0.000038	0.0562	0.0025	0.64	$0.9 \pm 0.2$ 7.0 ± 0.2
AMB1Z-14 AMB1Z-15	0.001090	0.000039	0.0519	0.0025	0.59	$7.0 \pm 0.2$ $7.1 \pm 0.2$
AMB1Z-15 AMB1Z-16	0.001110	0.000039	0.0543	0.0020	0.39	$7.1 \pm 0.2$ $7.1 \pm 0.2$
AMB1Z-10 AMB1Z-17	0.001100	0.000039	0.0615	0.0021	0.68	$7.1 \pm 0.2$ $6.9 \pm 0.2$
AMB1Z-17 AMB1Z-18	0.001070	0.000038	0.0548	0.0025	0.65	$6.9 \pm 0.2$ $6.9 \pm 0.2$
	0.001160	0.000038		0.0025	0.65	
AMB1Z-19			0.0514			7.5 ± 0.3
AMB1Z-20	0.001090	0.000038	0.0574	0.0027	0.63	$7.0 \pm 0.2$
Mandrosohasina-1	0.001040	0.000051	0.0074	0.0100	0.20	67.00
MAN1-1	0.001040	0.000051	0.0674	0.0126	0.26	6.7 ± 0.3
MAN1-2	0.001090	0.000052	0.0565	0.0129	0.21	$7.0 \pm 0.3$
MAN1-3	0.001050	0.000051	0.0779	0.0069	0.53	$6.8 \pm 0.3$
MAN1-4	0.001120	0.000032	0.0602	0.0034	0.44	$7.2 \pm 0.2$
MAN1-5	0.001200	0.000033	0.0572	0.0031	0.44	7.8 ± 0.2
MAN1-6	0.001210	0.000034	0.0602	0.0032	0.45	7.8 ± 0.2
MAN1-7	0.001140	0.000032	0.0560	0.0029	0.46	7.3 ± 0.2
MAN1-9	0.001150	0.000033	0.0558	0.0031	0.43	$7.4 \pm 0.2$
MAN1-10	0.001160	0.000033	0.0537	0.0031	0.42	$7.4 \pm 0.2$
MAN1-11	0.001160	0.000033	0.0581	0.0032	0.45	7.5 ± 0.2
MAN1-12	0.001140	0.000032	0.0663	0.0044	0.38	$7.4 \pm 0.2$
MAN1-13	0.001190	0.000048	0.0684	0.0068	0.39	7.7 ± 0.3
MAN1-14	0.001120	0.000047	0.0651	0.0072	0.37	$7.2 \pm 0.3$
MAN1-15	0.001140	0.000047	0.0688	0.0079	0.35	7.3 ± 0.3
MAN1-16	0.001130	0.000047	0.0497	0.0074	0.28	7.3 ± 0.3
MAN1-17	0.001040	0.000031	0.0586	0.0034	0.45	$6.7 \pm 0.2$
MAN1-18	0.001110	0.000032	0.0552	0.0031	0.44	$7.2 \pm 0.2$
MAN1-19	0.001070	0.000031	0.0625	0.0032	0.49	$6.9 \pm 0.2$
MAN1-20	0.001090	0.000032	0.0526	0.0028	0.46	$7.0 \pm 0.2$



**Fig. 7.** U–Pb ages obtained by ICP-MS laser ablation analysis on zircons associated with the sapphires from the central and northern corundum gem alkali basalt Provinces of Madagascar. (A) <sup>238</sup>U/<sup>206</sup>Pb vs. <sup>207</sup>Pb/<sup>206</sup>Pb vs. <sup>207</sup>Pb/<sup>206</sup>Pb diagram for Mandrosohasina-1 (Antsirabe area). (B) <sup>238</sup>U/<sup>206</sup>Pb vs. <sup>207</sup>Pb/<sup>206</sup>Pb diagram for Ambatomainty-1Z (Antsirabe area). (C) <sup>238</sup>U/<sup>206</sup>Pb vs. <sup>207</sup>Pb/<sup>206</sup>Pb diagram for Befotaka-1 (Nosy Be island, Antsiranana Province). (D) <sup>238</sup>U/<sup>206</sup>Pb vs. <sup>207</sup>Pb/<sup>206</sup>Pb diagram for Maramokotra-230 (Ambondromifehy mines, Antsiranana Province).

 $^{208}\text{Pb}-^{232}\text{Th}$  age of 0.78  $\pm$  0.05 Ma. The zircon from Maramokotra-320 yielded a weighted mean  $^{206}\text{Pb}-^{238}\text{U}$  age of 40.6  $\pm$  0.5 Ma (Fig. 7D).

Montagne d'Ambre, Ambato peninsula and Nosy Be-Nosy Mitsio and Nosy Komba islands belong to the Mesozoic volcanism from the eastern Indian Ocean. K-Ar dating on whole rocks of felsic and mafic lavas yielded ages between 14 and 0.8 Ma (Karche, 1973; Emerick and Duncan, 1982; Tucker et al., 2008; Cucciniello et al., 2011; Fig. 8).  $^{40}$ Ar/ $^{39}$ Ar dating on whole rock of kaersutite from Nosy Komba and on K-feldspar of a quartz monzonite from the Lokobe complex in Nosy Be, respectively yielded ages of 20.7 and 21 Ma (Tucker et al., 2008). <sup>40</sup>Ar/<sup>39</sup>Ar dating on plagioclase of hawaiite from Montagne d'Ambre and on a sanidine of a phonolite dyke from the Bobaomby peninsula, respectively yielded ages of 12.1 and 10.5 Ma (Cucciniello et al., 2011). The syenitic dyke of Andavakoera in the southern part of the Montagne d'Ambre gave a whole rock <sup>40</sup>Ar/<sup>39</sup>Ar age at 52 Ma (Tucker et al., 2008) and the zircon associated with sapphire in the deposit of Maramokotra yielded an U/Pb age at  $40.6 \pm 0.5$  Ma (this work). These oldest ages measured from Montagne d'Ambre suggest that some sapphire may have formed in the Eocene (Lutetian/Bartonian boundary). In contrast, a Befotaka zircon age at 0.78 ± 0.05 Ma represents the youngest likely age for sapphire crystallization in Nosy Be island.

# 4. Metallogenic model: mechanism of formation and transport of gem corundum in alkali basalts from Madagascar

The three corundum-bearing basaltic Provinces of Madagascar exhibit mixed features. The main differences are the absence of ruby in Antsiranana Province; the presence of a primary ruby deposit in Antananarivo Province; and the almost total absence of blue sapphire in Toamasina Province. The chemistry, nature of mineral inclusions and oxygen isotopic composition imply a metamorphic origin for ruby in all the deposits: (i) metamorphic s.s. in mafic or ultramafic rocks as described in the southern Madagascar Precambrian (Nicollet, 1986, 1988, 1990); and (ii) metasomatichydrothermal in plumasite. The ruby from Soamiakatra is hosted by metagabbros and garnet-bearing pyroxenite enclaves. The paragenesis formed at T > 1100 °C for  $P \sim 20$  kb in the eclogite facies (Rakotosamizanany, 2009). The other likely sources for ruby in both Provinces are mafic rocks such as amphibolite found in the granulitic basement located in southern Ihosy (Nicollet, 1986; Rakotondrazafy et al., 2008).

The sapphires have either a metamorphic or magmatic origin. Magmatic sapphires in the three Provinces are iron and titaniumrich for blue and green colours and with mineral inclusions (columbite-(Fe), pyrochlore group, samarskite group, Nb-rich tantalite and Nb-bearing rutile) typical of syenite or nepheline syenite. These syenitic magmas are enriched in incompatible and volatile elements (Aspen et al., 1990; Upton et al., 1999; Sutherland et al., 1998a,b; Peucat et al., 2007; Graham et al., 2008). The sapphire-bearing albitites from Urdach and Espechère in the Pyrénées have low Rb contents (2-11 ppm) but high to very high contents of Sr, Ba, Nb (167-299 ppm) and Ta (6-17 ppm), and extreme contents of light REE (Pin et al., 2006). In addition, the presence of micro-xenoliths of sapphire-bearing anorthoclasite in the Mandrohasina mining district indicates that these sapphires have crystallized from a Na-K-rich syenitic magma. This sapphire association with anorthoclasite is similar to the rocks described in the French Massif Central by Brousse and Varet (1966) and the Western Carpathians by Uher et al. (2012). Pin et al. (2006) proposed the formation of sapphire by fractional crystallization of a mantle magma source with an initial  $\delta^{18}$ O of 5–6% (Yui et al., 2006). The alkaline magma would result from a very low degree of partial melting (<1%) of a spinel-bearing lherzolite. A similar model has been proposed by Giuliani et al. (2009) for the formation of the anorthoclasites from Menet in the French Massif Central.

Corundum-bearing alkali-basalts are formed in intra-continental rifts (Guo et al., 1996a,b; Sutherland, 1996; Sutherland and Coenraads, 1996; Merle et al., 1998; Sutherland et al., 1998a; Saminpanya, 2000; Sutthirat et al., 2001; Graham et al., 2008; Giuliani et al., 2010; Saminpanya and Sutherland, 2011). The extension and thinning of the continental crust resulted in the formation of sedimentary basins limited by fault blocks that swivel towards the rift axis. The tilted blocks are limited by a network of complex faults and fractures parallel to the direction of the rift. In the example of the French Pyrénées Iherzolites, the basin formed by pull apart and is controlled by strike slip faults (Lagabrielle and Bodinier, 2008). The formation of the alkaline magmatism and its rise to the surface is related to the presence of a thermal aureole due to mantle upwelling (Sutherland et al., 1998a).

Mantle upwelling was identified in the central, eastern and northern parts of Madagascar (Nougier et al., 1986; Piqué et al., 1999; de Wit, 2003; Kusky et al., 2006). In Antananarivo Province and more precisely in the Ankaratra volcanic massif, the gravity and seismic data show undulations of the isobaths of the mantle and the presence of a thinned crust at 32–30 km (Rakotondraompiana et al., 1999; de Wit, 2003). Rasimanana (1996) established the causal relationship between asthenospheric rise and the elevation of the Malagasy basement in the area with concomitant volcanism since the Miocene. U/Pb dating obtained on zircons associated with 'basaltic' sapphires from the Mandrohasina–Kianjanakanga gave

Table 4

U-Pb ages obtained by excimer laser ablation (ELA)-ICP-MS laser on zircon grains from the Maramokotra (Ambondromifey mines) and Befotaka sapphire deposits (Nosy Be island) in the Antsiranana Province.

Analyse	<sup>206</sup> Pb/ <sup>238</sup> U	Error $2\sigma$	<sup>207</sup> Pb/ <sup>206</sup> Pb	Error $2\sigma$	Rho	$^{206}$ Pb/ $^{238}$ U	
Antsiranana Province (Montagne d'Ambre)						Ages $(2\sigma)$	
Maromokotra-320							
MAR320-1	0.006770	0.000251	0.0466	0.0065	0.26	43.5 ± 1.6	
MAR320-2	0.006390	0.000230	0.0674	0.0076	0.31	41.1 ± 1.5	
MAR320-4	0.006210	0.000227	0.0511	0.0071	0.26	39.9 ± 1.5	
MAR320-5	0.006160	0.000227	0.0686	0.0066	0.37	39.6 ± 1.5	
MAR320-6	0.006150	0.000227	0.0495	0.0059	0.30	39.5 ± 1.5	
MAR320-7	0.006380	0.000230	0.0630	0.0066	0.33	$41.0 \pm 1.5$	
MAR320-8	0.006360	0.000205	0.0551	0.0046	0.38	40.9 ± 1.1	
MAR320-9	0.006450	0.000207	0.0580	0.0053	0.35	$41.4 \pm 1.1$	
MAR320-10	0.006340	0.000205	0.0652	0.0062	0.34	40.7 ± 1.1	
MAR320-11	0.006390	0.000222	0.0670	0.0059	0.39	41.0 ± 1.1	
MAR320-12	0.006220	0.000220	0.0585	0.0053	0.38	$40.0 \pm 1.1$	
MAR320-13	0.006560	0.000224	0.0506	0.0066	0.26	42.1 ± 1.1	
MAR320-14	0.006440	0.000222	0.0673	0.0056	0.41	$41.4 \pm 1.1$	
MAR320-15	0.006110	0.000202	0.0631	0.0066	0.31	39.3 ± 1.1	
MAR320-16	0.006340	0.000221	0.0706	0.0062	0.39	40.7 ± 1.1	
MAR320-17	0.006310	0.000221	0.0568	0.0066	0.30	$40.5 \pm 1.1$	
MAR320-17 MAR320-18	0.006350	0.000221	0.0668	0.0056	0.30	$40.3 \pm 1.1$ $40.8 \pm 1.1$	
MAR320-18 MAR320-19	0.006240	0.000221	0.0456	0.0036	0.41	$40.8 \pm 1.1$ $40.1 \pm 1.1$	
MAR320-19 MAR320-20	0.006080	0.000204	0.0681	0.0048	0.32	$40.1 \pm 1.1$ $39.1 \pm 1.1$	
WAR520-20		0.000202		0.0000	0.57		
Analyse	<sup>206</sup> Pb/ <sup>238</sup> U	Error $2\sigma$	<sup>207</sup> Pb/ <sup>206</sup> Pb	Error $2\sigma$	Rho	<sup>206</sup> Pb/ <sup>238</sup> U	<sup>208</sup> Pb/ <sup>232</sup> T
Antsiranana Province (Nosy Be)						Ages $(2\sigma)$	Ages $(2\sigma)$
Befotaka-1							
BEF1-1	0.000110	0.000004	0.0296	0.0169	0.04	$0.71 \pm 0.08$	-
BEF1-2	0.000100	0.000004	0.1097	0.0201	0.11	$0.60 \pm 0.08$	$1.1 \pm 0.1$
BEF1-3	0.000100	0.000004	0.0564	0.0199	0.06	$0.65 \pm 0.08$	$0.9 \pm 0.1$
BEF1-4	0.000090	0.000004	0.0645	0.0244	0.05	$0.58 \pm 0.08$	$0.8 \pm 0.1$
BEF1-5	0.000100	0.000004	0.0607	0.0181	0.07	$0.64 \pm 0.04$	$0.9 \pm 0.1$
BEF1-6	0.000110	0.000004	0.0372	0.0173	0.04	$0.71 \pm 0.04$	$0.8 \pm 0.1$
BEF1-7	0.000100	0.000004	0.1151	0.0154	0.15	$0.70 \pm 0.04$	$0.7 \pm 0.1$
BEF1-8	0.000100	0.000004	0.0822	0.0122	0.14	$0.60 \pm 0.04$	$0.8 \pm 0.1$
BEF1-9	0.000100	0.000004	0.0898	0.0138	0.13	$0.70 \pm 0.04$	$0.7 \pm 0.1$
BEF1-10	0.000110	0.000004	0.0941	0.0128	0.14	$0.70 \pm 0.04$	$0.7 \pm 0.1$
BEF1-11	0.000100	0.000004	0.0598	0.0169	0.07	$0.70 \pm 0.04$	$0.9 \pm 0.1$
BEF1-12	0.000100	0.000006	0.0804	0.0136	0.11	$0.64 \pm 0.04$	$0.9 \pm 0.1$
BEF1-13	0.000100	0.000006	0.1281	0.0166	0.14	$0.70 \pm 0.04$	$0.8 \pm 0.1$
BEF1-14	0.000110	0.000007	0.0692	0.0100	0.14	$0.70 \pm 0.04$ $0.70 \pm 0.04$	$0.6 \pm 0.1$
BEF1-14 BEF1-15	0.000110	0.000007	0.0593	0.0105	0.12	$0.70 \pm 0.04$ $0.70 \pm 0.04$	$0.0 \pm 0.1$ $0.7 \pm 0.1$
BEF1-15 BEF1-16	0.000110	0.000007	0.0595	0.0108	0.10	$0.70 \pm 0.04$ $0.71 \pm 0.04$	$0.7 \pm 0.1$ $0.8 \pm 0.1$
BEF1-10 BEF1-17	0.000110	0.000007	0.0953	0.0104	0.11		
						$0.70 \pm 0.04$	$0.9 \pm 0.1$
BEF1-18	0.000110	0.000007	0.0588	0.0141	0.08	$0.71 \pm 0.04$	$0.6 \pm 0.1$
BEF1-19	0.000100	0.000006	0.0596	0.0164	0.07	$0.64 \pm 0.04$	$1.0 \pm 0.1$
BEF1-20	0.000100	0.000006	0.0907	0.0187	0.09	$0.70 \pm 0.04$	$0.9 \pm 0.1$

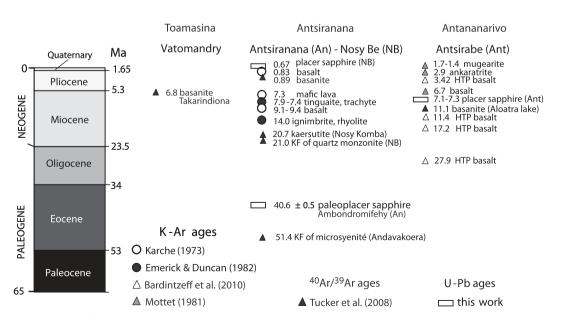
ages around 7 Ma and confirm the role played by the alkaline volcanism for the transport and formation of the sapphires in the Miocene at Ankaratra.

Farther east, the extensional tectonics are characterized by the formation of the rifts of Alaotra and Ankai (Laville et al., 1998; Melluso et al., 2011) covered by Neogene to Quaternary sediments (de Wit, 2003), and a succession of parallel basins outlined by normal faults oriented north-south. At the scale of the African continent, the orientation of the central Malagasy rift is parallel to the direction of the extensional structures from East Africa and the Mozambique Channel (i.e. parallel to the Davie ridge; Chorowicz et al., 1987; Kusky et al., 2006). <sup>40</sup>Ar/<sup>39</sup>Ar dating on whole rock of basanite from Alaotra lake (Fig. 1) yielded an age of 11.1 Ma (Fig. 8; Tucker et al., 2008) and confirms the Miocene age found for the majority of the volcanic rocks dated in the Ankaratra massif (Fig. 8). These faults have the same direction parallel to the Indian Ocean coast in the Vatomandry region which is characterized by the presence of the Takarindiona-Vohibalaina alkali-basalt massif and the corundum mining districts. The whole rock <sup>40</sup>Ar/<sup>39</sup>Ar age at 6.8 Ma of a basanite from the Takarindonia massif confirms the Miocene age of the basalt and opens the question of the age of sapphires and rubies in the associated placers. The ankaratrites (olivine melilitite and olivine nephelinite) of the Alaotra and Takarindiona volcanics shows enrichment in the most strongly incompatible elements such as Ba and Nb (Melluso et al., 2011). Their genesis is related to the rifting events which a low-partial melting of a garnet-peridotite in the lowermost lithosphere.

The northern part of Madagascar formed by the Montagne d'Ambre volcanic complex, the Ampasindava peninsula and the Comores archipelago belongs to the Mesozoic–Cenozoic volcanism of the eastern Indian Ocean. Various studies of the Cenozoic volcanism allowed characterization of different petrological facies and determination of the ages of the lavas (Nougier et al., 1986; Class et al., 1998; Melluso and Morra, 2000). The age of the volcanism characterized by K–Ar data on micas increases from the Grande Comore (0–1 Ma) to Mayotte (1.5–5.8 Ma); then Nosy Be (7–10 Ma). The ages of alkaline volcanism from Montagne d'Ambre are between 0.8 and 52 Ma but the volcanism was active until the Quaternary (Emerick and Duncan, 1982; Nougier et al., 1986; Tucker

et al., 2008). The U/Pb age of circa 41 Ma obtained on a zircon associated with a 'basaltic' sapphire from Maramokotra is Eocene (Fig. 8). Such an older age is feasible within the evolution of the volcanic field, as the ages of the Seychelles volcanism, the northeast of Montagne d'Ambre, are between 60 to 70 Ma (Nougier et al., 1986). The Th/Pb age of 0.78 Ma found for a zircon associated with the sapphires of Befotaka in Nosy Be is coeval with the <sup>40</sup>Ar/<sup>39</sup>Ar age of 0.83 obtained by whole rock dating of a basanite from Nosy Be (Fig. 8; Tucker et al., 2008). This age indicates that a small part of the volcanism on the island was active simultaneously with the volcanic emissions of the Grande Comore or Montagne d'Ambre (age 0.8 Ma; Emerick and Duncan, 1982; BGS-USGS-GLW, 2008). The absence of sapphires associated with the volcanism of the Comorian islands can be explained by the nature and the composition of the lithosphere (de Wit, 2003). The Comores Islands are located on a 135-142 Ma old and  $\sim$ 100 km thick oceanic lithosphere (Class and Goldstein, 1997: Class et al., 1998). The basanites of Nosy Be, however, formed through low degrees of partial melting (4-5%) of the subcontinental lithospheric mantle spinel peridotite enriched in incompatible elements during the Pan-African events (Melluso and Morra, 2000). Indeed, the Sm-Nd isotope systematics of the volcanic rocks from Nosy Be indicate significant participation of a subcontinental lithospheric source enriched during the Neoproterozoic (Nd model age = 550-610 Ma; Melluso and Morra, 2000). This Nd model age is within the range of U/Pb ages defined for the Precambrian formations and associated gem corundums from southern Madagascar (Paquette et al., 1994; Paquette and Nédélec, 1998; Giuliani et al., 2007a).

Thus, the rifting episodes at the end of the Pan-African orogenesis were responsible for the changes of the mantellic lithosphere under the African continent (Ashwal and Burke, 1989), with fertile mantle coming from the lithosphere. The melting of this lithosphere, enriched in incompatible elements and volatiles, under the northern part of Madagascar would explain the presence of K and Ti-rich lavas in Montagne d'Ambre. The basanites, alkali basalts, transitional and tholeiitic basalts have Zr/Nb, Ba/Nb and La/Nb ratios typical of incompatible element-rich (Nb and K) within plate basalts (Cucciniello et al., 2011). The Montagne d'Ambre



**Fig. 8.** Synthesis of the K–Ar and <sup>40</sup>Ar/<sup>39</sup>Ar ages obtained respectively on whole rocks and K-feldspar of volcanic rocks (Karche, 1973; Emerick and Duncan, 1982; Mottet, 1981; Tucker et al., 2008; Bardintzeff et al., 2010). U–Pb ages (this work) were obtained on zircons associated with the sapphires from respectively the Ambondromifehy (Maromokotra deposit) and Nosy Be (Befotaka deposit) mining districts in the Antsiranana Province, and the Mandrosohasina and Ambatomainty deposits in the Antsirabe region, Antananarivo Province.

primitive compositions are the product of variable degrees of partial melting (4 to 12%) of element-enriched mantle source. This main difference with the Comores would explain the presence of K-Ti-Nb-Ta-rich magmas thatgenerated sapphire-bearing anorthoclasites. The presence of metamorphic sapphires in the Antsiranana deposits indicates that Precambrian basement was intruded by the alkali-basalts during their ascent to the surface. The absence of ruby indicates the absence of mafic to ultramafic rocks in the basement.

The central and eastern corundum Provinces are affected by intense extensional tectonics registered in the Ankaratra mountains and Vatomandry area. The E-W extension greatly affected the lower part of the continental crust and the upper mantle of Pan-African age that contains ruby in mafic and/or ultramafic rocks or 'plumasites' and sapphires in the different types of corundum deposits found in granulites (Giuliani et al., 2007a; Rakotondrazafy et al., 2008). The intrusion of these rocks by the alkali-basalts explains their presence as metamorphic-metasomatic rubies and/or sapphires in the melt and their transport to the surface. The ruby deposit of Soamiakatra shows that some of these rubies formed at the boundary of the eclogite and granulite domains  $(HT \sim 1100 \text{ °C}, HP \sim 20 \text{ kb})$  in metagabbros and pyroxenites. The magmatic sapphires result from a complex evolution probably starting with very low degrees of partial melting (<1%) of a spinel-bearing lherzolite to produce a syenitic magma as proposed by Pin et al. (2006).

### 5. Conclusions

Madagascar gem corundums related to basaltic environments are found in the Provinces of Antsiranana (Montagne d'Ambre and Nosy Be), Antananarivo (Ankaratra massif) and Toamasina (Vatomandry area). Most of the deposits are either largely secondary as in placers or paleoplacers with only one known deposit of primary origin (viz. Soamiakatra).

Mineralogical and chemical studies of 'basaltic' corundum coupled to their oxygen isotopic composition have advanced the understanding of their origin. Iron is the main chromophore element for sapphire and the variation in the Fe<sup>2+</sup>/Fe<sup>3+</sup> ratio combined with chromium or titanium contents in the parental fluid will determine the final colour of the crystals. Chromium is the main chromophore element for ruby, but iron and vanadium also act as colour elements. High quality rubies have  $Fe_2O_3/Cr_2O_3 < 1$ . Chemical composition and oxygen isotopes confirmed the metamorphic origin of all the rubies. They are xenocrysts or crystals in xenoliths extracted by the basalts from mafic-ultramafic rocks or 'plumasites'. Sapphires are either metamorphic xenocrysts coming from 'plumasite', skarn or biotite schist, from the Precambrian basement, or magmatic euhedral crystals that have grown in an alkaline-alumina-rich magma derived from very low degree of partial melting of a spinel lherzolite. The alkali-basalts are related to intracontinental volcanism associated with an extensional rift system developed in three distinct geographic zones. In the Ankaratra plateau and Montagne d'Ambre, the volcanism and the presence of magmatic gem corundum is due to a thermal anomaly and the upwelling of the asthenosphere. Volcanic conduits formed by extensional tectonics that extended from the Ankaratra massif to the Vatomandry region.

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### Appendix A

### A.1. Analytical methods

The polished and thin sections have been examined under backscattered electron imaging (BSE) with a Hitachi S-4800 scanning electron microprobe (SEM at the University of Lorraine, Vandœuvre-lès-Nancy). The electron-probe micro-analyses (EPMA) were obtained on a Cameca SX100. The operating conditions for the corundum analysis were: acceleration voltage 15 kV, beam current 10 nA, 1  $\mu$ m beam size, collection time of 20 s for aluminium, and 25 kV, 150 nA, 120 s for trace elements (Mg, Ti, V, Cr, Fe and Ga). The detection limits for the trace elements (in ppm) are: Mg = 34; Ti = 22; V = 22; Cr = 26; Fe = 24; Ga = 43. Natural and synthetic standards were used and the PAP program (Pouchou and Pichoir, 1991).

Oxygen isotope analyses of corundum were performed using a modification of the laser-fluorination technique (Giuliani et al., 2005). The analyzed samples were 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  $\sim$ 1 mg of powdered corundum, heated by a CO<sub>2</sub> laser, with ClF<sub>3</sub> as the fluorine reagent. The released oxygen is passed through an in-line Hg-diffusion pump before conversion to CO<sub>2</sub> 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 per mg were taken as likely evidence of analytical artifact. Precision and accuracy on quartz standards are ±0.1‰  $(1\sigma)$ , 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).  $\delta^{18}O$  (%) = [(( $R_{sample}/R_{standard}) - 1$ )] × 10<sup>3</sup>, where *R* is the isotopic ratio  ${}^{18}O/{}^{16}O$ .

Zircon grains for U–Pb dating were imaged by back-scattered electrons (BSE) with a Hitachi 2500 SEM. The excimer laser ablation (ELA)-ICPMS laser on zircon grains were performed at the University of Pavia (Italy) by J.-L. Paquette (within the PGRM program from Madagascar). The analytical procedure for ICPMS laser ablation is detailed in Tiepolo (2003). Instrumental and laser-induced U/Pb fractionations were corrected using the 91500 zircon standard. The spot size was set to 20 or 10 µm and laser fluence to 12 J/cm<sup>2</sup>.

### Appendix B. Supplementary material

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.jafrearsci. 2013.06.003.

#### References

Abduriyim, A., Kitawaki, H., 2006. Applications of laser ablation-inductively coupled plasma-mass spectrometry (LA-ICP-MS) to gemmology 42, 98–118.

Alt, J.C., 1995. Subseafloor processes in mid-ocean ridge hydrothermal systems. In: Zierenberg, R.A., Humphris, S.E., Mullineaux, L.S., Thomson, R.E. (Eds.), Seafloor Hydrothermal Systems: Physical, Biological Interactions. AGU, WA, pp. 85–114.

- Ashwal, L.D., Burke, K., 1989. African lithospheric structure, volcanism and topography. Earth and Planetary Science Letters 96, 8–14.
- Aspen, P., Upton, B.G.J., Dickin, A.P., 1990. Anorthoclase, sanidine and associated megacrysts in Scottish alkali-basalts: high-pressure syenitic debris from upper mantle sources. European Journal of Mineralogy 2, 503–517.
- Bardintzeff, J.-M., Liégeois, J.-P., Bonin, B., Bellon, H., Rasamimanana, G., 2010. Madagascar volcanic provinces linked to the Gondwana break-up: geochemical and isotopic evidences for contrasting mantle sources. Gondwana Research 18, 295–314.
- Besairie, H., 1971. Mise au point sur le volcanisme de l'Ankaratra (Madagascar). Archives Service Géologique de Madagascar, N°2169, 19 p.
- Besairie, H., Brenon, P., Boulanger, J., Emberger, A., De Saint Ours, J., 1957. Le volcanisme à Madagascar. Travaux Bureau Géologique de Madagascar, N°83, 237 p.
- BGS-USGS-GLW, 2008. Révision de le cartographie géologique et minière des zones Nord, Centre et Centre-est de Madagascar (Zones A, B et D). République de Madagascar, Ministère de l'Energie et des Mines (MEM/SG/DG/UCP/PGRM). NERC 2008, Keyworth, Nottingham, 1049 p.
- Brousse, R., Varet, J., 1966. Les trachytes du Mont Dore et du Cantal septentrional et leurs enclaves. Bulletin Société Géologique de France VII (7), 246–262.
- Chorowicz, J., le Fournier, J., Vidal, G., 1987. A model for rift development in eastern Africa. Geological Journal 22, 495–513.
- Class, C., Goldstein, S.L., 1997. Plume–lithosphere interactions in the ocean basins: constraints from the source mineralogy. Earth and Planetary Science Letters 150, 245–260.
- Class, C., Goldstein, S.L., Altherr, R., Bachelery, P., 1998. The process of plumelithosphere interaction in the ocean basins: the case of Grande Comore. Journal of Petrology 39, 881–903.
- Cucciniello, C., Melluso, L., Morra, V., Storey, M., Rocco, I., Franciosi, L., Grifa, C., Petrone, C.M., Vincent, M., 2011. New <sup>40</sup>Ar-<sup>39</sup>Ar ages and petrogenesis of the Massif d'Ambre volcano, northern Madagascar. Geological Society of America Special Papers 478, 257–281.
- de Wit, M.J., 2003. Madagascar: heads it's a continent tails it's an island. Annual Review of Earth Planetary and Science 31, 213–248.
- Emerick, C.M., Duncan, R.A., 1982. Age progressive volcanism in the Comores Archipelago, western Indian Ocean and implications for Somali plate tectonics. Earth and Planetary Science Letters 60, 415–428.
- Gaillou, E., 2003. Les saphirs du Massif Central: étude minéralogique des saphirs du Sioulot, du Mont Coupet et de Menoyre. Détermination de leur origine. Mémoire de DEA, Université Blaise Pascal, Clermont-Ferrand, France, 45 p.
- Gaillou, E., Devouard, B., Vielzeuf, D., Boivin, P., Rochalut, J., Valley, J., Harris, C., 2010. Le Mont Coupet, Menet et le Sioulot: trois gisements de saphirs du massif central. le Règne Minéral 93, 28–36.
- Garnier, V., Giuliani, G., Ohnenstetter, D., Schwarz, D., 2004. Saphirs et rubis. Classification des gisements de corindons. le Règne Minéral 55, 4–47.
- Giuliani, G., Fallick, A.E., Garnier, V., France-Lanord, Ch., Ohnenstetter, D., Schwarz, D., 2005. Oxygen isotope composition as a tracer for the origin of rubies and sapphires. Geology 33, 249–252.
- Giuliani, G., Fallick, A.E., Rakotondrazafy, A.F.M., Ohnenstetter, D., Andriamamonjy, A., Ralantoarison, Th., Rakotosamizanany, S., Razanatseheno, M., Offant, Y., Garnier, V., Dunaigre, Ch., Schwarz, D., Mercier, A., Ratrimo, V., Ralison, B., 2007a. Oxygen isotope systematics of gem corundum deposits in Madagascar: relevance for their geological origin. Mineralium Deposita 42, 251–270.
- Giuliani, G., Ohnenstetter, D., Garnier, V., Fallick, A.E., Rakotondrazafy, A.F.M., Schwarz, D., 2007b. The geology and genesis of gem corundum deposits. In: Raeside, E.R. (Ed.), Geology of Gem Deposits, Mineralogical Association of Canada, Short Course Series 37. Yellowknife, Canada, pp. 23–78.
- Giuliani, G., Fallick, A.E., Ohnenstetter, D., Pegere, G., 2009. Oxygen isotopes of sapphire from the French Massif Central: implications for the origin of gem corundum in basaltic fields. Mineralium Deposita 44, 221–231.
- Giuliani, G., Lasnier, B., Ohnenstetter, D., Fallick, A.E., Pegère, G., 2010. Les gisements de corindons de France. le Règne Minéral 93, 28–36.
- Giuliani, G., Ohnenstetter, D., Fallick, A.E., Groat, L., Feneyrol, J., 2012. Geographic origin of gems linked to their geographical history. In Color 19, 16–27.
- Graham, I., Sutherland, L., Zaw, K., Nechaev, V., Khanchuk, A., 2008. Advances in our understanding of the gem corundum deposits of the West Pacific continental margins intraplate basaltic fields. Ore Geology Reviews 34, 200–215.
- Guo, J., O'Reilly, J., Griffin, W.L., 1996a. Corundum from basaltic terrains: a mineral inclusion approach. Contributions to Mineralogy and Petrology 122, 368–386.
- Guo, J., O'Reilly, J., Griffin, W.L., 1996b. Zircon inclusions in corundum megacrysts: I. Trace element geochemistry and clues to the origin of corundum megacrysts in alkali-basalts. Geochimica et Cosmochimica Acta 60, 2347–2363.
- Hirschmann, M.M., Kosigo, T., Baker, M.B., Stolper, E.M., 2003. Alkalic magmas generated by partial melting of garnet pyroxenite. Geology 31, 481–484.
- Hong-sen, X., Wei-Guo, Z., Wei, H., Wen-ge, Z., Jie, G., Ji-an, X., 2002. Discovery of corundum in alkali basalt at high pressure and high temperature. Journal of Physics: Condensed Matter 14, 11365–11368.
- Karche, J.P., 1973. Le massif volcanique d'Ambre et les régions voisines du Nord de Madagascar. Etude volcanologique et pétrologique. Thèse Annales Sciences de l'Université de Besançon, France, 19, 1, 173 p.
- Khin Zaw, Sutherland, F.L., Dellapasqua, F., Ryan, C.G., Yui, T.Z., Mernagh, T.P., Duncan, D., 2006. Contrasts in gem corundum characteristics, eastern Australian basaltic fields: trace elements, fluid/melt inclusions and oxygen isotopes. Mineralogical Magazine 70, 669–687.

- Kornprobst, J., Piboule, M., Roden, M., Tabit, A., 1990. Corundum-bearing garnet clinopyroxenites at Beni Bousera (Morocco): original plagioclase-rich gabbros recrystallized at depth within the mantle? Journal of Petrology 31, 717–745.
- Kusky, T.M., Toraman, E., Raharimahefa, T., 2006. The great rift valley of Madagascar: an extension of the Africa–Somalia diffusive plate boundary ? Gondwana Research 11, 577–579.
- Lagabrielle, Y., Bodinier, J.-L., 2008. Submarine reworking of exhumed subcontinental mantle rocks: field evidence from the Lherz peridotites, French Pyrenées. Terra Nova 20, 11–21.
- Laville, E., Piqué, A., Plaziat, J.-C., Gioan, P., Rakotomalala, R., Tidahy, E., 1998. Le fossé d'Ankay-Aloatra, témoin d'une extension crustale récente et actuelle à Madagascar. Bulletin de la Société Géologique de France 169, 775–788.
- Mattey, D., Lowry, D., Macpherson, C.G., 1994. Oxygen isotope composition of mantle peridotite. Earth and Planetary Science Letters 128, 231–241.
- Melluso, L., Morra, V., 2000. Petrogenesis of Late Cenozoic mafic alkaline rocks of the Nosy Be archipelago (northern Madagascar): relationships with the Comorean magmatism. Journal of Volcanology and Geothermal Research 96, 129–142.
- Melluso, L., Morra, V., Brotzu, P., Razafiniparany, A., Ratrimo, V., Razafimahatratra, D., 1997. Geochemistry and Sr-isotopic composition of the late Cretaceous flood basalt sequence of northern Madagascar: petrogenic and geodynamic implications. Journal of African Earth Sciences 24, 371–390.
- Melluso, L., le Roex, A.P., Morra, V., 2011. Petrogenesis and Nd-, Pb-, Sr-isotope geochemistry of the Cenozoic olivine melilities and olivine nephelinites ("ankaratrites") in Madagascar. Lithos 127, 505–521.
- Merle, O., Michon, L., Camus, G., De Goer, A., 1998. L'extension Oligocène sur la transversale septentrionale du rift du Massif Central. Bulletin de la Société Géologique de France 169, 615–626.
- Monchoux, P., Fontan, F., De Parseval, Ph., Martin, R.F., Wang, R.C., 2006. Igneous albitite dikes in orogenic lherzolites, western Pyrénées, France: a possible source for corundum and alkali feldspar xenocrysts in basaltic terranes. I. Mineralogical associations. Canadian Mineralogist 44, 811–836.
- Mottet, G., 1981. L'Ankaratra et ses bordures. Thèse Université de Lyon III, 730 p.
- Muntener, O., Hermann, J., 1996. The Val Malenco lower crustal mantle complex and its field relations (Italian Alps). Schweizer Mineralogische und Petrographische Mitteilungen 76, 475–500.
- Nicollet, C., 1984. Le volcanisme dans le Sud Ouest de Madagascar. Journal of African Earth Sciences 2, 383–388.
- Nicollet, C., 1986. Saphirine et staurotide riche en magnésium et chrome dans les amphibolites et anorthosites à corindon du Vohibory Sud, Madagascar. Bulletin de Minéralogie 109, 599–612.
- Nicollet, C., 1988. Métabasites granulitiques, anorthosites et roches associées de la croûte inférieure. Exemples pris à Madagascar et dans le Massif Central Français. Arguments d'un métamorphisme associé à l'extension lithosphérique. Thèse de Doctorat d'Etat, Université Blaise Pascal. Clermont-Ferrand, France, 315 p.
- Nicollet, C., 1990. Crustal evolution of the granulites of Madagascar. In: Vielzeuf, D., Vidal, Ph. (Eds.), Granulites and Crustal Evolution. Kluwer, Dorchet, pp. 291– 310.
- Nougier, J., Cantagrel, J.M., Karche, J.-P., 1986. The Comores archipelago in the western Indian Ocean/volcanology, geochronology and geodynamic setting. Journal of African Earth Sciences 5, 135–145.
- Paquette, J.-L., Nédélec, A., 1998. A new insight into Pan-African tectonics in the East–West Gondwana collision zone by U–Pb zircon dating of granites from central Madagascar. Earth Planetary and Science Letters 155, 45–56.
- Paquette, J.-L., Nédélec, A., Moine, B., Rakotondrazafy, A.F.M., 1994. U–Pb single zircon Pb-evaporation and Sm–Nd isotopic study of a granulite domain in SE Madagascar. Journal of Geology 102, 523–538.
- Pearson, D.G., Davies, G.R., Nixon, P.H., Greenwood, P.B., Mattey, D.P., 1993. Oxygen isotope evidence for the origin of pyroxenites in the Beni Bousera peridotite massif, North Morocco: derivation from subducted oceanic lithosphere. Earth and Planetary Science Letters 102, 289–301.
- Peucat, J.J., Ruffault, P., Fritsch, E., Bouhnik-Le Coz, M., Simonet, C., Lasnier, B., 2007. Ga/Mg ratios as a new geochemical tool to differentiate magmatic from metamorphic blue sapphires. Lithos 98, 261–274.
- Pham, V.L., Hoang, Q.V., Garnier, V., Giuliani, G., Ohnenstetter, D., Lhomme, Th., Schwarz, D., Fallick, A.E., Dubessy, J., Phan, T.T., 2004. Gem corundum deposits in Vietnam. Journal of Gemmology 29, 129–147.
- Pin, C., Monchoux, P., Paquette, J.-L., Azambre, B., Wang, R.C., Martin, R.F., 2006. Igneous albitite dikes in orogenic lherzolites, western Pyrénées, France: a possible source for corundum and alkali feldspar xenocrysts in basaltic terranes. II. Geochemical and petrogenetic considerations. Canadian Mineralogist 44, 837–850.
- Piqué, A., Laville, E., Chotin, P., Chorowicz, J., Rakotondraompiana, S., Thouin, C., 1999. L'extension à Madagascar du Néogène à l'actuel: arguments structuraux et géophysiques. Journal of African Earth Sciences 28, 975–983.
- Pouchou, J.-L., Pichoir, F., 1991. Quantitative analysis of homogenous or stratified microvolumes applying the model "PAP". In: Kerrich, K.F.J., Newbury, D.E. (Eds.), Electron Probe Quantification. Plenum Press, New York, pp. 31–75.
- Rakotondraompiana, S., Albouy, Y., Piqué, A., 1999. Modèle de lithosphère pour l'île de Madagascar (océan indien occidental): nouvelle interprétation des données gravimétriques. Journal of African Earth Sciences 28, 961–973.
- Rakotondrazafy, M., Moine, B., Cuney, M., 1996. Mode of formation of hibonite (CaAl<sub>12</sub>O<sub>19</sub>) within the U–Th skarns from the granulites of the S–E Madagascar. Contributions to Mineralogy and Petrology 123, 190–201.

- Rakotondrazafy, A.F.M., Giuliani, G., Fallick, A.E., Ohnenstetter, D., Andriamamonjy, A., Rakotosamizanany, S., Ralantoarison, Th., Razatseheno, M., Offant, Y., Garnier, V., Maluski, H., Dunaigre, C., Schwarz, D., Mercier, A., Ratrimo, V., Ralison, N., 2008. Gem corundum deposits in Madagascar: a review. Ore Geology Reviews 34, 134–154.
- Rakotosamizanany, S., 2009. Les corindons gemmes dans les basaltes alcalins et leurs enclaves à Madagascar: significations pétrologique et métallogénique. Thèse de Doctorat, Université Henri Poincaré, Nancy I, France, 414 p.
- Ralantoarison, Th., Offant, Y., Giuliani, G., Rakotondrazafy, A.F.M., Schwarz, D., Ohnenstetter, D., Fallick, A.E., Razatseheno, M., Rakotosamizanany, S., Moine, B., Baillot, P., 2006. Les saphirs multicolores de Sahambano et Zazafotsy, région granulitique d'Ihosy, Madagascar. Revue de l'Association Française de Gemmologie 158, 4–13.
- Rasimanana, G., 1996. Caractérisations géochimiques et géochronologiques de trois épisodes magmatiques (Crétacé, Miocène terminal et Quaternaire) à Madagascar liés aux phénomènes de rifting. Thèse de Doctorat, Université d'Orsay, Paris, France, 234 p.
- Saminpanya, S., 2000. Mineralogy and Origin of Gem Corundum Associated with Basalt in Thailand. PhD Thesis, University of Manchester, Great Britain, 395 p.
- Saminpanya, S., Sutherland, F.L., 2011. Different origins of Thai area sapphire and ruby derived from mineral inclusions and co-existing minerals. European Journal of Mineralogy 23, 683–694.
- Schwarz, D., Schmetzer, K., 2001. Rubies from the Vatomandry area, eastern Madagascar. Journal of Gemmology 27, 409–416.
- Schwarz, D., Kanis, J., Schmetzer, K., 2000. Sapphires from Antsiranana Province, Northern Madagascar. Gems & Gemology 36, 216–233.
- Seifert, A., Hyrsl, J., 1999. Sapphire and garnet from Kalalani, Tanga Province, Tanzania. Gems & Gemology 35, 108–120.
- Simonet, C., 2000. La géologie des gisements de saphir et de rubis. L'exemple de la John Saul Ruby Mine, Mangare, Kenya. Thèse de Doctorat, Université de Nantes, France, 349 p.
- Simonet, C., Fritsch, E., Lasnier, B., 2008. A classification of gem corundum deposits aimed towards gem exploration. Ore Geology Reviews 34, 127–133.
- Sutherland, F.L., Abduriyim, A., 2009. Geographic typing of gem corundum: a test case from Australia. Journal of Gemmology 31, 203–210.
- Sutherland, F.L., 1996. Alkaline rocks and gemstones, Australia. Australian Journal of Earth Sciences 43, 323–343.
- Sutherland, F.L., Coenraads, R.R., 1996. An unusual ruby-sapphire-sapphirinespinel assemblage from the Tertiary Barrington volcanic Province, New South Wales, Australia. Mineralogical Magazine 60, 623–638.
- Sutherland, F.L., Schwarz, D., 2001. Origin of gem corundums from basaltic fields. Australian Gemmologist 21, 30–33.
- Sutherland, F.L., Hoskin, P.W.O., Fanning, C.M., Coenraads, R.R., 1998a. Models of corundum origin from alkali-basaltic terrains: a reappraisal. Contributions to Mineralogy and Petrology 133, 356–372.

- Sutherland, F.L., Schwarz, D., Jobbins, E.A., Coenraads, R.R., Graham, I.T., Webb, G., 1998b. Distinctive gem corundum suites from discrete basalt fields: a comparative study of Barrington, Australia, and West Pailin, Cambodia, gemfields. Journal of Gemmology 26, 65–85.
- Sutherland, F.L., Coenraads, R.R., Schwarz, D., Raynor, L.R., Barron, B.J., Webb, G.B., 2003. Al-rich diopside in alluvial ruby and corundum-bearing xenoliths, Australian and SE Asian basalt fields. Mineralogical Magazine 67, 717–732.
- Sutherland, F.L., Khin Zaw, Meffre, S., Giuliani, G., Fallick, A.E., Webb, G.B., 2009. Gem-corundum megacrysts from east Australian basalt fields: trace elements, oxygen isotopes and origins. Australian Journal of Earth Sciences 56, 1003– 1022.
- Sutthirat, C., Saminpanya, S., Droop, G.T.R., Henderson, C.M.B., Manning, D.A.C., 2001. Clinopyroxene-corundum assemblages from alkali-basalt and alluvium, eastern Thailand: constraints on the origin of Thai rubies. Mineralogical Magazine 63, 277–295.
- Tiepolo, M., 2003. In situ Pb geochronology of zircon with laser ablation-inductively coupled plasma-sector field mass spectrometry. Chemical Geology 199, 159–177.
- Tucker, R., Conrad, J., Key, R., Pitfield, P., Randriamananjara, Th. Taylor, C., Goodenough, K., Thomas, B., 2008. Géochronologie 40Ar/39Ar des roches ignées du Mésozoïque et ceux d'âge plus jeunes. in: BGS-USCS-GLW, 2008. Révision de le cartographie géologique et minière des zones Nord, Centre et Centre-est de Madagascar (Zones A, B et D). République de Madagascar, Ministère de l'Energie et des Mines (MEM/SG/DG/UCP/PGRM). NERC 2008, Keyworth, Nottingham, pp. 385–439.
- Hey Word, Hotting, Karl, P. 505 152.
  Uher, P., Giuliani, G., Szakall, S., Fallick, A.E., Strunga, V., Vaculovic, D., Ozdin, D., Greganova, M., 2012. Sapphires related to alkali basalts from the Cerová Highlands, Western Carpathians (southern Slovakia): composition and origin. Geologica Carpathica 63, 71–82.
- Upton, B.G.J., Hinton, R.W., Aspen, P., Finch, A.A., Valley, J.W., 1999. Megacrysts and associated xenoliths: evidence for migration of geochemically enriched melts in the upper mantle beneath Scotland. Journal of Petrology 40, 935–956.
- Upton, B.G.J., Finch, A.A., Slaby, E., 2009. Megacryst and salic xenoliths in Scottish alkali basalts: derivatives of deep crustal intrusions and small-melt fractions from the upper mantle. Mineralogical Magazine 73, 943–956.
- Yager, R.Y., Menzie, W.D., Olson, D.W., 2008. Weight of Production of Emeralds, Rubies, Sapphires, and Tanzanite from 1995 through 2005. Open File Report 2008–1013. USGS Reston, Virginia, USA, 9 p.
- Yui, T.F., Zaw, K., Limkatrum, P., 2003. Oxygen isotope composition of the Denchai sapphire, Thailand: a clue to its enigmatic origin. Lithos 67, 153–161.
- Yui, T.F., Wu, C.M., Limkatrum, P., Sricharn, W., Boonsoong, A., 2006. Oxygen isotope studies on placer sapphire and ruby in the Chanthaburi-Trat alkali-basaltic gemfield, Thailand. Lithos 86, 197–211.