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RAPID COMMUNICATION

Nd–Sr–Pb isotopic composition and mantle sources of Triassic rift units in the Serbo-Macedonian and the western Rhodope massifs (Bulgaria–Greece)

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Abstract

We report on the field occurrence and isotopic compositions of metamafic rocks exposed in the Serbo-Macedonian (Volvi and Therma bodies) and western Rhodope (Rila Mountains) massifs of Bulgaria and Greece. These metamafic units consist of high- and low-Ti gabbroic and basaltic rocks, whose Nd–Sr–Pb isotopes are compatible with mantle-derived MORB and OIB components with a small amount of crustal material involved in their melt source. These isotopic features combined with the field observations are consistent with an intra-continental rift origin of the metamafic rocks protolith, and are comparable to those of the Triassic rift-related mafic rocks in the northern Aegean region.

Keywords: metamafic rocks, whole-rock Nd–Sr–Pb isotopes, Triassic rifting, Serbo-Macedonian–Rhodope massifs, Bulgaria, Greece

1. Introduction

The Serbo-Macedonian and Rhodope metamorphic massifs constitute the crystalline basement of the Alpine orogenic belt in the Balkan Peninsula and display a structural record of a late Mesozoic contractional deformation episode overprinted by an early Cenozoic extensional deformation (e.g. Ricou *et al.* 1998). Both massifs comprise amphibolite-facies metamorphic rocks of continental and oceanic origin (e.g. Liati, 2005; Himmerkus, Reischmann & Kostopoulos, 2009) that are intruded by a series of Late Cretaceous to Miocene granitoids (Zagorchev, Moorbath & Lilov, 1987; Dinter *et al.* 1995; Christofides *et al.* 2001). The S-vergent, imbricate crustal architecture of the Serbo-Macedonian and Rhodope massifs was assembled during a contractional phase in the hanging wall of a N-dipping Cretaceous–Tertiary subduction zone that was located within the Vardar Ocean farther to the SSW (in the present-day coordinate system) (Ricou *et al.* 1998). This shortening and nappe-stacking event resulted in crustal thickening, amphibolite-facies regional metamorphism and topographic build-up, reminiscent of many other collision zones (Dilek, 2006). The subsequent extensional collapse of this young orogenic belt led to metamorphic core complex formation starting in early Cenozoic time (e.g. Bonev & Beccaletto, 2007 and references therein).

The high-grade metamorphic units of the Serbo-Macedonian and Rhodope massifs are locally intercalated with mafic-ultramafic rocks, which are pervasively deformed and metamorphosed up to amphibolite facies. The same structural and metamorphic fabric elements observed in these mafic-ultramafic rock assemblages and in the surrounding rocks of the two massifs indicate that they all experienced the same deformational events, and that they were already part of the same crustal mosaic in the Balkan Peninsula by latest Cretaceous time. However, how these mafic-ultramafic rocks were incorporated into the protoliths of the Serbo-Macedonian and Rhodope massifs and the nature of their melt source(s) and the tectonic setting of their formation, are not well understood. The existing models consider them either as ophiolitic allochthonous tectonic sheets, representing the remnants of a Tethyan oceanic lithosphere, or as late-orogenic intrusive-extrusive complexes (Dixon & Dimitriadis, 1984; Robertson *et al.* 1996; Himmerkus *et al.* 2005; Himmerkus, Reischmann & Kostopoulos, 2009). These different interpretations have major implications for the crustal evolution of the Balkan Peninsula and need to be validated with field-based structural, geochemical, isotopic and geochronological studies.

In this paper, we report on the field occurrence and Nd–Sr–Pb radiogenic isotope geochemistry of the Volvi and Therma mafic-ultramafic units in the Serbo-Macedonian Massif and a series of other mafic complexes in the western Rhodope Massif exposed in southern Bulgaria and northern Greece (Fig. 1). Our data and field observations suggest that these mafic-ultramafic rock units represent para-rift assemblages emplaced during rifting of the Serbo-Macedonian and Rhodope massifs during Triassic time. This inferred rifting was aborted, however, before the onset of continental break-up and seafloor spreading, and thus no ocean basin was created between these rifted blocks. In the first part of the paper, we present a brief account of the

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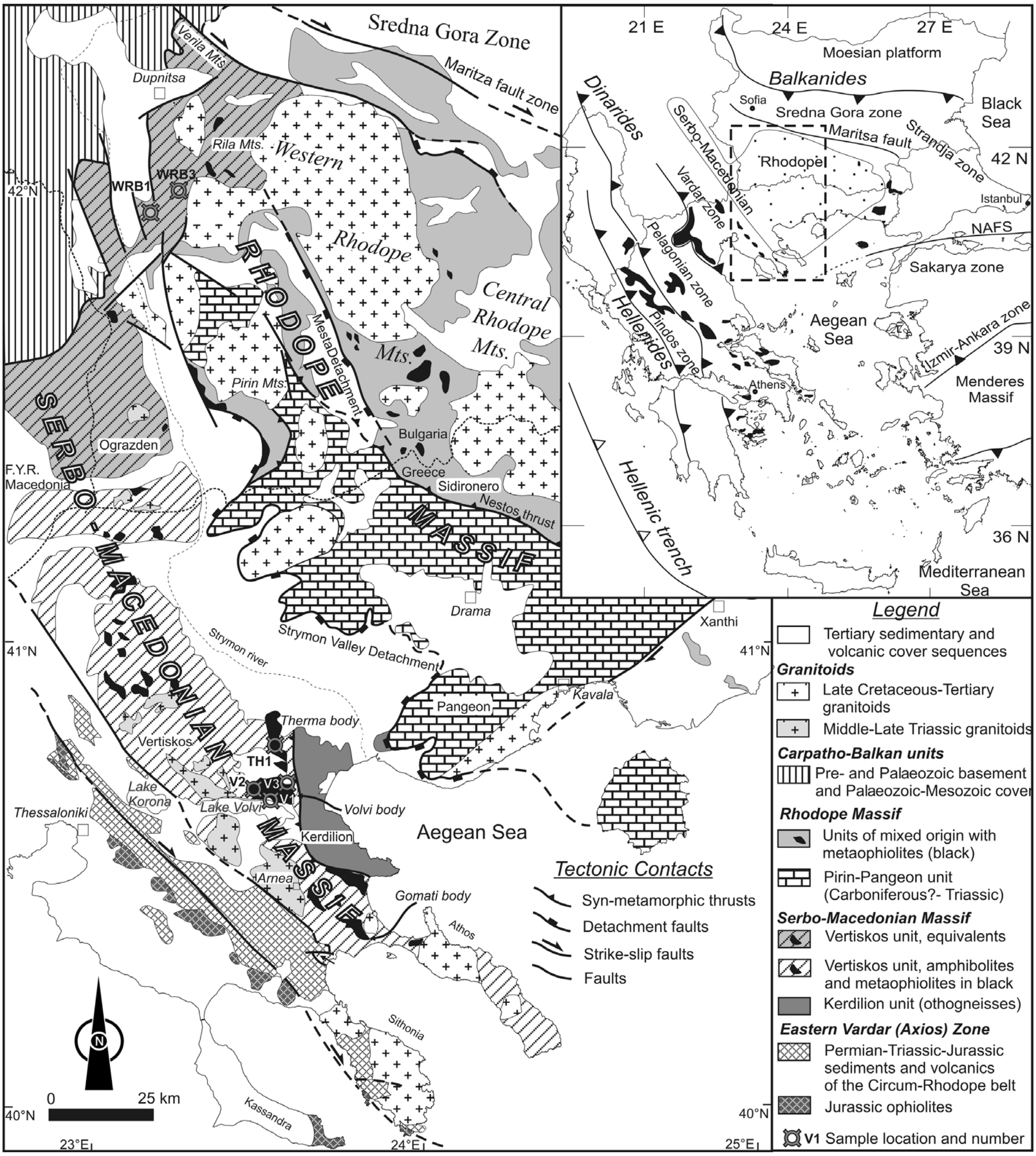

Figure 1. Tectonic map of the Serbo-Macedonian Massif and the western-central Rhodope Massif in southern Bulgaria and northern Greece and adjacent units (adapted from Kockel, Mollat & Walther, 1977; Zagorčev, 1994; Ricou *et al*. 1998), showing the locations of studied samples listed in Table 1. Inset: Tectonic framework of the Alpine system in the Aegean domain of the eastern Mediterranean region. The main exposures of Jurassic ophiolites are shown in black using data from Papanikolaou (2009).

regional geology, geochemistry and isotopic signature of the Triassic mafic-ultramafic rocks, and then discuss their origin and significance for the Mesozoic–Cenozoic tectonic evolution of the region.

2. Regional geology and geochemistry of Triassic mafic-ultramafic rocks

In the northern Aegean region, there are three major continental blocks in the crustal mosaic of the Balkan Peninsula. These include, from west to east, the Pelagonia, Serbo-Macedonian and Rhodope massifs (Fig. 1, inset). Jurassic ophiolites and Triassic mafic rift-related rocks occur on both sides of Pelagonia, whereas the same oceanic crust elements were emplaced from the south or occur within the Serbo-Macedonian and Rhodope massifs.

The Serbo-Macedonian Massif is separated from the Pelagonia microcontinent to the SW by the Vardar suture zone, which is part of the innermost Hellenides. To the north, the Serbo-Macedonian Massif continues laterally into the Rhodope Massif, which delimits the Alpine Balkan thrust–fold belt in the north (Fig. 1). The crustal basement units of the Serbo-Macedonian and the Rhodope massifs

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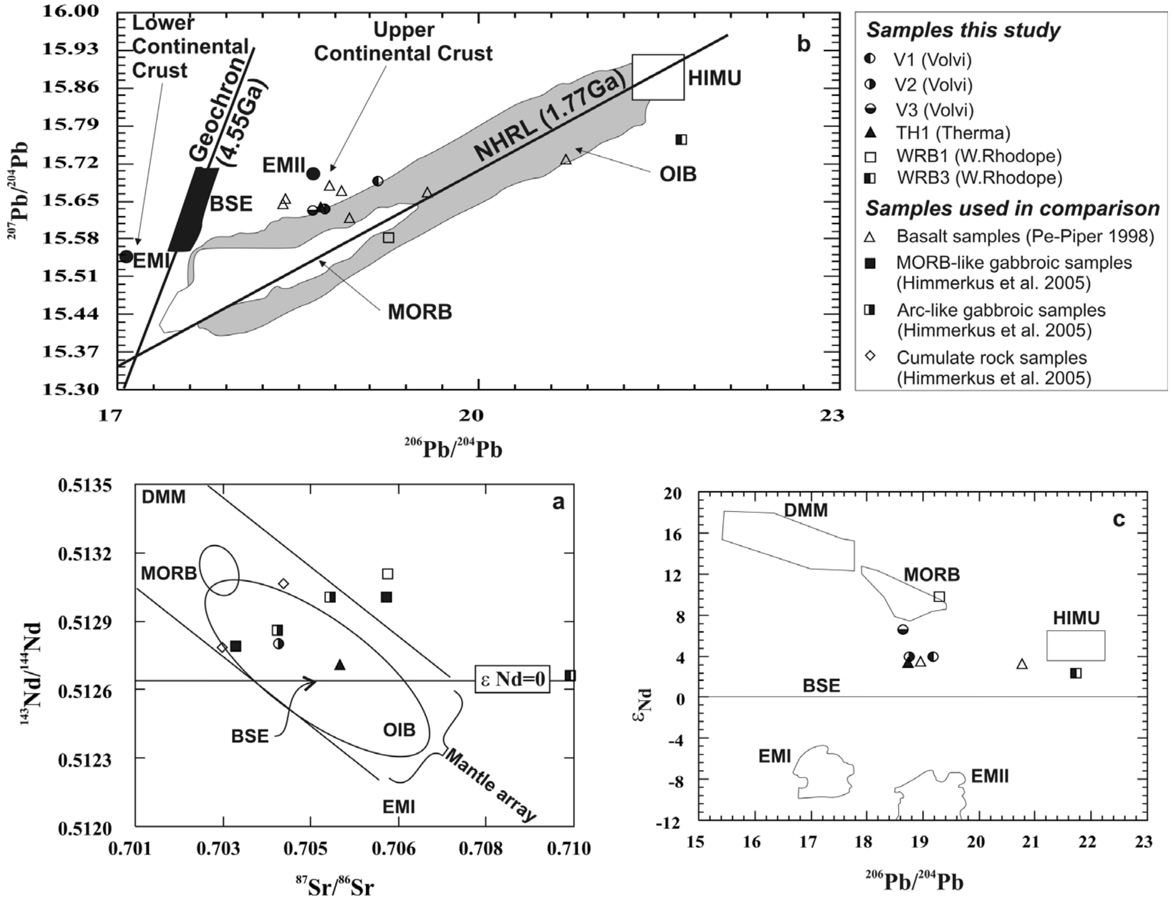

Figure 2. Correlation diagrams for Nd–Sr–Pb isotopes of the metamafic rocks. (a) $^{143}Nd/^{144}Nd$ v. $^{87}Sr/^{86}Sr$ diagram. DMM and EMI mantle reservoirs from Hart (1984); (b) $^{207}Pb/^{204}Pb$ v. $^{206}Pb/^{204}Pb$ correlation diagram. The NHRL and the fields of reference mantle reservoirs are after Zindler & Hart (1986), and the fields of OIB and MORB from Rollinson (1993); (c) εNd_t v. $^{206}Pb/^{204}Pb$ diagram. Mantle reservoirs are after Zindler & Hart (1986).

are structurally related to the Mesozoic development of the Neotethyan Vardar oceanic realm at the southern continental margin of Eurasia (Koukouvelas & Doutsos, 1990; Burg *et al.* 1996; Robertson *et al.* 1996; Dinter, 1998; Ricou *et al.* 1998; Bonev & Beccaletto, 2007). The Serbo-Macedonian and Rhodope massifs comprise mainly amphibolite-facies metamorphic basement rocks consisting of pre-Alpine (Neoproterozoic–Permian) and Alpine (e.g. Lips, White & Wijbrans, 2000; Liati, 2005; Himmerkus, Reischmann & Kostopoulos, 2009) units of continental and oceanic affinities. All these metamorphic units are intruded by granitoid plutons that range in age from Late Cretaceous to Miocene (Zagorchev, Moorbath & Lilov, 1987; Dinter *et al.* 1995; Christofides *et al.* 2001).

The high-grade crystalline rocks of the western-central Rhodope Massif are best exposed in the Verila, Rila and Pirin mountains in Bulgaria, and extend into the Central Rhodope Mountains in Bulgaria and Greece (Fig. 1). Almost all of these basement units in the western-central Rhodope Massif contain bodies and lenses of mafic-ultramafic rocks, showing mid-ocean ridge (MORB)-like geochemical affinities (Kolcheva & Eskenazy, 1988; Liati & Mposkos, 1990). Both high-grade metamorphic units and mafic-ultramafic intercalations with *c.* 245–250 Ma old protoliths structurally overlie along the WSW-vergent Nestos thrust fault the Upper Palaeozoic–Triassic (Kronberg, Meyer & Pilger, 1970; Liati, Gebauer & Fanning, 2011) platform carbonates represented by the Pirin-Pangeon unit (Papanikolaou & Panagopoulos, 1981; Zagorčev, 1994). The metamorphic basement of the Rila Mountains in the western Rhodope Massif is regarded as the northeastward extension of the Ograzden unit in the Serbo-Macedonian Massif (Zagorčev, 1994; Zagorchev, 2001). In the Rila Mountains, the metagabbro-metadiorite bodies within the high-grade metamorphic units show island arc tholeiitic (IAT) and calc-alkaline (CA) affinities (Machev, 2002). These mafic rocks are locally associated with sheared boudins of lherzolite?–clinopyroxenite cumulates (Bazylev *et al.* 1999). A complex Alpine ultra-high, high- and medium pressure tectonometamorphic history of the western-central Rhodope units is bracketed between 149 and 40 Ma (Liati, 2005 and references therein). Zircon U–Pb geochronology in the Rhodope units has revealed ubiquitous Permo-Carboniferous (310–253 Ma) and Late Jurassic–Early Cretaceous (160–134 Ma) granitoid protolith ages in the high-grade basement (e.g. Peytcheva *et al.* 2004; Liati, 2005; Turpaud & Reischmann, 2010) (Fig. 1).

The Serbo-Macedonian Massif (Mercier, 1966; Dimitriević, 1974) extends northwards into Serbia, F.Y.R. Macedonia, Greece and Bulgaria, and has been considered as either a separate crustal unit within the Internal Hellenides (Papanikolaou, 2009) that was thrust onto the Rhodope Massif (Kockel & Walther, 1965), or is part of a single continental block sharing together with the Rhodope Massif the same Alpine nappe-stacking kinematics (Ricou *et al.* 1998). In the Chalkidiki Peninsula of northern Greece, it has been subdivided into two separate series. The Lower Kerdilion series is separated by a SW-dipping fault (Kockel, Mollat & Walther, 1971, 1977) or ductile shear zone (Burg,

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Godfriaux & Ricou, 1995) from the Upper Vertiskos series. The Vertiskos series hosts mafic-ultramafic bodies, which are known as the Therma-Volvi-Gomati (TVG) complex (Dixon & Dimitriadis, 1984) (Fig. 1). Recent zircon U–Pb geochronology has shown the presence of Neoproterozoic (586 Ma) and Silurian (433–428 Ma) igneous basement rocks in the Vertiskos series gneisses, which are intruded by the Triassic (mean age 228 Ma), A-type Arnea granite (Himmerkus, Reischmann & Kostopoulos, 2009 and references therein) (Fig. 1). The Permian–Lower Triassic Examili clastic and volcanic successions overlying the Vertiskos series have been interpreted as the products of Triassic rifting (Dimitriadis & Asvesta, 1993).

Dixon & Dimitriadis (1984) identified the Volvi mafic rocks as 'transitional basalts' of mixed 'alkalic' and 'above-subduction-zone' geochemical affinities generated in an '*in situ*' Mesozoic intra-continental rift. Sr–Nd isotope chemistry of the TVG complex revealed MORB and IAT affinities interpreted in terms of formation in a supra-subduction zone environment (Himmerkus *et al.* 2005). The Volvi body mainly consists of isotropic coarse-grained massive and pegmatoid metagabbros and rare basaltic dykes, whereas the Therma body consists of basalts, rare altered peridotites and gabbros. All these mafic-ultramafic rocks of both the Therma and Volvi bodies are metamorphosed into massive or layered amphibolites, which largely preserve primary igneous textures and mineral phases. Liati, Gebauer & Fanning (2011) have reported a zircon U–Pb SHRIMP protolith age of 252 ± 13 Ma for the Volvi body metagabbros, implying Late Permian–Early Triassic magmatic crystallization.

In the western foothills of the Rila Mountains, along the Bistrica river valley, the amphibolite-facies gneiss and schist successions of the Rhodope Massif host thick metadiorite-metagabbro layers. These gabbro-diorite bodies commonly form massive or banded amphibolites, and locally show in weakly metamorphosed and deformed domains massive or pegmatoid textures similar to these observed in the Volvi mafic body. In the northern part of the Rila Mountains, massive amphibolites are intercalated with schists and overlain by migmatitic gneisses that texturally strongly resemble those of the Ograzden unit of the Serbo-Macedonian Massif. Between the Greek Serbo-Macedonian Massif and the Rila Mountains in the western-northern Rhodope Massif, other occurrences of metamafic rocks that are texturally similar to the Volvi body metagabbros are observed in the foothills of the Pirin Mountains (Fig. 1).

We have identified high- and low-Ti groups in the Volvi and Therma bodies (Serbo-Macedonian Massif) and in the Rila Mountains of the western Rhodope Massif based on the trace elements and rare earth element (REE) characteristics of the mafic-ultramafic rocks (SiO_2 = 45–56 wt %) (Bonev & Dilek, 2010). The high-Ti group rocks with high high-field-strength element (HFSE) and REE contents show E-MORB and mild within-plate oceanic floor signatures (OIB). The low-Ti group rocks exhibit, on the other hand, both partly N-MORB/E-MORB depleted and fractionated HFSE and REE characteristics and similar enriched characteristics comparable to the high-Ti group, and also an arc-like signature. Both high- and low-Ti groups display low- to medium-K tholeiitic and mostly calc-alkaline affinities of the gabbroic-basaltic protoliths. These geochemical signatures, trace element and REE characteristics of the mafic-ultramafic rocks are consistent with a continental rift setting for their tectonic environment of formation (Bonev & Dilek, 2010). We interpret the arc-like geochemical signature of the low-Ti group to have been inherited from previous subduction events (Palaeotethyan) in the region.

3. Nd–Sr–Pb isotope results and comparison with other Triassic rift units

We analysed three samples from the Volvi body (V1, V2, V3; low-Ti group), one sample from the Therma body (TH1; low-Ti group), together with two samples from the western Rhodope Massif metamafic rocks (WRB-1; high-Ti group, WRB-3; low-Ti group) for whole-rock Sr, Nd and Pb isotopic compositions. We compared the Nd and Pb isotopes of our rock samples to the published Nd and Pb isotopic compositions of the Triassic rift-related mafic rocks in the adjacent Pelagonian zone of the western Hellenides. We also compared the Nd and Sr isotopes obtained in this study with the Nd and Sr isotopic compositions of mafic rocks from the Athos-Volvi zone (equivalent of the TVG complex) (Himmerkus *et al.* 2005).

Chemical separation of the samples and whole-rock isotopic analyses were done in the TERRA Facility in the Department of Earth Sciences at Memorial University of Newfoundland (Canada) and calibrated against both international and internal standards. Sr–Nd–Pb isotopes were measured on a multicollector Finningan MAT 262V thermal ionization mass spectrometer, and $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$, $^{208}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{206}Pb/^{204}Pb$ ratios were corrected for mass fractionation and normalized to international standards. The whole-rock Nd, Sr and Pb isotopic compositions are given in Table 1.

The $^{143}Nd/^{144}Nd$ ratios of the mafic rock samples fall in the range of 0.5127–0.5131, with positive ε_{Nd} values that are characteristic of mantle melts. When time corrected for the crystallization age of the Arnea granite (228 Ma) adjacent to the Volvi body, the $\varepsilon_{Nd(t)}$ values vary from +2.4 to +9.8 (Table 1). The $^{87}Sr/^{86}Sr$ ratios of the Volvi (V1 = 0.7039), Therma (TH1 = 0.7051) and the western Rhodope samples (WRB-1 = 0.7061) display values characteristic of the oceanic crust. However, sample (WRB-3) from the western Rhodope massif has a higher $^{87}Sr/^{86}Sr$ ratio of 0.7098. These data indicate that the analysed mafic rocks except sample WRB-3 originated from magmas that were derived from a similar mantle source with a high time-integrated Sm/Nd ratio and with a moderate range of Rb/Sr ratios. In a correlative $^{143}Nd/^{144}Nd$ v. $^{87}Sr/^{86}Sr$ diagram (Fig. 2a), almost all of the samples (except sample WRB-3) show consistent values of obtained isotopic ratios to those of the TVG complex MORB-like, arc-like and cumulate mafic rock groups reported by Himmerkus *et al.* (2005).

The $^{206}Pb/^{204}Pb$ ratios of the analysed rocks show a relatively narrow range (18.64–19.28), reaching a higher $^{206}Pb/^{204}Pb$ value of 21.72 for sample WRB-3, which has the most radiogenic Pb (Table 1). These samples display a narrow range of $^{207}Pb/^{204}Pb$ ratios (15.58–15.77), and similarly uniform $^{208}Pb/^{204}Pb$ ratios (38.24–39.31), reaching a higher value in sample WRB-3. The Volvi–Therma mafic rocks exhibit a short linear trend straddling a progressive enrichment along the MORB–OIB line in the $^{207}Pb/^{204}Pb$–$^{206}Pb/^{204}Pb$ correlation diagram (Fig. 2b), plotting above the Northern Hemisphere Reference Line (NHRL) where enriched mantle reservoirs (EMI, EMII) are identified (Zindler & Hart, 1986). The Pb isotopic compositions plot within the large OIB field and close to the MORB field (Rollinson, 1993). Sample WRB-1 plots closest to the MORB field, while sample WRB-3, with distinctly high $^{206}Pb/^{204}Pb$, falls below NHRL approaching the HIMU field.

The analysed mafic rock samples have Pb isotope ratios that cluster close to those of Triassic, rift-related mafic rocks from Greece (Pe-Piper, 1998). Our samples from the Serbo-Macedonian and Rhodope massifs and those reported from Pelagonia by Pe-Piper (1998) all plot within the OIB field (closest to the MORB field), and mostly show

Table 1. Nd, Sr and Pb isotopic compositions of metamafic rocks from the Serbo-Macedonian and western Rhodope massifs

Sample	Nd (ppm)	Sm (ppm)	$^{147}Sm/^{144}Nd$	$^{143}Nd/^{144}Nd$	2σ	εNd_0	εNd_{228}	T	TDM2	$^{87}Sr/^{86}Sr$	2σ
V1	5.564	1.573	0.1710	0.512803	7	3.2	4.0	870	1240	0.703859	7
V2	12.900	3.759	0.1761	0.512805	4	3.3	3.9	975	1400	–	–
V3	5.616	1.624	0.1748	0.512941	5	5.9	6.6	455	821	–	–
TH1	5.579	1.184	0.1283	0.512708	4	1.4	3.4	616	790	0.705094	7
WRB-1	16.48	4.767	0.1749	0.513107	5	9.1	9.8	–	170	0.706098	7
WRB-3	19.30	4.193	0.1314	0.512662	5	0.5	2.4	720	905	0.709835	9

T (DePaolo, 1988)
TDM2 calculated using a linear evolution for a mantle separated from the CHUR at 4.55 Ga and having a present-day Epsilon value of +10
$^{143}Nd/^{144}Nd$ ratio corrected from the deviation from the JNdi-1 standard (0.512115; Tanaka *et al*. 2000). Mean value of MUN TIMS Lab: $^{143}Nd/^{144}Nd$ = 0.512137; Std Dev = 0.000019; n = 104

$^{87}Sr/^{86}Sr$ ratio corrected from the deviation from the NBS 987 standard (certified: 0.710248). Mean value of MUN TIMS Lab: $^{87}Sr/^{86}Sr$ = 0.710206; Std Dev = 33; n = 51

Sample	$^{208}Pb/^{204}Pb$	2σ	$^{207}Pb/^{204}Pb$	2σ	$^{206}Pb/^{204}Pbc$	2σ
V1	38.894	0.007	15.636	0.002	18.744	0.002
V2	39.310	0.009	15.687	0.003	19.172	0.002
V3	38.680	0.008	15.634	0.002	18.637	0.002
TH1	38.822	0.007	15.641	0.002	18.732	0.002
WRB-1	38.244	0.010	15.581	0.004	19.284	0.005
WRB-3	39.176	0.003	15.765	0.001	21.718	0.002

Ratios are reported corrected for mass fractionation.
Deviation from the certified values of repeated analyses of std NBS 981 yields a correction factor of 0.000982 ± 2 per amu.

Certified Todt *et al*. 1993

$^{208}Pb/^{204}Pb$	36.687
$^{207}Pb/^{204}Pb$	15.486
$^{206}Pb/^{204}Pb$	16.933

Mean of NBS 981 measurements (n = 15)

$(^{208}Pb/^{204}Pb)m$	Std Dev	$(^{207}Pb/^{204}Pb)m$	Std Dev	$(^{206}Pb/^{204}Pb)m$	Std Dev
36.634	0.010	15.467	0.003	16.920	0.003

Location of samples shown in Figure 1.

high $^{206}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ ratios. When $^{206}Pb/^{204}Pb$ is plotted against $\varepsilon Nd_{t=228}$, the isotopic ratios of our samples cluster between the Bulk Silica Earth (BSE) and MORB fields (Fig. 2c). The Nd isotopes are available only for two samples from the Triassic rift-related rocks in Pelagonia (Pe-Piper, 1998); when plotted as $\varepsilon Nd_{t=210}$ values together with our samples, both rock suites display strong similarities in terms of Nd–Pb isotopic compositions (Fig. 2c).

4. Discussion and conclusions

The Nd isotope compositions obtained in this study are consistent with N-MORB to E-MORB and OIB signatures of the samples displayed by their trace element and REE geochemistry (Bonev & Dilek, 2010). The range of Nd isotopes is consistent with the values of the oceanic crust developed in seafloor spreading centres, indicating their origin from a MORB mantle source, with contributions of enriched OIB-type, within-plate melts. We infer, therefore, the involvement of multiple mantle reservoirs in the mantle source region. The Pb isotope data suggest a more pronounced contribution of an OIB source, possibly due to a small degree of melting (e.g. Hickey-Vargas *et al*. 2007) of an OIB component mixed into the source mantle. The range of Sr isotopes also supports mixed mantle components in the source region, but in addition shows the enrichment process via crustal contamination, explaining the variations and an extremely high $^{87}Sr/^{86}Sr$ ratio in sample WRB-3. We infer a contribution from continental crust to explain the elevated $^{87}Sr/^{86}Sr$ ratio (0.7098) observed in this sample. This value is indistinguishable from that of regional crystalline basement of the Serbo-Macedonian Massif ($^{87}Sr/^{86}Sr$ = 0.7096) (Himmerkus, Reischmann & Kostopoulos, 2009).

Triassic rift-related mafic volcanic rocks showing enriched mantle and within-plate chemical signatures (E-MORB and OIB) are abundant in the Hellenides to the west. These volcanic assemblages have Nd–Pb isotopic signatures indicating an enriched, hydrated mantle source and plume-related HIMU component (Pe-Piper, 1998), although the sample closest to HIMU compositions is contaminated sample WRB-3. This feature of sample WRB-3, together with similar Pb isotopes of the Triassic rift-related rocks in Pelagonia that are both shifted toward the upper crust EMII reservoir (Fig. 2b), point to likely crustal influence in the melt source of the mafic rock assemblages in the region. This latter feature additionally points to an isotopic continuity of our TVG samples with the Triassic rift-related mafic volcanic suites in Pelagonia.

Our findings highlight coherent Nd–Sr–Pb isotopic ratios of the mafic rocks, indicating generation of magmas by partial melting of a mantle source ranging from MORB- to OIB-source mantle. These isotopic data indicate variable contamination of magmas by continental crust material. These features are compatible with an intra-continental rift to spreading centre setting. Comparison of the Nd–Sr–Pb isotope results with analogous data from Triassic rift-related mafic rocks in the Aegean region demonstrates regionwide similarity of the isotopic compositions, which in turn provides additional support for the proposed rift-related origin for the metamafic rocks of the western Rhodope and the Serbo-Macedonian massifs.

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