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The Upper Cretaceous ophiolite of North Kozara — remnants of an anomalous mid-ocean ridge segment of the Neotethys?

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Abstract: This study sheds new light on the origin and evolution of the north Kozara ophiolite, a part of the Sava-Vardar Zone. The Sava-Vardar Zone is regarded as a relict of the youngest Tethyan realm in the present-day Balkan Peninsula. The north Kozara ophiolite consists of a bimodal igneous association comprising isotropic to layered gabbros, diabase dykes and basaltic pillow lavas (basic suite), as well as relicts of predominantly rhyodacite lava flows and analogous shallow intrusions (acid suite). The rocks of the basic suite show relatively flat to moderately light-REE enriched patterns with no or weak negative Eu-anomaly, whereas those of the acid suite exhibit steeper patterns and have distinctively more pronounced Eu- and Sr- negative anomalies. Compared to the known intra-ophiolitic granitoids from the Eastern Vardar Zone, the acid suite rocks are most similar to those considered to be oceanic plagiogranites. The new geochemical data suggest that the basic suite rocks are similar to enriched mid-ocean ridge basalts. The geochemical characteristics of the acid suite rocks indicate that their primary magmas most probably originated via partial melting of gabbros from the lower oceanic crust. Our study confirms the oceanic nature of the north Kozara Mts rock assemblage, and suggests that it may have formed within an anomalous ridge setting similar to present-day Iceland.

Key words: Balkan ophiolites, Sava-Vardar Zone, E-MORB, rhyodacite, acid magmatism.

Introduction

One of the major conundrums in geodynamic reconstructions of the Balkan Peninsula is the problem of the final closure of the Tethys Ocean. The most recent and widely accepted interpretation suggests that the Sava-Vardar Zone (SVZ) is the relict of the youngest Tethyan realm. According to this standpoint, most Balkan ophiolites, which occur along three sub-parallel belts roughly extending NNW–SSE, were emplaced during the Late Jurassic (Robertson & Karamata 1994; Bortolotti et al. 2002; Schmid et al. 2008; and references therein). These ophiolite belts have been given different names, and those most often used are East and West Vardar and Dinaric ophiolites (see Schmid et al. 2008). Although there are strong disagreements about how many oceans were involved in Mesozoic geodynamic events, most authors share the opinion that an oceanic realm still existed during Late Cretaceous. The term Sava-Vardar Zone was first suggested by Pamić (1993, 2002) who regarded it as the continuation of the Periadriatic Zone. Accepting his view that it was the latest suture in this region, Schmid et al. (2008) slightly changed the name into Sava Zone in order to distinguish it from other Vardar ophiolitic units. In this study we use the name Sava-Vardar Zone by defining this unit which had survived the Late Jurassic ocean(s) closure and the emplacement of the West and East Vardar ophiolites, and which supposedly represents the last suture between the Tisza/Dacia and Dinarides (Karamata 2006; Schmid et al. 2008; Robertson et al. 2009). The age of the final closure of this last oceanic realm is constrained by a regional metamorphic overprint at ~65 Ma, which was recorded on the Maastrichtian siliciclastic rocks belonging to the deepest parts of the SVZ accretionary wedge (Ustaszewski et al. 2010).

One of the key SVZ localities is exposed on the northern slopes of the Kozara Mts (north Bosnia and Herzegovina). This is the place where it was first documented that, besides the generally known Upper Jurassic ophiolites, remnants of Upper Cretaceous oceanic crust also exist (Jelaska & Pamić 1979; Karamata et al. 2005; Ustaszewski et al. 2009). Ustaszewski et al. (2009) reported the first U/Pb radiometric ages and provided a detailed geological and petrological study of the north Kozara ophiolite-related igneous rocks. In addition to providing accurate age data, the authors conclude that the north Kozara ophiolite represents a bimodal igneous association. They suppose that the north Kozara ophiolite is the relict of an oceanic plateau, leaving a possibility that it formed in a back-arc setting still open. Their hypothesis about an intra-oceanic geotectonic setting was postulated using a combination of geological and geochemical arguments and the latter was mainly based on the observation that the north Kozara basic rocks are geochemically more enriched than normal mid-ocean ridge basalts (NMORB).

In this study we report and discuss a new set of major element and trace element data of igneous rocks of the north Kozara ophiolite complex. The main aim of this study was twofold: firstly, to try to further constrain the geotectonic setting of this ophiolite by taking a closer look at the geochemistry of the basic igneous rocks, and secondly to address the petrogenesis of the acid rocks of this bimodal association, which was neglected in the previous research. Our results confirm earlier views that there is a close petrogenetic link between the basic and the acid rocks of this bimodal association and that this has important implications for geodynamic reconstructions in this area.

Geological setting

The SVZ trends N–S in central Serbia and, then, toward the northwest, it bends parallel to the Sava River (see inset of Fig. 1). In central Serbia, the SVZ suture assemblage is represented by Senonian flysches, whereas more to the north along the southern margin of the Pannonian Basin it is exposed in the form of several scattered inselbergs (Belak et al. 1998; Pamić 2002; Slovenec et al. 2010). One of these is the north Kozara body, the largest exposure of ophiolites in this area.

The north Kozara ophiolite was thrust onto the ophiolite melange of south Kozara, which belongs to the Upper Jurassic West Vardar ophiolite belt during the latest Cretaceous to Early Paleogene (Schmid et al. 2008; Ustaszewski et al. 2009). In map view (Fig. 1), the north Kozara complex appears as a partly dismembered slice of ophiolite rocks, which is mostly covered by Cenozoic formations. It is approximately 15 km long and extends ENE–WSW, starting from Gornji Podgradci to Maglajci in the east and the west, respectively (Fig. 1). In map view, the ophiolite slice consists of several blocks surrounded by unconformably overlying Maastrichtian to Paleocene mostly siliciclastic sediments. They are represented by arkoses, lithic sandstones, conglomerates and shallow-water limestones (Ustaszewski et al. 2009). The ophiolite blocks show a regular WNW–ESE distribution, with pillowed basalts situated in the south, a series of diabase dykes in the middle part and gabbros in the north. A similar ophiolite slice is found approximately 20 km northwest, in the area of Kostajnica (Fig. 1). The north Kozara ophiolites are separated from the Upper Jurassic ophiolites of south Kozara by a major N-dipping thrust (Ustaszewski et al. 2009). In contrast to the south Kozara ophiolite complex, the north Kozara ophiolite does not have exposed ultramafic rocks and has distinctively larger masses of acid rocks. Moreover, the north Kozara acid rocks are characterized by a strong predominance of volcanic rocks.

Approximately 5 km north of the north Kozara ophiolite there is the series of Prosara Mt (Fig. 1). It is another inselberg of the SVZ, which consists of two tectonic units separated by a northward dipping thrust. This structure was recently re-defined by Ustaszewski et al. (2010) as a low-angle detachment recording latest Oligocene/Miocene extensional unroofing of the Prosara inselberg. The lower, southern, unit is slightly or non-metamorphosed whereas the upper, northern, unit consists of various rocks metamorphosed under up to greenschist facies conditions (Šparica & Buzeljko 1984; Jovanović & Magaš 1986). Both units are cut by decametric intrusions of alkali feldspar granites that were dated by U/Pb zircon age to 82.68 ± 0.13 Ma (Ustaszewski et al. 2009). The granites show the same schistosity as their host rocks indicating that deformation is post-Late Cretaceous.

Samples and analytical methods

A set of 23 samples of both groups of the north Kozara bimodal magmatic association was analysed optically and chemically. For the basic rock group only diabases and fine-grained isotropic gabbros were sampled, whereas the samples of acid rocks cover all rock types found in the field, from almost aphyric silicic lava flows, through fine-grained rhyolite/rhyodacite dykes to medium-grained granite. Sixteen samples were analysed on major and selected trace elements in the Laboratory of the Department of Earth Sciences — University of Perugia (Italy) using an XRF device. The XRF included an X-ray tube with Rn and W anode, with acceleration voltage and electric current ranging from 40 kV to 45 kV to and from 30 mA to 35 mA, respectively. An LiF-200 crystal analyser was used for radiation separation in working regime without vacuum. In addition, eight samples were analysed for major and full range trace elements in the ACME Laboratories Ltd. Vancouver (Canada). Major element oxides were determined using ICP atomic emission spectrometry (detection limits around 0.001–0.04 %). Concentrations of trace and rare earth elements were measured by ICP-MS (detection limits 0.01–0.5 ppm). STD SO-17 was certified in-house against 38 Certified Reference materials including CANMET SY-4 and USGS AGV-1, G-2, GSP-2 and W-2. The accuracy of the analyses is within limits of 2–5 % for major elements, 10–15 % for trace elements and 1–5 % for REEs.

Results

Rock classification and petrography

The accurate rock classification of the north Kozara igneous rocks is difficult because some rocks are, at least to some extent, affected by low temperature metamorphic processes. In general, by combining fieldwork data, petrography and chemical characteristics, the studied rocks are distinguished into: (1) the basic and (2) the acid suite. The basic suite, which may comprise some intermediate rocks as well, vastly predominates, however, in spite of that, the relative proportion of acid rocks is very high. The relative abundance of acid rocks is remarkably higher than the relative proportion of acid magmatic rocks in other ophiolites of the Balkan Peninsula (e.g. Šarić et al. 2009). Ustaszewski et al. (2009) reported a detailed petrographic study of the north Kozara basic rocks, including field images and photomicrographs. Thus, in this study we provide only brief descriptions for the basic suite, whereas a special emphasis is put on the lithology and petrography of the acid rocks, which was mainly neglected by earlier researchers.

Basic suite

The basic suite is mostly represented by gabbros, diabases and basalts. **Gabbros** compose the lower part of the north Kozara ophiolitic sequence. They mainly occur in the north of the complex, where they appear as km-sized irregular masses (Fig. 1). The rocks are commonly hypidiomorphic, coarse- to

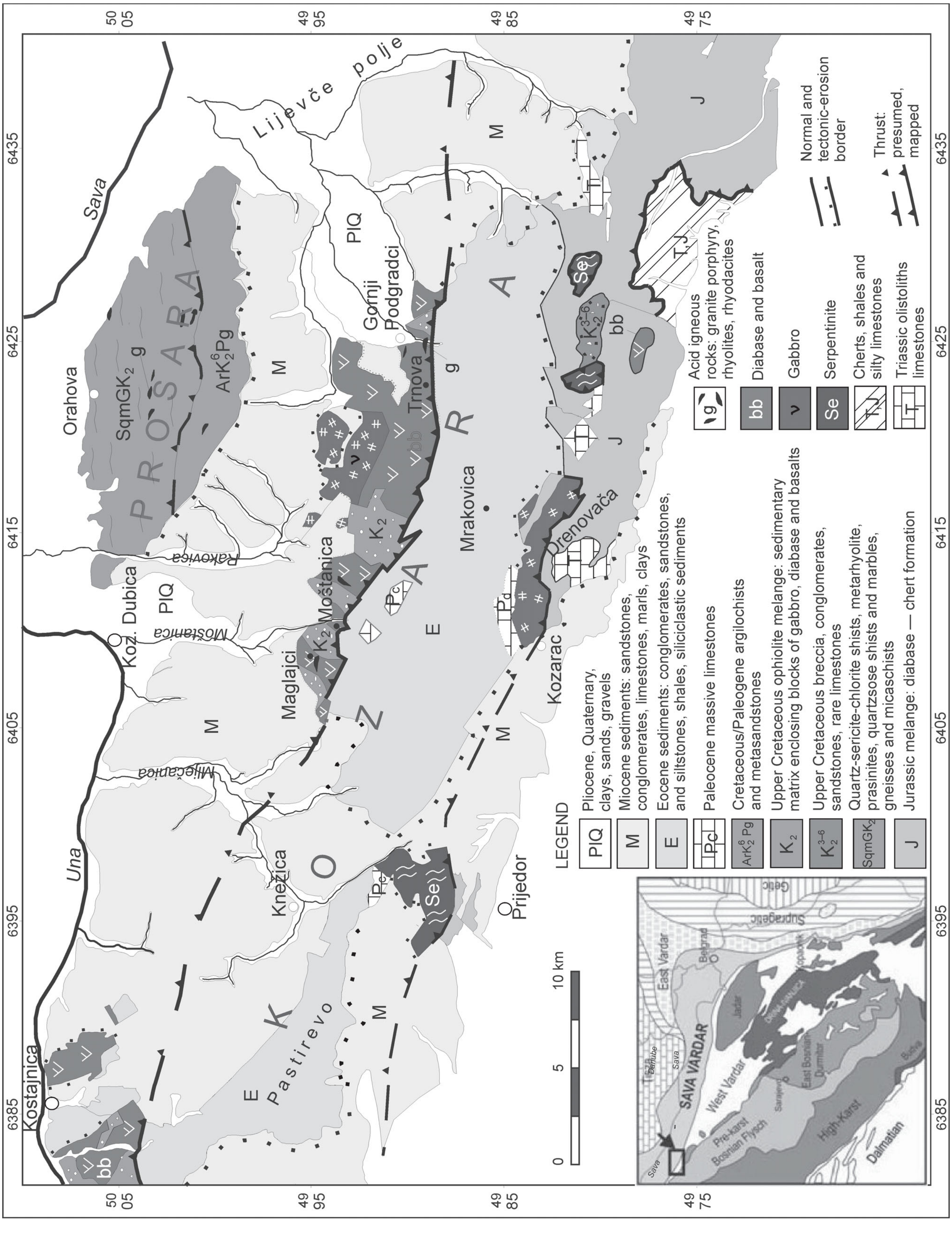

Fig. 1. Geological map of the Kozara Mts and the surrounding areas. Compiled from the Basic Geological Map of SFR Yugoslavia 1:100,000, Sheets: Banjaluka (Mojićević et al. 1976, 1977), Prijedor (Đerković et al. 1975), Kostajnica (Jovanović & Magaš 1986) and Nova Gradiška (Šparica et al. 1983). The inset shows geotectonic regionalization after Schmid et al. (2008).

medium-grained textures and show isotropic fabric. Occasionally, coarser-grained varieties with observable cumulitic textures are also observed. The gabbros consist of hypidiomorphic plagioclase and commonly uralitized clinopyroxene, the interstitial space is filled by fine-grained Fe-Ti oxide and secondary minerals, mostly albite, chlorite, epidote and calcite. At some places, very coarse-grained gabbro masses and veins of gabbro pegmatite are found. **Diabase** mostly occurs within typical sheeted dyke complexes, the best exposed are found near Trnova (Fig. 1). More rarely, diabase appears as isolated dykes and irregularly shaped shallow intrusions, which cut the gabbro zone. In general, diabase dykes vary in thickness from a few cm up to 0.5 m. Diabase is composed of idiomorphic to hypidiomorphic plagioclase and hypidiomorphic clinopyroxene as primary phases, whereas Fe-Ti oxide, sphene and apatite appear as accessories. Like the gabbros, the diabases are also affected by low temperature ocean floor metamorphism. The most altered varieties usually contain albite, epidote, chlorite, prehnite, uralite, secondary opaque minerals and leucoxene. **Basalts** appear as pillow lavas or coherent and volcaniclastic effusions. More rarely, they form feeding dykes cutting the extrusive facies. Most basalts contain scarce phenocrysts or are almost aphyric with only rare elongated laths of plagioclases enclosed by volcanic glass commonly replaced by chlorite or clay minerals. The pillow lavas are found intercalated with 'Scaglia Rossa' red pelagic limestones. This field observation was taken as first evidence that the north Kozara basalts are late Campanian to early Maastrichtian in age (Karamata et al. 2000, 2005; Grubić et al. 2009; Ustaszewski et al. 2009; Vishnevskaya et al. 2009).

Acid suite

Acid rocks are generally distinguished into volcanic and subvolcanic/intrusive facies. The largest outcrops of **acid volcanics** are found south and south-west of Gornji Podgradci (Fig. 1). There relicts of presumably larger piles of silicic lava crop out. These lavas do not have direct magmatic contacts with the adjacent masses of basalt and diabase, but they commonly display a similar N-dipping orientation as the bulk of the north Kozara ophiolite. The silicic lavas appear as up to 15 m high roadcuts and cliffs (Fig. 2a). The lava is commonly platy jointed with individual plates commonly between 0.3 and 1 m in thickness. Because they can be variable in colour, these rocks were sometimes mistaken for altered diabase, and this is probably a reason why their abundance (and significance) was merely underestimated. They are predominantly aphyric or have few phenocrysts and usually show banding or foliated fabric related to magmatic flow. Plagioclase is most abundant among the phenocrysts, whereas quartz and K-feldspar are rare. The groundmass typically displays banding in combination with classical perlitic texture (Fig. 2c). Most samples underwent high-temperature devitrification that resulted in microspherulitic and micropoikilitic textures (Fig. 2e,d). Devitrification bands are sometimes folded (Fig. 2f) suggesting that devitrification occurred while the acid magma was still hot and deformable. Plagioclase is idiomorphic, sometimes displaying perfect crystal shapes against the glassy groundmass. It appears as individual phenocrysts and only some samples show the presence of irregular mm-sized glomeroporhyritic nests. Quartz phenocrysts are usually ~0.3 mm in diameter. They are commonly rounded and partially embayed. Quartz also appears as a product of secondary recrystallization filling lens-like voids and lithophysae in the groundmass. Alkali feldspars as phenocrysts are very rare. More often, they appear as submilimetric laths in the groundmass, mostly as product of devitrification. The **subvolcanic/intrusive acid rocks** are predominantly represented by leucocratic dykes that are cutting the sheeted diabase complexes. The acid dykes are up to 1–2 m in thickness and are often parallel to adjacent diabase (Fig. 2b). They are plagioclase- rarely also quartz-phyric rocks with a holocrystalline groundmass composed of the same phases (Fig. 2g). Subordinate amounts of devitrified volcanic glass can be observed extremely rarely. Granitoid rocks are found only near Moštanica (Fig. 1). The granite is leucocratic and has a hypidiomorphic granular texture. It is predominantly composed of variable amounts of plagioclase, quartz and K-feldspar. Plagioclase is tabular and hypidomorphic, whereas K-feldspar and quartz are anhedral and usually fill the interstitial space, sometimes forming micrographic intergrowths (Fig. 2h). Primary mafic minerals are replaced by fine-grained chlorite aggregates. The form of these aggregates suggests that biotite was originally present. Opaque minerals, apatite and zircon are the main accessories.

Rock geochemistry

The results of chemical investigations are given in Table 1a,b. In most diagrams data reported by Ustaszewski et al. (2009) are also plotted. Most studied igneous rocks show less than 53 wt. % and more than 65 wt. % SiO_2, with the exception of three samples having ~57, ~61 and ~64 wt. % SiO_2. Given that these three rock samples contain magmatic quartz, they are plotted within the acid suite. Because of petrographic evidence of low-temperature alteration processes, and relatively high loss on ignition (LOI) values (mostly between 2 and 6 wt. %), the chemical classification is based on so-called 'immobile elements'. The Nb/Y vs Zr/Ti diagram (Winchester & Floyd 1977; Fig. 3) shows that the basic suite samples predominantly plot within the corner of the subalkaline basalt field and toward the andesite/basalt and andesite fields, whereas those of the acid suite stretch along the rhyolite/rhyodacite/dacite fields.

Most samples of the basic suite comprise a relatively narrow silica range of 45–50 wt. % SiO_2. They are also characterized by relatively high titanium contents, ranging from 1.2 to 2.5 wt. % TiO_2. MgO and CaO contents vary 4–8 wt. % and 4–9.5 wt. %, respectively. Mg#mol[MgO/(MgO+FeOt)] values are mostly between 0.6 and 0.75. MgO and CaO show negative correlations with silica contents, whereas other major oxides display either a poor correlation or data scattering (Fig. 4). The acid suite samples display a much wider silica range of 57–75 wt. % SiO_2. They also show remarkable variations in the abundance of other major oxides, in particular Al_2O_3 (9–18 wt. %), Na_2O (1–9 wt. %) and K_2O (<1–7 wt. %). Although some of these variations can be the result of alter-

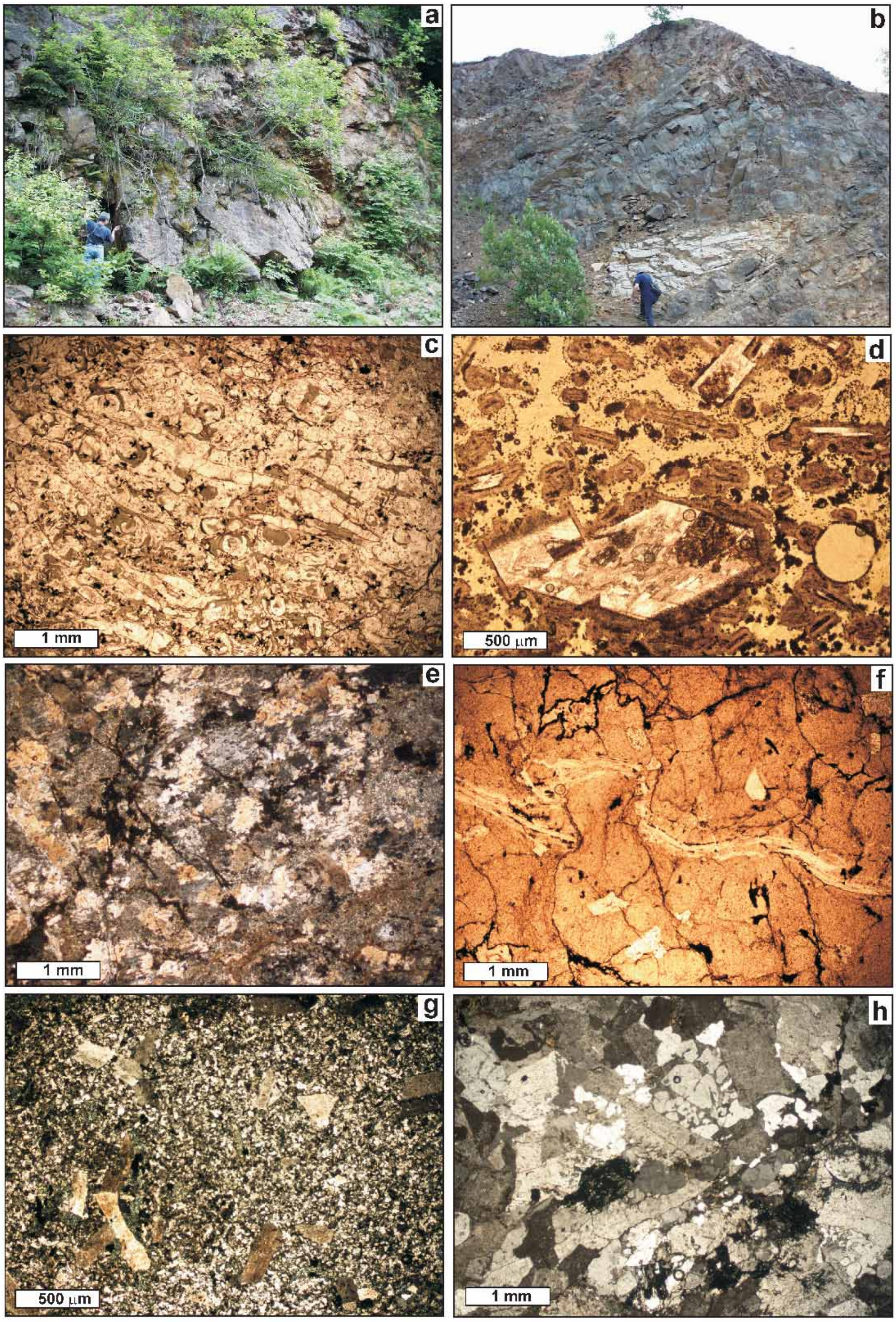

Fig. 2. Field photos and photomicrographs of the north Kozara acid rocks. **a** — Relicts of rhyodacite lava flows outcropping as steeply dipping 'walls' along the roadcuts between Mrakovica and Gornji Podgradci; **b** — A leucocratic rhyodacitic dyke cutting diabases of the Trnova sheeted dyke complex; **c** — Banded to perlitic rhyodacite (plane parallel light — PPL); bands are recrystallized into fine-grained mosaic quartz aggregates; **d** — Microspherulitic texture in devitrified rhyodacite (PPL); **e** —Micropoikilitic texture in devitrified dacite; note patchy quartz crystals that are optically continuous and enclose partially sericitized feldspar laths (plane crossed light — PCL); **f** — Folding in rhyodacite; the band which is folded formed in response to devitrification processes implying that devitrification occurred during slow solidification and under high temperature conditions (PPL); **g** — Holocrystalline rhyodacite cutting diabase of the Trnava sheeted dyke complex (PCL); **h** — Granophyre texture in granitoid rock near Moštanica (PCL).

Table 1a: Chemical composition of the rocks of the northern Kozara basic suite.

No.	*KZ-53/2*	MAG 11/2	KZ-44	KZ-46	*KZ-54*	MAG 11/4	KZ-40	TRNAV 1/2	TRNAV 1/1	KZ-26	*KZ-26*
Rock	*FG-G*	FG-G	D	D	*MG-G*	MG-G	D	D	D	D	*D*
SiO_2	*46.80*	47.64	47.79	48.14	*48.60*	48.70	48.79	48.89	50.02	50.96	*53.50*
TiO_2	*1.68*	1.43	1.33	1.48	*1.86*	1.54	1.84	1.33	2.42	2.25	*1.87*
Al_2O_3	*14.15*	15.67	14.55	13.16	*12.90*	15.69	12.17	16.67	14.81	12.24	*12.45*
$Fe_2O_3^t$	*11.30*	11.55	11.26	12.30	*10.75*	11.63	14.21	10.29	13.28	13.18	*10.65*
MnO	*0.16*	0.20	0.16	0.18	*0.15*	0.19	0.19	0.15	0.22	0.23	*0.17*
MgO	*7.21*	7.69	7.91	7.98	*7.38*	7.12	6.86	7.25	4.75	6.20	*3.65*
CaO	*9.20*	6.95	9.61	8.75	*6.21*	7.23	7.31	9.50	7.20	6.05	*4.28*
Na_2O	*2.88*	2.97	2.64	3.01	*3.88*	4.30	3.16	2.61	4.20	3.80	*5.03*
K_2O	*0.35*	0.68	0.23	0.18	*0.75*	0.34	0.30	0.28	0.84	0.50	*0.45*
P_2O_5	*0.22*	0.25	0.20	0.21	*0.19*	0.25	0.27	0.17	0.37	0.28	*0.28*
LOI	*5.16*	5.89	3.95	3.88	*6.83*	3.75	3.91	3.38	2.79	3.79	*6.05*
Tot	*99.11*	100.92	99.64	99.27	*99.50*	100.74	99.02	100.52	100.90	99.48	*98.38*
Ba	*63.5*	141	72	61	*147.5*	108	90	97	188	86	*45.3*
Co	*42*	49	52	71	*36.3*	48	76	41	40	74	*30.7*
Cs	*0.53*				*0.62*						*4.98*
Ga	*17.8*	27			*15.9*	26		30	28		*18.5*
Hf	*3*				*3.3*						*5.3*
Nb	*6.2*	14	2	2	*7.1*	13	3	7	23	12	*14*
Rb	*8.7*	13	9	13	*20.5*	5	18	8	23	12	*11.7*
Sr	*183.5*	203	193	174	*224*	163	140	169	259	77	*64.7*
Ta	*0.4*				*0.5*						*1*
Th	*0.91*	10			*1.08*	9		15	3		*3.92*
U	*0.27*				*0.49*						*1.56*
V	*271*	250	225	271	*287*	218	323	233	312	287	*229*
W	*1*				*1*						*2*
Zr	*110*	134	118	134	*120*	127	175	95	248	236	*205*
Y	*29.4*	30	27	28	*32.6*	29	40	27	53	44	*38*
La	*7.4*	16			*8.8*	17		17	22		*16.9*
Ce	*18.6*	11			*21.2*	8		0	34		*39*
Pr	*2.87*				*3.28*						*5.41*
Nd	*13.8*				*15.5*						*23.5*
Sm	*3.93*				*4.71*						*6.07*
Eu	*1.47*				*1.66*						*1.62*
Gd	*4.78*				*5.45*						*6.77*
Tb	*0.95*				*1.07*						*1.21*
Dy	*5.58*				*5.98*						*7*
Ho	*1.23*				*1.35*						*1.49*
Er	*3.33*				*3.8*						*4.23*
Tm	*0.5*				*0.57*						*0.63*
Yb	*3.11*				*3.44*						*3.84*
Lu	*0.46*				*0.53*						*0.6*
Pb	*<5*	16			*13*	10		20	10		*7*
Ni	*63*	38	97	84	*49*	33	59	46	32	39	*17*
Cr	*260*	114	301	345	*240*	113	93	272	38	63	*40*

Explanation: $Fe_2O_3^t$ — Total iron as Fe_2O_3. The analyses are ordered according to silica contents with ICP-MS analyses (Acme, Canada) given in italic. D — Diabase, FG-G — Fine-grained gabbro, MG-G — Medium-grained gabbro, LOI — Loss on ignition.

ation processes, these effects should not be very large because there is no correlation between LOI values and Na_2O, K_2O, Rb or Ba contents and simultaneously the concentrations of these elements are very well correlated ($R^2_{K-Na}>0.7$, $R^2_{Ba-Rb}>0.9$). It is generally observed that the samples of acid lava flows are relatively homogeneous with high silica (>65 wt. % SiO_2) and high potassium contents (~5 wt. % K_2O). These rocks are also slightly to moderately peraluminous with Alumina Saturation Index [ASI=mol(Al_2O_3/($CaO+Na_2O+K_2O$))] ranging from 1 to 1.4. On the other hand, four samples of leucocratic dykes have invariably low potassium contents and approach trondhjemitic composition. The variations between SiO_2 contents and CaO and MgO are generally negatively correlated both for the acid and basic rocks. However, the contents of some major oxides, in particular Al_2O_3, form almost parallel trends whereas others, such as TiO_2 and P_2O_5 display inflections (Fig. 4).

The basic suite samples show generally elevated concentrations of compatible trace elements, for instance Ni contents are up to 100 ppm, Cr up to 350 ppm and Co up to 50 ppm (Table 1a). The contents of these trace elements are strongly positively correlated with MgO contents and negatively with SiO_2 contents (Fig. 5). Compatible trace elements are present in low concentrations in the rocks of the acid suite (Ni<40 ppm, Cr mostly <50 ppm). Sr also behaves compatibly in both suites and is more enriched in the rocks of the basic suite (Fig. 5). The concentrations of most incompatible trace elements are higher in the rocks of the acid suite

Table 1b: Chemical composition of the rocks of the northern Kozara acid suite.

No.	***KZ-21/1***	**KZ-21/1**	**MOST 13/1**	**MAG 11/3**	***KZ-32***	**TRN 5**	**KZ-27**	**KZ-27**	***KZ-23/2***	**TRN 3**	**CR 8**	**MOST 14**
Rock	***QD***	**RD**	**RD**	**RD**	***RH***	**RD**	**RH**	**RH**	***RH***	**RH**	**RH**	**G**
SiO_2	*57.30*	61.13	64.25	66.13	*66.66*	68.92	69.42	70.10	*70.70*	73.85	74.29	72.14
TiO_2	*0.80*	0.65	1.36	0.39	*0.36*	0.47	0.36	0.30	*0.31*	0.27	0.21	0.30
Al_2O_3	*17.95*	18.51	14.59	14.13	*15.77*	14.55	11.75	12.00	*15.11*	14.14	13.75	13.80
Fe_2O_3	*3.89*	3.75	7.67	3.62	*2.10*	1.87	5.50	5.35	*1.84*	1.91	2.04	4.45
MnO	*0.02*	0.03	0.07	0.12	*0.01*	0.04	0.02	0.02	*0.01*	0.02	0.04	0.04
MgO	*1.84*	2.19	2.60	2.18	*0.58*	2.03	0.87	0.49	*0.39*	0.40	0.97	0.19
CaO	*2.77*	0.86	0.54	2.64	*0.34*	1.82	0.20	0.05	*0.15*	0.51	0.27	0.23
Na_2O	*9.00*	8.06	5.18	4.09	*2.38*	7.18	1.49	1.46	*2.58*	5.81	5.57	5.49
K_2O	*1.19*	0.21	0.44	2.65	*6.40*	0.25	5.83	5.72	*6.51*	2.21	1.70	2.53
P_2O_5	*0.17*	0.25	0.24	0.10	*0.09*	0.12	0.08	0.01	*0.17*	0.04	0.06	0.05
LOI	*4.38*	4.24	3.28	4.07	*5.11*	2.83	4.30	2.72	*1.94*	0.88	1.19	0.82
Tot	*99.31*	99.87	100.22	100.12	*99.80*	100.08	99.82	98.22	*99.60*	100.04	100.09	100.04
Ba	*27.1*	20	103	371	*31.5*	95	468	419	*835*	236	219	354
Co	*10.4*	14	11	8	*7.3*	5	12	1.5	*1.8*	5	4	2
Cs	*0.3*				*4.47*			11.6	*5.81*			
Ga	*16.1*		29	25	*29.6*	18		24.7	*24.7*	22	21	29
Hf	*9.2*				*61.8*			17.1	*13.1*			
Nb	*17.5*	14	23	15	*81.9*	15	29	34.3	*18.7*	18	18	48
Rb	*4.5*	2	12	65	*7.7*	6	114	111.5	*214*	55	41	62
Sr	*66.3*	80	47	102	*58.9*	93	47	35.4	*111*	52	88	34
Ta	*1.3*				*6.5*			2.2	*1.5*			
Th	*14.05*		28	29	*33.8*	26		13.35	*24*	36	33	30
U	*2.08*				*14.25*			4.73	*6.76*			
V	*84*	122	101	21	*26*	41	7	7	*14*	7	7	7
W	*1*				*5*			2	*2*			
Zr	*336*	337	347	334	*2290*	345	764	710	*453*	315	289	809
Y	*32.6*	35	61	41	*116.5*	35	92	73.8	*54*	46	48	116
La	*33.3*		43	36	*72.7*	44		49.7	*48.6*	34	33	56
Ce	*73.8*		83	68	*181.5*	85		99.4	*82.9*	68	89	151
Pr	*8.94*				*21.3*			13.6	*11.9*			
Nd	*33*				*78.8*			52.4	*41.7*			
Sm	*6.87*				*18.75*			12.45	*8.49*			
Eu	*1.34*				*2.23*			1.3	*0.86*			
Gd	*7.01*				*21*			13	*8.8*			
Tb	*1.13*				*4.17*			2.29	*1.55*			
Dy	*6.21*				*25.5*			12.85	*9.16*			
Ho	*1.36*				*5.65*			2.86	*2.09*			
Er	*4.04*				*17.25*			8.53	*6.15*			
Tm	*0.61*				*2.79*			1.34	*0.96*			
Yb	*4.19*				*18.85*			8.31	*6.01*			
Lu	*0.64*				*3.01*			1.31	*0.9*			
Pb	*6*		23	43	*<5*	21		10	*17*	22	39	19
Ni	*36*	33	18	26	*8*	22	14	9	*5*	6	17	35
Cr	*80*	95	14	7	*20*	23	1	10	*10*	4	9	2

Explanation: $Fe_2O_3^t$ — Total iron as Fe_2O_3. The analyses are ordered according to silica contents with ICP-MS analyses (Acme, Canada) given in italic. G — Granite, QD — Quartzdiorite, RD — Rhyodacite, RH — rhyolite.

(e.g. Nb = 14–80 ppm, Zr = 300–800 ppm, Th = 13–36 ppm) than in the rocks of the basic suite (e.g. Nb < 15 ppm, Zr < 250 ppm, Th < 10 ppm).

Chondrite- and primitive mantle-normalized multi-element diagrams for rare earth elements (REE) and incompatible trace elements, respectively, are shown in Fig. 6a–f. Only samples that were analysed by ICP-MS method are plotted and they are compared with data reported by Ustaszewski et al. (2009). The patterns of the basic suite are compared with those shown by basalts from known geotectonic settings (normal mid ocean-ridge basalts — NMORB; enriched mid ocean-ridge basalts — EMORB and ocean island basalts — OIB) (Hofmann 1997), whereas the rocks of the acid suite are compositionally compared with intra-ophiolite acid rocks of the Eastern Vardar Zone (Šarić et al. 2009) and rhyolites from the Torfajökull and Ljósufjöll volcanic fields (South Iceland Volcanic Zone; Martin & Sigmarsson 2007). The data reported in this study show very similar patterns to those of the north Kozara samples reported by Ustaszewski et al. (2009). The basic suite rocks display relatively flat or slightly LREE- and LILE-enriched patterns with low to moderate Eu and Sr negative anomalies (Fig. 6a,b). Their normalized patterns REE and trace element patterns are most similar to the pattern of EMORB. The samples of the acid suite are characterized by LREE- and LILE-enriched chondrite- and primitive mantle-normalized patterns, respectively, and exhibit pronounced Eu and Sr negative anomalies (Fig. 6c–f). Four samples of the north Kozara acid rocks were analysed by

ICP-MS. Two of them are from the Si-rich lavas near Gornji Podgradci and the other two are leucocratic dykes that cut the sheeted dyke complex of Trnova. Both subgroups show subparallel normalized REE and trace element patterns with only differences in lower Rb and Ba contents in the leucocratic dykes. Compared to ophiolite-related granitoid rocks of the Upper Jurassic Eastern Vardar Zone (Šarić et al. 2009), the north Kozara acid rocks are similar to the rocks interpreted as oceanic plagiogranites and partly to those believed to be produced by obduction-induced melting (Fig. 6e,f). By contrast, they differ from the presumed pre-collisional granites in having much higher contents of heavy REE (HREE) and higher concentrations of high field strength elements (Zr, Hf, Nb, Ta, and Y). Most chondrite- and primitive mantle-normalized values of the north Kozara acid rocks are within the range shown by Pleistocene and Holocene peralkaline rhyolites from Torfajökull and Ljósufjöll (Martin & Sigmarsson 2007). The only difference is found in the higher contents of Nb-Ta and LREE in the samples from Iceland.

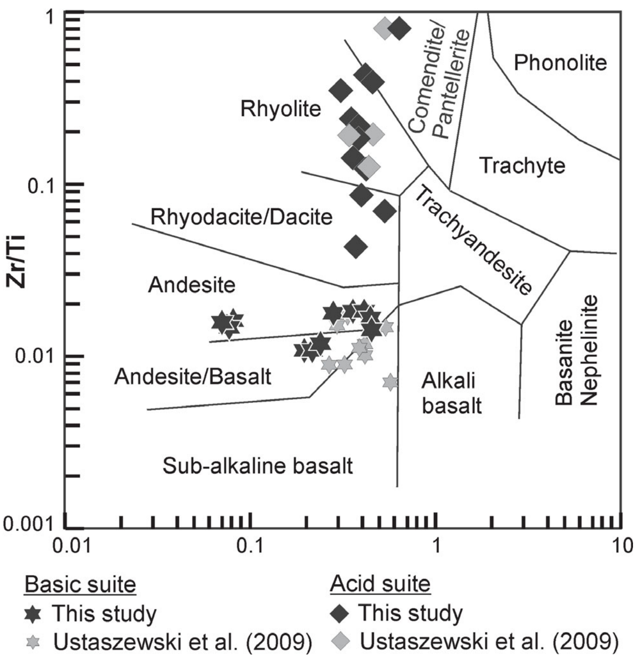

Fig. 3. The Nb/Y and Zr/Ti diagram of classification of volcanic rocks (Winchester & Floyd 1977).

Discussion

Ustaszewski et al. (2009) provided the most detailed geological reconstruction so far of the entire Kozara ophiolite complex. They unequivocally proved earlier suggestions that the north and south Kozara ophiolites do not represent a single ophiolite unit (Pamić 2002; Karamata et al. 2005). They based their conclusions on the following grounds: i) the north Kozara ophiolite is Upper Cretaceous, whereas the south Kozara ophiolite is Upper Jurassic in age (Karamata et al. 2000, 2005), ii) the north Kozara has characteristics of a bimodal association (e.g. Šparica & Buzeljko 1984; Karamata et al. 2000), and 3) the north and the south Kozara basic rocks show different trace element patterns. Moreover, Ustaszewski et al. (2009) suggested that the north Kozara ophiolite represents a

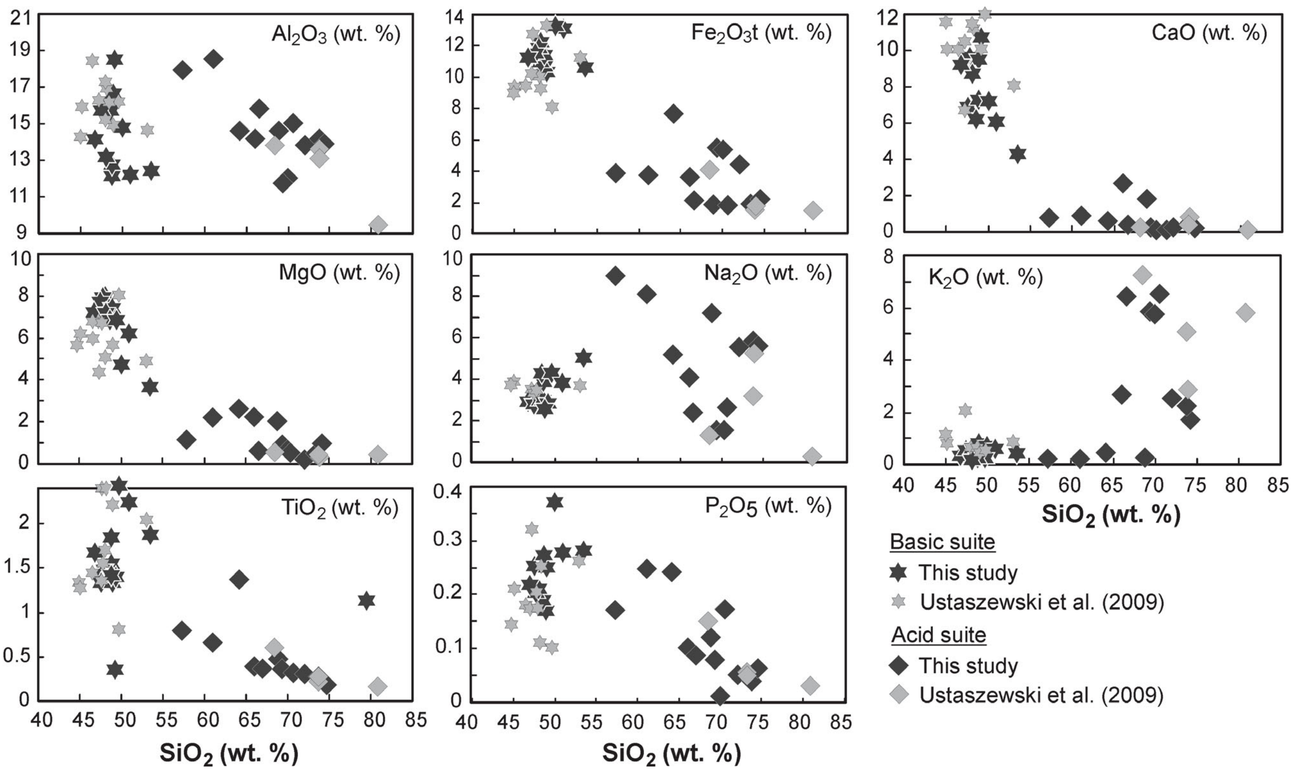

Fig. 4. Harker's diagrams of variations of major oxides with silica contents for the rocks of the basic and the acid suite of north Kozara.

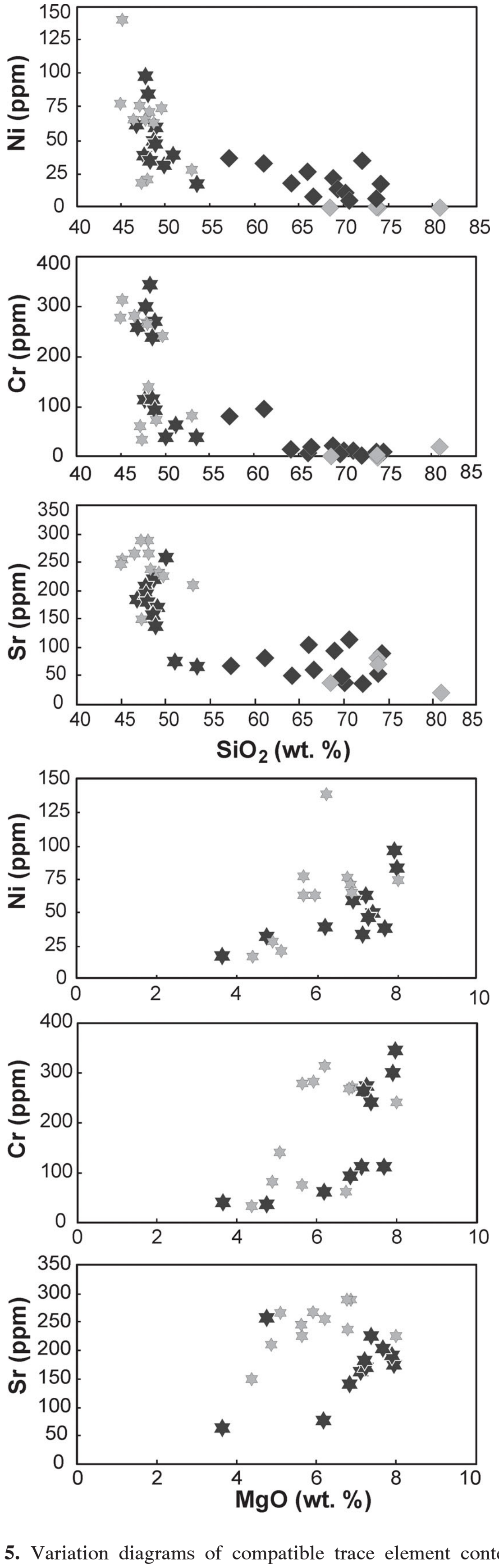

Fig. 5. Variation diagrams of compatible trace element contents with MgO contents as index of differentiation for the north Kozara basic suite. Symbols as for Fig. 4.

slice of an intra-oceanic island lithosphere. According to this interpretation, the north Kozara ophiolite would be a part of an oceanic plateau that was formed during the latest Cretaceous within the Sava-Vardar Ocean and later tectonically emplaced within a complex suture between the north Dinarides and the southern Tisia (e.g. Schmid et al. 2008). However, for corroborating this hypothesis geochemical evidence was used only to a limited extent. Ustaszewski et al. (2009) stated only that the basic rocks of north Kozara cannot be derived from MORB or island arc settings emphasizing LREE-enrichments and 'no significant depletion of HFSE', as well as εNd(T) and initial $^{87}Sr/^{86}Sr$ isotopic values ranging from +4.4–+6.3 and from 0.70346–0.70507 respectively. Although they generally left open whether north Kozara formed in an ocean-island or back-arc basin setting, they favoured the former scenario by the fact that the Maastrichtian and younger sediment cover of north Kozara is represented by abundant alluvial (i.e. above sea-level) facies. In the following discussion we first discuss some aspects of the geochemical/geotectonic affinity of both basic and acid rock suites and, then, we explore a possible petrogenetic relationship between them.

The north Kozara basic suite revisited: OIB or EMORB setting?

Geochemical data shown in this study suggest that the source of primary magmas of the north Kozara basic suite is certainly more enriched than a depleted MORB-like mantle. This conclusion is robust even taking into account that the studied rock samples, including those reported by Ustaszewski et al. (2009), have low contents of MgO (≤8 wt. %), Cr (<350 ppm) and Ni (<100 ppm). Such compatible element concentrations are lower than the values of primitive magmas that would directly originate by partial melting of mantle material (Roeder & Emslie 1970; Sato 1977). The effects of differentiation, presumably fractionation processes are also evident from negative correlations between SiO_2 and MgO and CaO contents (Fig. 4) and the positive ones between MgO and Ni, Cr, and Sr contents (Fig. 5). This, along with the observation that their normalized trace element patterns have a weak Eu anomaly, indicates that the north Kozara basic rocks crystallized from magmas that underwent some crystal fractionation. This fractionation was most likely controlled by removal of olivine and pyroxene, whereas the accumulation of plagioclase was subordinate. A geochemical quantification of fractional crystallization processes is beyond the scope of this study. However, for this discussion it is very important to understand how much these differentiation processes could have changed the geochemical signature of the primary melts.

As was already mentioned, the north Kozara basic rocks are compositionally more similar to EMORB and OIB than to MORB. Fig. 7a shows that there is a clear negative correlation between Nb concentrations and Ni contents, the latter taken as an index of fractionation. This correlation suggests that processes of fractional crystallization likely produced the increase of absolute abundances of highly incompatible elements. However, the fractionation processes could not have significantly affected the ratios between incompatible trace elements of similar or slightly different partition coefficients. Fig. 7b

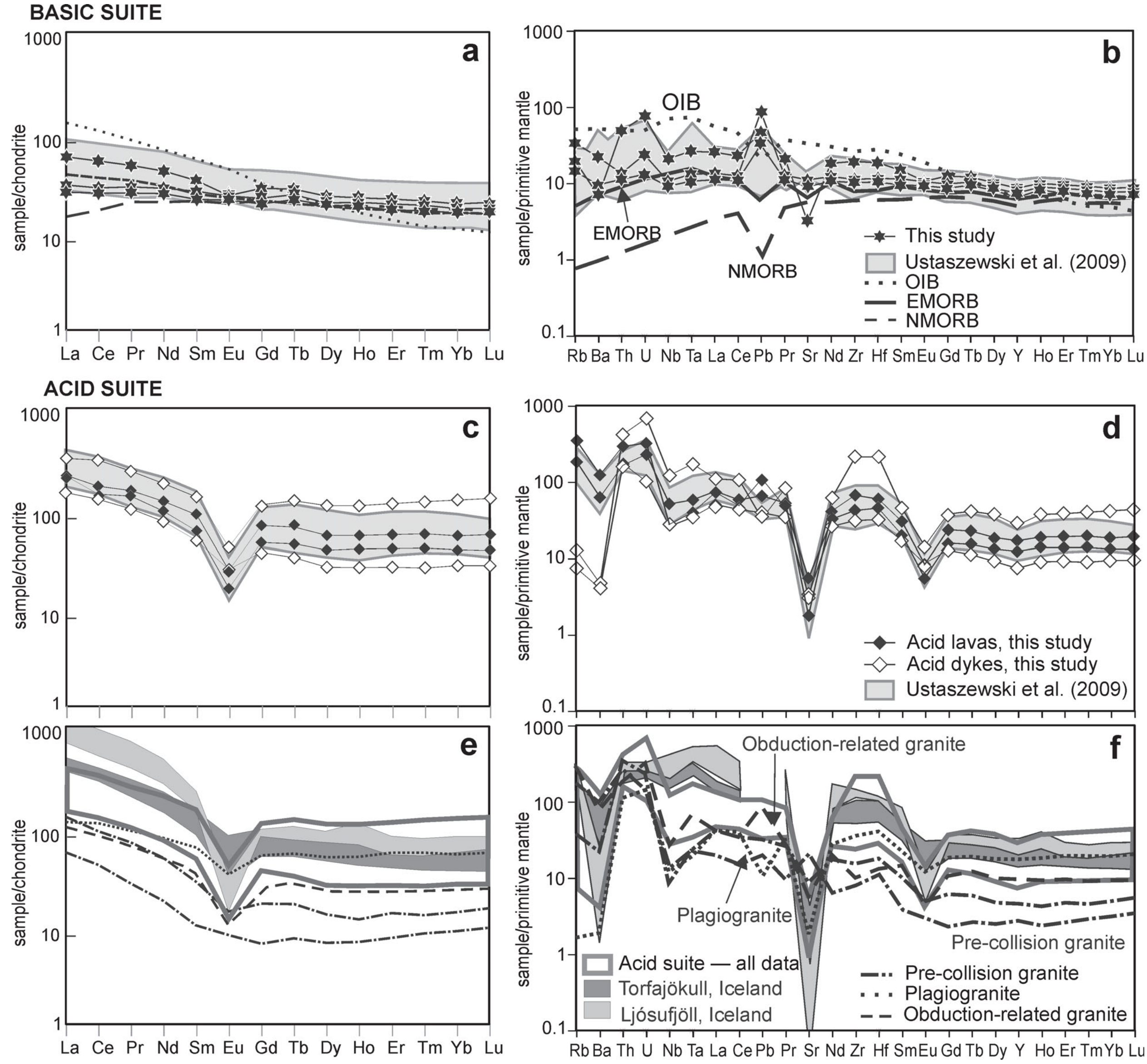

Fig. 6. Chondrite-normalized REE (a, c, e) and primitive mantle-normalized incompatible trace element patterns (b, d, f) for north Kozara igneous rocks. Data previously reported by Ustaszewski et al. (2009) are also given. The patterns of NMORB, EMORB and OIB are from Hofmann (1997), those for obduction-related granites of the Eastern Vardar Zone are from Šarić et al. (2009) and for rhyolites from the Torfajökull & Ljósufjöll volcanic fields (South Iceland Volcanic Zone) are from Martin & Sigmarsson (2007). Coefficients of normalization are from McDonough & Sun (1995).

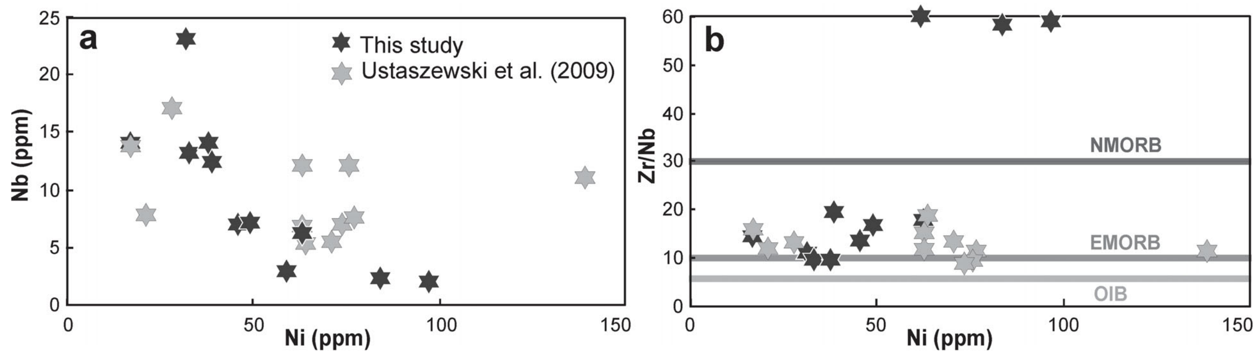

Fig. 7. Diagram of variations between Ni contents and Nb contents and Nb/Zr ratios. The values of NMORB, EMORB and OIB are from Hofmann (1997).

shows that although Nb concentrations generally increase with nickel concentrations, Zr/Nb ratios remain unaffected. Such roughly uniform Zr/Nb ratios suggest that they were mostly insensitive to partial melting and fractionation processes, and, importantly, that this ratio can be used to elucidate the mantle source geochemistry. Most basic rocks of north Kozara have Zr/Nb ratios varying ~15 and that is close to primordial mantle values (McDonough & Sun 1995). These values are slightly higher than the average EMORB Zr/Nb ratio of ~10, but distinctively lower and higher from the average Zr/Nb ratios of NMORB (~30) and OIB (~6), respectively. Three samples analysed by XRF show remarkably low Nb contents (2–3 ppm) and, therefore, they have very high Zr/Nb ratios (~60). Apart from possible analytical problems (i.e. higher detection limits of XRF analyses), potential explanations can be that these rocks either crystallized from magmas originating from a strongly depleted upper mantle, or they do not compositionally represent melts but crystal cumulates.

The previous discussion indicates, first, that the conclusion of Ustaszewski et al. (2009) that the basic rocks of north Kozara did not originate from an NMOR environment or from volcanic arc settings is robust even taking into consideration possible effects of fractionation processes. The prediction that this ophiolite segment formed in an OIB setting should, at least, be reconsidered on the basis of our new data. The above presented values of the Zr/Nb ratio indicate that the north Kozara primary magmas formed by partial melting of an EMORB-like source or, more likely, of a mixture between EMORB and NMORB mantle sources. This conclusion has certain geodynamic significance because it would mean that the ophiolitic segment of the north Kozara represents part of an anomalous mid-ocean ridge or, alternatively, an island plateau that was presumably situated not far from the ridge.

The origin of acid magmatism: A petrogenetic link to the basic suite

In general, the acid magmatic rocks that are spatially related to ophiolites are most frequently represented by so-called oceanic plagiogranites (Coleman & Peterman 1975; Pedersen & Malpas 1984; Floyd et al. 1998). Alternatively, they correspond to granitoids and their volcanic equivalents that result from pre- or post-collisional geodynamic events (Brown & D'Lemos 1991; Li & Li 2003; Kamei 2004; Karsli et al. 2007). However, the intra-ophiolitic acid rocks of north Kozara show important differences with respect both to the plagiogranites and the pre- to post-collisional granites. First, the north Kozara acid rocks have greater relative abundance in respect to mafic products than the intra-ophiolitic acid rocks of any known ophiolite segment of the Balkan Peninsula, and, second, they have a much larger proportion of volcanic products.

Šarić et al. (2009) gave a detailed geochemical and Sr-Nd-Pb overview of granitoids and related acid/intermediate rocks that occur within the Upper Jurassic East Vardar Zone ophiolites. Apart from oceanic plagiogranites, the authors identified two additional types of intra-ophiolite granitoids and interpreted them as originating from either pre-collisional subduction-related magmas or from magmas which formed due to obduction-induced melting of various protoliths. The authors suppose that the latter event was associated with the ophiolite emplacement. As already demonstrated, the normalized trace element patterns of the north Kozara acid rocks are generally similar to those shown by the oceanic plagiogranites and partly to granites deriving from obduction-induced melting (Fig. 6). The geochemical similarity between the north Kozara acid rocks and the plagiogranites occurring in Upper Jurassic ophiolites of the Eastern Vardar Zone is present, although the former do not have the trondhjemitic composition, typical for oceanic plagiogranites (Coleman & Peterman 1975; Coleman & Donato 1979). Plagiogranites are generally interpreted as originating from: i) extensive fractionation of parental tholeiitic magma (e.g. Montanini et al. 2006), ii) partial melting of basaltic protolith (e.g. Pedersen & Malpas 1984), or iii) liquid immiscibility processes (e.g. Dixon & Rutherford 1979). However, irrespectively of their true origin, most authors agree that there is a tight petrogenetic link between the plagiogranites and the host basic rocks and a similar hypothesis can be postulated for the north Kozara acid suite (see also Ustaszewski et al. 2009). The geochemical similarity of the north Kozara acid rocks and the obduction-related East Vardar granitoids may be used to support such a hypothesis, because obduction of hot ophiolites is capable of producing melts of various underlying protoliths (Cox et al. 1999; Whitehead et al. 2000). If the protoliths are basic rocks, than the resulting partial melts can easily have geochemical characteristics similar to oceanic plagiogranites and, therefore, to the north Kozara acid suite rocks, as well.

The genetic relationship between the two suites of north Kozara is also inferred by comparing the north Kozara acid rocks with the modern acid volcanic rocks of Iceland. As previously shown in Fig. 6, there is a general similarity between the normalized REE and trace element patterns between the north Kozara and rocks of the South Iceland Volcanic Zone (Martin & Sigmarsson 2007). Moreover, these two groups of acid rocks are similar because: i) both are part of bimodal igneous associations with relative abundance of acid igneous rocks in excess of 1 %, ii) there is a vast predominance of volcanic products within the acid suite, and iii) in both regions the basic rocks mostly have an EMORB (±NMORB) geochemical signature. These petrological and geochemical similarities imply that both acid suites can have a similar origin. In general, the origin of the Icelandic acid rocks is explained by extensive fractionation of primary basic tholeiitic melts (e.g. Prestvik et al. 2001) or by direct partial melting of altered gabbros and amphibolites in the oceanic crust (e.g. Sigmarsson et al. 1991; Martin & Sigmarsson 2007).

Diagrams Rb vs Ba*/Ba and Rb vs Sr*/Sr (Fig. 8) show fields reflecting chemical compositions of acid rocks originating by crystal fractionation of basic magmas (grey field) and those formed by partial melting of hydrated basaltic oceanic crust (stippled field). The Ba*/Ba and Sr*/Sr ratios represent a quantification of strontium and barium anomalies, respectively (see figure caption for details). Most samples of silica rich lava of north Kozara plot within the field of melts formed by 1–10 % partial melting of altered basaltic crust. This field overlaps with the field of melts produced by fractionation of basaltic magmas but the required amounts of fractionation are far too high (>80 %) to be considered possible. More-

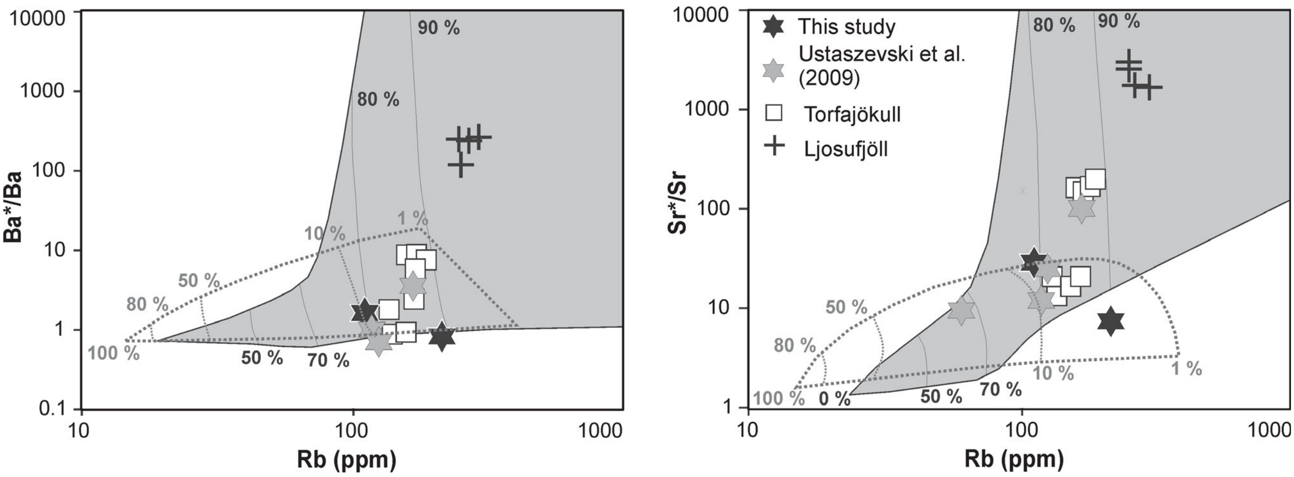

Fig. 8. Rb vs Ba*/Ba (a) and Sr*/Sr (b) for distinguishing melts formed by fractional crystallization of basic primitive magma (grey field) and partial melting of oceanic crust material (stippled field) using the example of Iceland bimodal associations (Martin & Sigmarsson 2007). Ba* is defined as $10^{(\log[Rb]+\log[Nb])/2}$ and Sr* as $10^{(\log[Ce]+\log[Nd])/2}$. All concentrations are normalized to primitive mantle values using the coefficients of Sun & McDonough (1989).

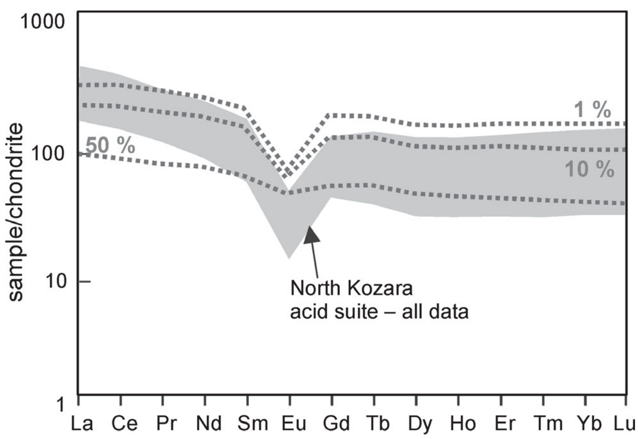

Fig. 9. REE batch melting model of partial melting of a basic protolith. Average REE contents of the north Kozara basic suite is adopted for the geochemical proxy of the protolith. The residual mineralogy is plagioclase (~0.5), clinopyroxene (~0.4), spinel (~0.4), olivine (0.05) and apatite (0.05). For partial melting the formulae of Shaw (1970) is used. Coefficients of normalization on chondritic composition are from McDonough & Sun (1995).

over, the north Kozara rocks are compositionally similar to the Torfajökull rhyolites and are likely to have a similar origin (Martin & Sigmarsson 2007). Similar scenarios for explaining bimodal associations were, at least partly, proposed for the Katla (Lacasse et al. 2007), Askja and Öræfajökull volcanoes (Prestvik 1980; Sigurdsson & Sparks 1981).

In order to further corroborate this hypothesis we performed REE batch melting modelling (Fig. 9). The model demonstrates that the chondrite-normalized REE patterns of the north Kozara acid rocks can generally be produced by melting of 1–10 % of basic rocks. The average REE contents of the north Kozara basic suite is adopted for the proxy of the source rock composition. The residual mineralogy in the model is plagioclase (~0.5), clinopyroxene (~0.4), spinel (~0.4), and traces of olivine and apatite. This assemblage corresponds to the near-solidus mineralogy obtained by calculations of equilibrium crystallization using the MELTS thermodynamic approach of Ghiorso & Sack (1995). The calculations assume a starting composition of the average sample of the north Kozara basic suite with 2.5 wt. % H_2O, $O_2f=+3$ (QMF) and temperature and pressure of 850 °C and 1 kb, respectively. This assumes that the source was similar to hydrothermally altered oceanic crust because it is generally accepted that fresh tholeiitic protoliths are not likely to produce rhyolitic magmas with more than 3 % K_2O (Beard & Lofgren 1989; Thy et al. 1990).

The above discussion does not unequivocally demonstrate that all the north Kozara acid rocks originated by partial melting of altered oceanic crust as suggested by modelling. It is likely that at least some leucocratic dykes that intrude diabase sheeted complexes represent small volume trondhjemitic melts, similar to those found in many ophiolites worldwide (Coleman & Peterman 1975; Pedersen & Malpas 1984).

Volcanological constraints

One of the most striking features of the north Kozara ophiolites is the presence of large masses of primary glass-rich silicic lavas. Although later erosion events and low-temperature alteration and weathering processes could have obliterated some primary features of these rocks, there is solid evidence that they likely originated from subaerial high-temperature lava flows.

Most acid volcanic rocks of north Kozara contain few phenocrysts or are almost aphyric, implying that this acid magma was emplaced at temperatures close to the liquidus temperature. A primary glass-rich nature of these rocks is principally inferred from evidence of high-temperature devitrification processes, which is a typical feature of glassy rhyodacite/rhyolite lavas. There are samples with preserved classical perlite (Allen 1988) and microspherulitic textures, which are partly obliterated by subsequent recrystallization to microcrystalline mosaic quartz aggregates. The presence of micropoikilitic or so-called snow-flake texture (Andersen 1969; Lofgren 1971), in the form of patchy quartz crystals enclosing laths of alkali feldspar, strongly suggests that these lavas emplaced at high temperatures and underwent slow cooling devitrification (Ryan & Sammis 1981; Manley 1992; Orth & McPhie 2003). Such slow cooling is likely associated with subaerial emplacement and that is also supported by the lack of hyaloclastic deposits.

Concluding remarks

The discussion presented above allows us to derive four major conclusions. First, the rocks of the basic suite of the north Kozara have an E-MORB geochemical signature. Second, there is a close petrogenetic link between the basic and acid/intermediate suite. Third, the acid magmas most probably originated through partial melting of hydrated oceanic crust, similar in composition to the rocks of the basic suite. And fourth, the most voluminous acid magma most likely emplaced as subaerial high-temperature rhyodacite/rhyolite lava.

The most important geodynamic implication provided by these conclusions is that the entire north Kozara ophiolite slice could represent the remnant of an anomalous ridge segment that is similar to present day Iceland. The above recognition that the north Kozara acid volcanic originated by partial melting of basic rocks is tightly related to the thermal state of the oceanic crust. Partial fusion of hydrated basaltic material in shallow crust is only possible in regions with elevated geothermal gradient (Sigmarsson et al. 1991, 1992). This is so because a cold crust generally needs a larger input of heat to reach its solidus, while a hotter crust is more readily melted. The high-temperature emplacement of the north Kozara acid lavas further supports this opinion. It can imply that the rhyodacitic magma had travelled through hot oceanic crust and reached the surface relatively fast, because of a large density contrast in combination with relatively low viscosity. The presented evidence that is provided by the study of acid rocks, is in accordance with a typical E-MORB geochemical signature of the host basic volcanic and shallow intrusive.

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