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PETROLOGY, SR AND ND ISOTOPE SIGNATURE OF THE LATE CRETACEOUS MAGMATISM IN THE SOUTH-EASTERN PART OF ETROPOLE STARA PLANINA, SREDNOGORIE MAGMATIC ZONE

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ABSTRACT
The Chelopech volcano is the host of one of the largest Au-Cu deposits in Europe. It includes three phases: dome-like bodies (andesites and latites to trachydacites), lava to agglomerate flows (andesites, latites, dacites to trachydacites), a lava breccia neck (andesites to shoshonites and latites) and dykes (andesites to dacites). The age of those magmatic products is probably Turonian. The volcanic rocks are porphyric with plagioclase and amphibole phenocrysts, rarely quartz (in the dome-like bodies and dykes) and biotite. The groundmass is microlitic. The lava flows contain fully crystallized fine-grained inclusions with more basic compositions indicating mingling between two parental magmas. The chemical evolution from more acid to more basic lavas, and the absence of an Eu anomaly probably indicate a chemically zoned magmatic chamber. The trace element content is similar to that of the active continental margin (Andean type). Sr isotopic compositions display a small range between 0.7049 and 0.7054 (corrected for 90 Ma) and Nd rations are from 0.5124 to 0.5125 (corrected for 90 Ma).

INTRODUCTION

The Chelopech volcanics and dykes outcrop in the southeastern part of Etropole Stara planina. They are part of Late Cretaceous Central Srednogorie volcano-intrusive area. The Chelopech volcano is the host of one of the largest Au-Cu deposits in Europe, containing well in excess of 5.5 million ounces of Au and >10 million ounces Au equivalent (Andrew, 1997). It is situated about 65 km east of Sofia at the foot of the Stara Planina Mountain. This deposit has been an object of many investigations connected to its geology and structures (Popov and Mutafchiev, 1980; Popov et al., 2000, 2002), hydrothermal alteration (Mutafchiev and Chipchakova, 1969), mineralogy, stratigraphy of the Upper Cretaceous sequences (Moev and Antonov, 1978a; Dimitrova et al., 1984), structures in the region (Moev and Antonov, 1978b; Popov et al., 2000, 2002), and radiogenic age (Lilov and Chipchakova, 1999; Velichkova et al., 2001), because of its large economical interest. The petrographic and age characteristics of the surrounding area of the Chelopech deposit have received less attention (Mutafchiev and Chipchakova, 1969; Moev and Antonov, 1978a; Stoykov et al, 2002; Stoykov and Pavlishina, 2003). The aim of present paper is to complete this information and to show new data about the geological, petrochemical, Sr and Nd isotope, mineralogical and age characteristics of the magmatic rocks, pert of the Srednogorie magmatic zone.

GEOLOGICAL SETTING OF STUDIED MAGMATIC ROCKS

The region of the Chelopech volcano (Fig. 1) is built up by metamorphic basement rocks and a Late Cretaceous volcanic and sedimentary rock succession. The basement appears in the northeastern part of the region and it is composed by the metamorphic rocks of the Pirdop and the Bercovitza Groups in tectonic contact with each other. The Pirdop Group consists of two-mica migmatites with thin intercalations of amphibolites, hornblende-biotite and biotite gneisses (Dabovski, 1988). The Bercovitza Group is a Late Precambrian-Cambrian sedimentary-volcanic complex of island-arc association (Haydoutov, 2001). It consists of equal parts of sedimentary and volcanic rocks (spilites, keratophyres and their pyroclastic rocks) metamorphosed under greenschist facies conditions. Late Cretaceous (Turonian - Maastrichtian) sedimentary and volcanic rocks, more than 2000 m in thickness transgressively overlie this basement. The metamorphics is also cut by east-west oriented andesitic, latitic, dacitic to trochydacitic dykes.

The Late Cretaceous sedimentation starts with conglomerates and coarse sandstones with coal-bearing interbeds (Coal-bearing formation, according to Moev and Antonov, 1978a) covered by polymictic, argilleous and arcose sandstones to siltstones (Sandstone formation) with up to 500 m thick. Both formations are probably of Turonian age (Nikolaev, 1947; Moev and Antonov, 1978a) as confirmed by the new pollen data of Stoykov and Pavlishina (2003). These sedimentary rocks are cut by volcanic bodies and overlaid by the sedimentary and volçanic rocks of the Chelopech Formation according to Moev and Antonov (1978a) or the Tuff formation according to Dimitrova et al. (1984). The products of the Chelopech volcano form the Vozdol member of this Formation. After the Subhercinian tectonic deformations (Popov et al., 2002) the rocks of this units have been eroded and transgressively covered by the sedimentary rocks of the Mirkovo Formation - reddish limestones and marls (Moev and Antonov, 1978a) or the limestone-marls formation after Dimitrova et al. (1984). They are covered by the flysch of the Chugovo Formation (Moev and Antonov, 1978a) or the flysch formation after Dimitrova et al. (1984). The rocks of the last two Formations build up the Chelopech syncline (Moev and Antonov, 1978b). The size of this structure is 10 × 2 km. The volcanic rocks preserved by erosion form the limbs of this syncline that is cut and covered in the eastern part (Fig. 1) by the Chelopech thrust (Moev and Antonov, 1978b). The last structure is recovered by the Neogene-Quaternary Zlatitsa graben.

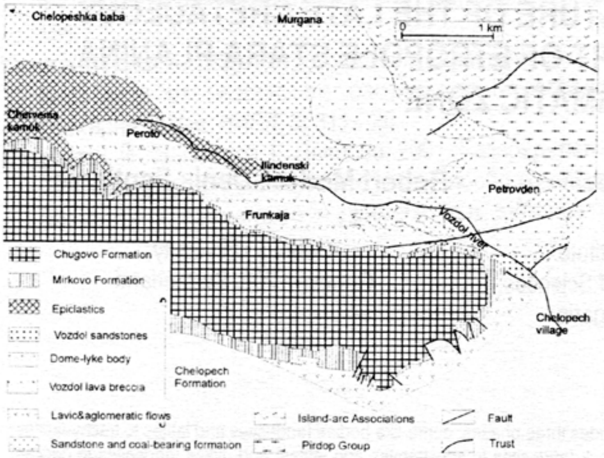

Figure 1. Geological map of the Southeastern part of Etropole Stara planina

The geophysical data show the presence of a positive anomaly 20 km in diameter, which is located between the studied magmatic rocks and the Elatsite pluton in the north (Popov et al., 2002). This magnetic anomaly is interpreted as a large magnetic-active body corresponding to a shallow magmatic chamber. These authors proposed that the Chelopech volcano and the Elatsite pluton are part of the same volcano-plutonic complex and one ore-magmatic system.

GEOLOGICAL STRUCTURE OF THE CHELOPECH VOLCANO

The basement of the Chelopech volcano

It is not exposed on the surface, but is cut by the boreholes in the underground mine of Chelopech. It is composed by the rocks of the Sandstone formation, with a thickness between 300 and 450 m (Moev and Antonov, 1978a). According to Popov et al. (2002), in the northern part of the Vozdol river, the basement of the volcano is built up by an olistrostrome unit with a limited development according to borehole data. These data can be interpreted in terms of blocks of metamorphic basement with a sedimentary rock cover, cut by volcanic bodies.

The Chelopech volcano

It (stratovolcano according to Popov et al., 2002) consists of 3 phases: (i) dome-like volcanic bodies, (ii) lava and agglomerate flows and (iii) a neck, locally known as the Vozdol neck (Popov et al., 2002).

Dome-like volcanic bodies. In the Murgana area (Fig. 1) the dome-like volcanic bodies are exposed on the surface without clear relationships with the products of the next phase. These bodies are intruded in the Turonian sediments where the bedding of the hosting rocks close to their contact is subvertical (e.g. in the Belishka river). The largest body is about 2 × 1 km in size. It has a complicated morphology probably reflecting its composite character. Some parts of the bodies (to the south of the Murgana summit) have a dome-like morphology (according to the data of Moev and Antonov, 1978a), corresponding to their petrographic characteristics (see below). Popov and Mutafchiev (1980) described these bodies as subvolcanic, and later, as subvolcanic intrusion (Popov et al., 2000). These authors distinguished an early and a late group of subvolcanic bodies. Lilov and Chipchakova (1999) attributed a 65-67 Ma age according to K-Ar dating of some of the bodies, which probably reflects a younger, overprinting geological event (see below).

The lava flows grade into agglomerate flows (with fragments up to about 30 cm in size) in the upper levels. Subvertical columnar jointing is observed in the lava flows in some places (e.g. in the Ilindenska river). The total thickness of these volcanic products is up to 1200 m according to the drilling data (Popov et al., 2002). K-Ar data of non-altered andesite indicate a Turonian age (91 Ma according to Lilov and Chipchakova, 1999) and U-Pb zircon dating of andesite overprinted by alteration and mineralization in the mine $^{206}Pb/^{238}U$ age is 91.45 ± 0.15 Ma (Moritz et al., 2003).

The location of the volcanic center is not clear. It is probably situated in the area of the Chelopech deposit (respectively in the area of the Chugovo river) where two boreholes cut a very thick volcanic succession (1700-2000 m). The other boreholes in the deposit cut a 700-800 m thick succession of volcanic rocks only. This difference in thickness is too large to be connected to a caldera subsidence. There are also no geological and geophysical evidences for concentric faults related to caldera subsidence, as proposed by Popov et al. (2000, 2002). There are also volcanic breccia and tuffs in the deposit (Mutafchiev and Chipchakova, 1969; Popov and Mutafchiev, 1980). They are strongly hydrothermally altered rocks and probably more of them are epiclastic rocks.

In the western part of the volcano, nearby the Chervenia Kamak summit the upper levels of the agglomerate flows are intercalated with psephitic and psamitic epiclastic rocks, the latter are interbedded with the sandstones and marls of the Chelopech Formation.

The Vozdol neck. In the eastern part of the Vozdol valley (Fig. 1), to the northeast of the Petrovden fault a volcanic breccia is outcropping with a surface of 1.5 × 0.250 km. It is interpreted as the youngest neck of the Chelopech volcano, and is called Vozdol monovolcano by Popov et al. (2000, 2002). One $^{40}Ar/^{39}Ar$ age of biotite from this breccia gives a Turonian age of about 90 Ma (Velichkova et al., 2001). The former K-Ar age of 65 Ma obtained by Lilov and Chipchakova (1999) for samples from the same locality likely represents the age of a younger overprinting thermal event than the real magmatic crystallization age of the Vozdol volcanics. The Vozdol neck consists of clasts-supported lava-breccia with 20 to about 80 cm-sized fragments in a lavic matrix. In the eastern periphery of the body, sedimentary material occurs in the matrix (sandstones to gravelites), which increases volumetrically to the border of the body, where they form a small lens and layers. These features show sedimentation during the formation of this volcanic body and the beginning of its destruction and redeposition in the younger sandstones of the Vozdol area.

The cover of the Chelopech volcano

It is represented by the Vozdol sandstones (in the eastern part), the muddy limestones of the Mirkovo Formation (in the central part) and the sedimentary rocks of the Chelopech Formation (in the western part).

The Vozdol sandstones, which have not been described as a single litostratigraphic unit in previous contributions, are only locally developed. They are exposed on a surface of about 2.5 × 1 km and are partly covered by the Chelopech syncline. These sandstones have a variable thickness, with the largest one (up to 250 m) being located in the syncline and on the Vozdol river. They are probably of fluvial or coastal origin (Stoykov and Pavlishina, 2003) and of Turonian age (Nikolaev, 1947) confirmed by the new pollen data of Stoykov and Pavlishina (2003). The sandstones are coarse, thick bedded, and they show cross-bedding. Small coal lenses are present and two conglomerate layers can be recognized (described previously as tuff layers by Moev and Antonov, 1978a, and Popov and Mutafchiev, 1980) with fragments of different volcanic rocks (including from the Vozdol neck) and variable sizes up to 1 m. They can be interpreted as products of mud flows. In comparison to the sandstones of the Chelopech Formation, they also contain muscovite which corresponds to another source of terrigeneous material probably derived from the Pirdop Group to the north.

The partly eroded Chelopech volcano (in the central part of the region) and the Vozdol sandstones (in the eastern part of the region) are transgressively covered by reddish clayey limestones of the Mirkovo Formation (Moev and Antonov, 1978a). These limestones, with a thickness up to 30-40 m, comprise fragments up to 25 cm in size of different volcanic rocks and the Vozdol sandstones. Calcareous nannofossils from the limestones, mostly in the base of this sedimentary unit, indicate a Latest Santonian to Campanian age (unpublished data of K. Stoykova, Geological Institute). They are concordantly covered by flysch sedimentary rocks of the Chugovo Formation (Late Campanian - Early Maastrichtian according to K. Stoykova). The latter consist of an interbedding of calcareous sandstone, siltstone and argillite with a thickness up to 500 m. Volcanoclastic layers are not present in the region of the Chelopech volcano, which is in contrast with other parts of the Central Srednogorie area (Velichkova et al., 2002). The sedimentary rocks of these two formations form the Chelopech syncline.

The dykes

They have predominately east-west direction and are intruded into pre-upper Cretaceous basement rocks of the Bercovitza and the Pirdop Group without clear relationships with the products of the Chelopech volcano are not clear. The largest one is more than 7 km in length.

PETROLOGY OF THE STUDIED MAGMATIC ROCKS

Methods

The major and trace elements were analyzed by X-ray fluorescence (XRF) at the University of Lausanne (Switzerland). The rare earth elements (REE) were analysed by ICP-atomic emission spectrometry following the procedure of Voldet (1993). The representative analyses of the compositional variation of the rock recovered from the Chelopech volcanics are given in Tables 1 and 2. Trace elements (Table 2) were analyzed also by XRF at the University of Geneva. The petrological study was carried out on fresh samples. Mineral analyses on 10 samples of the different phases were carried out at University of Lausanne (Switzerland) on a CAMEBAX SX-50 electron microprobe.

Petrography

The volcanic rocks are shoshonites, andesites, latites to dacites and trachydacites (Fig. 2).

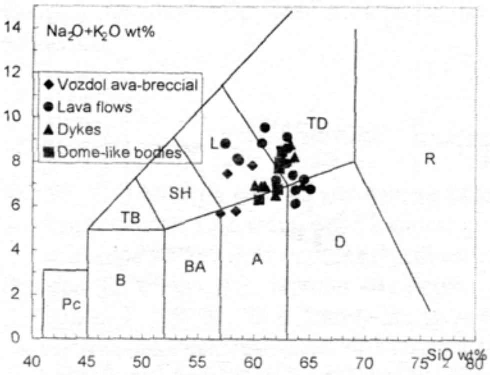

Figure 2. TAS diagram after Le Maitre (1989) for representative magmatic rocks from the studied region (B, basalt; BA, basaltic andesite; A, andesite; D, dacite; SH, shoshonite; L, latite; TD, trachidacyte)

In the Chelopech volcano the magma evolved from more acid volcanic rocks with 61-64 wt% SiO_2 of the earlier products (lava and agglomerate flows and dome-like bodies) to the more basic ones with 55.5-58 wt% SiO_2 of the Vozdol volcanic rocks (Table 1).

Table 1. Major element composition of the representative volcanic samples

Oxides wt. %	Dome-like body	Lava flows	Vozdol breccias	Dykes
SiO_2	61.22	63.01	57.11	60.07
TiO_2	0.54	0.51	0.65	0.47
Al_2O_3	17.98	16.36	18.35	16.46
Fe_2O_3	5.01	4.94	7.03	4.04
MnO	0.14	0.12	0.12	0.2
MgO	1.44	1.63	1.75	1.61
CaO	3.38	4.91	4.87	5.34
Na_2O	5.32	3.39	4.19	3.69
K_2O	2.70	2.74	3.27	3.2
P_2O_5	0.25	0.23	0.26	0.2
LOI	1.73	1.16	1.55	3.71
Total	99.71	99.00	99.15	98.99

The dome-like bodies are porphyric with a microlitic groundmass and an andesitic, latitic to trachydacitic chemistry. These volcanic rocks consist of plagioclase, zoned amphibole, minor biotite, quartz and titanite as phenocrysts, and microlites are presented by the same minerals.

The composition of the lava flows is mostly latitic. Subsidiary andesites, dacites and trachydacites are also present in minor amount too. These volcanic rocks are highly porphyric with microlitic groundmass. The phenocrysts (> 40 volume %) consist of plagioclase, zoned amphibole, minor biotite, and titanite; whereas the microlites consist of plagioclase and amphibole only. The accessory minerals are apatite, zircon, and Ti-magnetite. The lava flows contain fine-grained, fully crystallized inclusions consisting of the same minerals (plagioclase, amphibole and minor biotite), which comprise phenocrysts of different chemistry. The margins of the inclusions are marked by fine-grained quartz zone, which is interpreted as evidence of magma mingling.

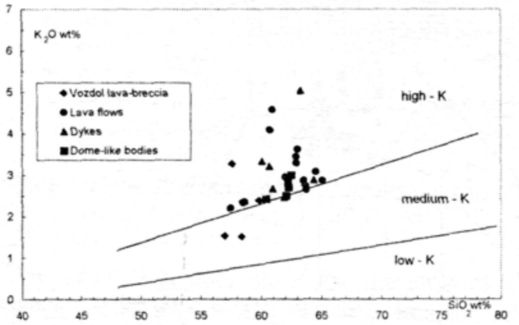

Figure 3. SiO_2 vs. K_2O diagram after Le Maitre (1989) for representative Chelopech volcanic rocks

The Vozdol andesites and latites to shoshonites display similar petrographic characteristics but their phenocrysts (plagioclase, amphibole, minor biotite, and titanite) are less abundant compared to the other magmatic rocks of the Chelopech volcano. The groundmass is composed of the microlites of the same minerals. K-feldspar is present as microlites only in the Vozdol andesitic rocks.

The composition of the dykes is andesitic, latitic to dacitic and trachydacitic. Plagioclase, amphibole, minor biotite, and titanite present phenocrysts of these rocks.

Mineral chemistry

The composition of plagioclase phenocrysts of the Murgana dome-like body $An_{38.5-42.2}$ (core) to $An_{38.7-46.2}$ (rim); those of the lava flows varies from $An_{42.5-48.2}$ (core) to $An_{30.1-53.9}$ (rim); for the Vozdol volcanic rocks phenocrysts display range from center $An_{50.8}$ to $An_{36.2}$ in the periphery; and for dykes $An_{44.1-46.2}$ (core) to $An_{40.7-44.2}$ (rim). The rims are variable in composition and substantially overlap the field of the phenocryst cores, the compositions of plagioclase microlites vary from An_{31} to An_{48}. K-feldspar microlites (Or_{86-93}) where only analyzed in the Vozdol volcanic rocks. The amphiboles for all volcanic rocks display Mg# between 0.48 and 0.67. The contents of Si p.f.u. range between 6.40 and 6.55 and they plot on the limit of the magnesiohastingsite, pargasite, ferropargasite, hastingsite and Fe-edenite field of Leake et al. (1997). The composition of the amphibole crystals of the inclusions is different to the one of the volcanic rocks. It displays higher values of Mg# between 0.70 and 0.83 and is classified as magnesiohastingsite. The contents of Si p.f.u. of the amphiboles from the inclusions range between 5.90 and 6.10.

Trace elements

The MORB normalized patterns for the described magmatic rocks indicate enrichment of LILE and in lesser degree of some HFSE (Ce, Zr, P and Hf) with a strong negative Nb anomaly and a depletion of the Fe-Mg elements (Table 2). All these features are typical for subduction-related magmatic sequences due to the melting of sedimentary material in the subducted slab. In comparison to the volcanic rocks of an Andean-type active continental margin, the studied magmatic rocks show small K_2O, Ba and Hf enrichments and depletions of Nb, TiO_2, Zr and P_2O_5.

Table 2. Trace element composition of the representative volcanic samples

Elements (in ppm)	Dome-like body	Lava flows	Vozdol breccia	Dykes
Nb	7	7	6	9
Zr	121	98	127	123
Y	23	20	18	22
Sr	1430	781	871	641
Rb	72	63	46	102
Th	4	3	3	4
Pb	17	16	15	13
Ga	18	19	18	19
Zn	46	72	137	49
Cu	25	26	35	7
Ni	3	2	4	2
Co	50	10	13	7
Cr	10	14	15	13
V	96	127	139	89
Ba	870	1441	768	726
S	12	113	29	11
Hf	7	6	6	6
Sc	6	10	9	10
As	11	6	3	7
La		22.9	21	25.2
Ce		49.3	44.7	53.3
Pr		5.3	5.2	6.4
Nd		24	22.8	24.8
Sm		4.9	4.6	4.9
Eu		1.26	1.27	1.23
Gd		3.3	3	3.6
Dy		3.1	3	3.2
Ho		0.66	0.64	0.67
Er		1.8	1.7	1.8
Tm		0.26	0.24	0.26
Yb		1.5	1.4	1.6
Lu		0.22	0.18	0.25

All rocks have fractionated LREE and relatively flat HREE patterns (Stoykov et al., 2002), as typically found in subduction related volcanic rocks. The LREE enrichment ranges from 33 to 105 times chondritic, whereas La_n/Yb_n ratios vary from 10 to 13. Middle and heavy REE show relatively flat patterns, generally within 5-30 times that of chondritic ones. An Eu anomaly is not observed, which suggests that there was no plagioclase fractionation involved in genesis of the studied andesitic rocks. The data can be interpreted in terms of a chemically zoned magmatic chamber (according to the model of Hildreth, 1981). The rocks from the Murgana dome-like body show slightly enriched values of the LREE compared to the lava flows and the Vozdol volcanic rocks.

Sr and Nd isotopes

The Sr isotope ratios of the magmatic rocks from the Chelopech volcano and dykes display a small range between 0.7049 and 0.7055 after a 90 Ma correction (Stoykov et at., 2002). Generally $^{87}Sr/^{86}Sr$ ratios fall within the field previously defined by Kouzmanov et al. (2001) values from 0.7046 to 0.7061 (after 80 Ma correction) for the volcanic (andesite and dacite) and plutonic (granodiorite and granite) rocks from the southern part of the Central Srednogorie volcano-intrusive area.

Table 3. Sr and Nd Isotope composition

	$^{87}Sr/^{86}Sr_{90Ma}$	$^{143}Nd/^{144}Nd_{90Ma}$
Murgana dome-like bodies	0.7054	0.5125
Lava flows	0.7050	0.5125
Vozdol lava-breccia	0.7049	0.5124
Dykes	0.7055	0.5125

The Nd isotope ratios of the magmatic rock from the Chelopech volcano and dykes display a small range 0.5123 to 0.5125 after a 90 Ma correction.

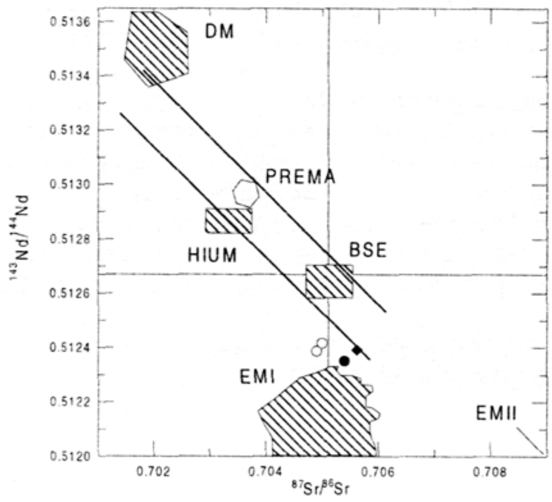

*Figure 4. Sr vs. Nd isotope correlation diagram, showing the main oceanic mantle reservoirs of Zindler and Hart (1986). Open circles, Vozdol lava-breccia; filed circle, lava flows; filed diamond, dome- like bodies. **DM**, Depleted Mantle; **BSE**, Bulk Silicate Earth; **EMI** and **EMII**, Enriched Mantle; **HIUM**, Mantle with High U/Pb ration; **PREMA**, frequently observed PREvalent MAntle composition*

CONCLUSIONS

The Upper Cretaceous volcanic rocks of the southeastern part of Etropole Stara planina are located in the central part of the Srednogorie zone. The magmatic products display Ca-alkaline to shoshonitic affinity. They are probably of Turonian age. The magma evolved from more acid volcanic rocks with 61-64 wt% SiO_2 of the earlier products (dome-like bodies, lava and agglomerate flows) to the to more basic ones with 55.5-58 wt% SiO_2 of the letters (Vozdol lava breccia neck). This chemical evolution and the absence of an Eu anomaly probably indicate a chemically zoned magmatic chamber. Magma mingling was a ubiquitous process and together with fractional crystallization controlled the evolution of the andesitic magmas of the Chelopech volcano. The behavior of the trace elements is similar to the andesitic rocks formed an active continental margin. The Sr and Nd isotope signature suggests derivation of melts generated in a mantle source modified by the addition of crustal material.

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