Geochemical implications for the magma origin of granitic rocks from the Ditrău Alkaline Massif (Eastern Carpathians, Romania)

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ABSTRACT

In addition to a series of ultramafic to mafic and alkaline igneous rocks, a granite body also occurs in the Ditrău Alkaline Massif, Eastern Carpathians, Romania. We present and discuss mineral chemical data, and major and trace element compositions of the granites in order to define their nature and origin and to determine the depth of the magma emplacement. The granites consist of K-feldspar, albite to oligoclase and quartz accompanied by Ti-rich annite ± calcic amphiboles. Depending on the amphibole content they are classified as less fractionated amphibole-bearing and amphibole-free varieties. Accessories include zircon, apatite, magnetite, ilmenite, and allanite or monazite. High Zr, Nb, Ga, Ce and Y content and Ga/Al and Fe/Mg ratios, together with low CaO, Sr and Ba contents and Y/Nb ratios of 0.04-0.88 are consistent with A1-type granites and mantle differentiates correspond to an intra-plate environment. The Ditrău Alkaline Massif granites were emplaced at middle – upper crustal levels between 14 and 4 km depth as indicated by the calculated crystallization pressure of 370 ± 40 MPa and the stability limit of calcic amphiboles.

Keywords: A-type granite; geochemistry; mantle differentiates; amphibole geobarometry; Ditrău Alkaline Massif, Eastern Carpathians, Romania

1. INTRODUCTION

Since the nineteenth century many studies have examined the mineralogy, petrology and geochemistry of the Ditrău Alkaline Massif (DAM) (e.g. KOCH, 1879; IANOVICI, 1938; STRECKEISEN, 1938, 1952, 1954; CODARCEA et al., 1957; STRECKEISEN & HUNZIKER, 1974; ANAS-TASIU & CONSTANTINESCU, 1982; PÁL-MOLNÁR, 1992, 1994a, 2000; PÁL-MOLNÁR & ÁRVA-SÓS, 1995; DALLMEYER et al., 1997; KRÄUTNER & BINDEA, 1998; JAKAB, 1998; MOROGAN et al., 2000; FALL et al., 2007; BATKI et al., 2014), however, there is still a great debate on the petrogenesis of the DAM.

A wide variety of igneous rocks have been described in the DAM from ultramafic to mafic ones (Tarnița Complex: peridotites, gabbros, diorites), felsic silica-saturated and oversaturated syenites and granites, as well as undersaturated alkaline rocks (nepheline syenites) (PÁL-MOLNÁR, 2000). The massif is the locus typicus of several magmatic rock types that were first identified here, e.g. ditróite, orotvite and ditró-essexite. Previously these names were widely accepted in the international petrographical literature, though by now the IUGS does not recommend their use. Numerous dykes, including lamprophyres, tinguaites and alkali feldspar syenites, cut across the whole complex (BATKI et al., 2014).
The north-eastern part of the DAM was previously considered to be a homogeneous granite body (JAKAB, 1998). However, KOVÁCS & PÁL-MOLNÁR (2005) and PÁL-MOLNÁR (2006) pointed out that there are various types of felsic granitic rocks in this area. Currently, two main hypotheses have emerged concerning the origin of the granite: (1) granites have resulted from the differentiation of mantle-derived melts (MOROGAN et al., 2000; PÁL-MOLNÁR, 2000) or (2) granites have been formed from silica-poor magmas contaminated by the felsic crust (STRECKEISEN & HUNZIKER, 1974; JAKAB, 1998), without mentioning any particular source of these magmas.

In this paper we discuss new geochemical and petrological data on granites and draw conclusions on processes of magma evolution in order to provide further constraints on the formation of the Ditrău Alkaline Massif.

2. GEOLOGICAL SETTING

The Ditrău Alkaline Massif forms the southern and south-western part of the Giurgeu Mountains (Eastern Carpathians, Romania). It is 19 km long and 14 km wide and ca. 200 km² in size on the surface (PÁL-MOLNÁR, 2000) (Fig. 1). In the structural system of the Alpine-Carpathian-Dinaric region it belongs to the Dacia Mega-Unit (Median Dacides; SĂNDULESCU, 1984) (Fig. 1A). The massif intruded the Variscan metamorphic rocks of the Eastern Carpathians, later participating in the Alpine tectonic events along with these metamorphic rocks (PÁL-MOLNÁR, 1994b). Structurally, the DAM is the part of the Alpine Bucovinian Nappe System having direct contact with three of its Pre-Alpine (Variscan) lithogroups: the Bretila (Rarău Nappe), Rebra (Rodna Nappe) and Tulgheș lithogroups (Putna Nappe) (BALINTONI, 1997, 1981) (Fig. 1B). The Bucovinian Nappe represents the upper unit of the Central Eastern Carpathian nappes which were formed during the Middle Cretaceous (SĂNDULESCU, 1984). The DAM is partly covered by Neogene-Quaternary andesitic pyroclastics and lava flows of the Călimani–Gurghiu–Harghita volcanic chain and by Pliocene–Pleistocene sediments and lignite-bearing lacustrine deposits of the Gheorgeni and Jolotca Basins (CODARCEA et al., 1957).

It is most likely that the intrusion was related to the opening events of the Meliata–Hallstatt Ocean (HOECK et al., 2009), where main rifting began during the Pelsonian Substage (Middle Triassic) (KOZUR, 1991).
Mineral phases of the studied granites were analyzed with a JEOL JXA-733 electron microprobe in wavelength-dispersive mode using a beam current of 15 nA and an acceleration voltage of 20kV at the Institute for Geological and Geochemical Research, Research Centre for Astronomy and Earth Sciences, Hungarian Academy of Sciences, Budapest, Hungary.

Direct contact of the massif with sedimentary rocks is not observed. The first K/Ar ages were published by BAGDASARIAN (1972) who determined a Neocomian (125±10 Ma) age of the DAM granites. Afterwards, PÁL-MOLNÁR & ÁRVA-SOÓS (1995) using K/Ar ages on amphibole and biotite separates, produced an average date of 206±7.8 Ma for the granites and suggested a late Triassic age for them.

The largest granite body crops out in the north-eastern part of the DAM, east of the Turcului Creek and north of the Jolotca Creek (Fig. 1C).

Mineral phases of the studied granites were analyzed with a JEOL JXA-733 electron microprobe in wavelength-dispersive mode using a beam current of 15 nA and an acceleration voltage of 20kV at the Institute for Geological and Geochemical Research, Research Centre for Astronomy and Earth Sciences, Hungarian Academy of Sciences, Budapest, Hungary.

4. PETROGRAPHY AND MINERAL CHEMISTRY

4.1. Petrography

The studied granites are generally light grey with a light reddish tone. The rocks are inequigranular and exhibit a phaneritic texture. The main constituents are quartz (20–37%), K-feldspar (up to 50%), plagioclase (ca. 10–35%) and subordinate biotite ± amphibole (ca. 2–16%) (Fig. 2). Amphibole is frequently altered and occurs in half of the granite samples. The presence of amphibole in the granite does not depend on the location; amphibole-bearing granite occurs randomly throughout the granite body. Chlorite, epidote, magnetite and titanite as alteration products commonly appear after amphibole (Fig. 2A). Quartz is observed in the rocks as a medium- to coarse-grained anhedral phase (250–1500 µm). Plagioclase is usually
Table 1: Representative biotite compositions of granites from the Ditrău Alkaline Massif, Romania.

| Sample | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 | VRG6835 |
|--------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|
|        | 1       | 2       | 3       | 4       | 5       | 6       | 7       | 8       | 9       | 10      | 11      | 12      | 13      | 14      |
| SiO₂   | 35.30   | 36.60   | 35.41   | 36.32   | 36.25   | 38.14   | 36.31   | 35.21   | 36.55   | 36.77   | 36.83   | 37.40   | 37.29   | 37.36   | 35.94   |
| TiO₂   | 2.66    | 1.35    | 1.99    | 2.11    | 2.53    | 2.44    | 2.59    | 3.17    | 3.00    | 3.40    | 2.99    | 3.13    | 2.74    | 3.23    | 2.96    |
| Al₂O₃  | 12.95   | 13.38   | 13.55   | 12.95   | 13.84   | 13.47   | 13.21   | 13.32   | 13.59   | 13.14   | 13.59   | 13.01   | 13.34   | 12.61   | 12.18   |
| FeO    | 22.86   | 23.06   | 23.04   | 22.94   | 22.93   | 23.20   | 24.11   | 23.29   | 23.44   | 24.88   | 24.72   | 21.51   | 21.23   | 21.87   | 20.67   |
| MnO    | 0.82    | 0.78    | 0.84    | 0.84    | 0.84    | 0.84    | 0.80    | 0.84    | 0.82    | 0.39    | 0.54    | 0.49    | 0.44    | 0.51    |
| MgO    | 8.66    | 8.80    | 8.42    | 8.26    | 8.00    | 8.71    | 8.04    | 7.22    | 7.47    | 7.31    | 7.59    | 9.63    | 9.19    | 9.24    | 9.92    |
| CaO    | 0.02    | 0.06    | 0.12    | 0.00    | 0.03    | 0.00    | 0.02    | 0.00    | 0.00    | 0.14    | 0.03    | 0.00    | 0.00    | 0.10    |
| Na₂O   | 0.53    | 0.03    | 0.18    | 0.15    | 0.03    | 0.34    | 0.11    | 0.21    | 0.21    | 0.07    | 0.00    | 0.33    | 0.12    | 0.40    |
| K₂O    | 9.38    | 9.94    | 9.38    | 9.34    | 9.76    | 9.95    | 10.00   | 9.60    | 9.93    | 9.61    | 9.89    | 9.62    | 9.75    | 9.79    | 9.47    |
| Total  | 93.15   | 94.00   | 92.93   | 92.67   | 94.21   | 96.66   | 95.42   | 92.76   | 94.81   | 96.35   | 86.33   | 94.87   | 94.36   | 94.66   | 92.15   |
| Oxygens| 22      | 22      | 22      | 22      | 22      | 22      | 22      | 22      | 22      | 22      | 22      | 22      | 22      | 22      | 22      |
| Si     | 5.67    | 5.81    | 5.70    | 5.83    | 5.74    | 5.86    | 5.72    | 5.69    | 5.76    | 5.74    | 5.74    | 5.81    | 5.83    | 5.84    | 5.77    |
| Al<sup>IV</sup> | 2.32 | 2.18 | 2.29 | 2.16 | 2.25 | 2.13 | 2.27 | 2.30 | 2.23 | 2.25 | 2.25 | 2.18 | 2.16 | 2.15 | 2.22 |
| Al<sup>V1</sup> | 0.13 | 0.32 | 0.27 | 0.29 | 0.32 | 0.29 | 0.18 | 0.24 | 0.28 | 0.16 | 0.24 | 0.20 | 0.28 | 0.16 | 0.08 |
| Ti     | 0.32    | 0.16    | 0.24    | 0.25    | 0.30    | 0.28    | 0.30    | 0.38    | 0.35    | 0.39    | 0.35    | 0.36    | 0.32    | 0.37    | 0.35    |
| Fe<sup>2+</sup> | 3.07 | 3.06 | 3.10 | 3.08 | 3.03 | 2.98 | 3.18 | 3.15 | 3.08 | 3.24 | 3.22 | 2.79 | 2.77 | 2.86 | 2.77 |
| Mn     | 0.11    | 0.10    | 0.11    | 0.08    | 0.11    | 0.05    | 0.10    | 0.11    | 0.09    | 0.10    | 0.05    | 0.07    | 0.06    | 0.05    | 0.06    |
| Mg     | 2.07    | 2.08    | 2.02    | 1.97    | 1.88    | 1.99    | 1.89    | 1.74    | 1.75    | 1.70    | 1.76    | 2.23    | 2.14    | 2.15    | 2.37    |
| Ca     | 0.00    | 0.01    | 0.02    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.01    | 0.03    | 0.02    | 0.00    | 0.00    | 0.00    | 0.01    |
| Na     | 0.17    | 0.01    | 0.05    | 0.04    | 0.01    | 0.08    | 0.10    | 0.03    | 0.06    | 0.06    | 0.00    | 0.01    | 0.10    | 0.03    | 0.12    |
| K      | 1.92    | 2.01    | 1.92    | 1.91    | 1.97    | 1.95    | 2.01    | 1.98    | 1.99    | 1.91    | 1.96    | 1.90    | 1.94    | 1.95    | 1.94    |
| Total  | 15.82   | 15.77   | 15.76   | 15.16   | 15.65   | 15.65   | 15.79   | 15.65   | 15.65   | 15.63   | 15.57   | 15.63   | 15.60   | 15.74   |
| mg#    | 0.40    | 0.40    | 0.39    | 0.39    | 0.38    | 0.40    | 0.37    | 0.35    | 0.36    | 0.34    | 0.35    | 0.44    | 0.43    | 0.46    |
| Fe/(Fe+Mg) | 0.60 | 0.60 | 0.61 | 0.61 | 0.62 | 0.60 | 0.63 | 0.64 | 0.64 | 0.64 | 0.56 | 0.56 | 0.57 | 0.54 |
| Mg/Fe  | 0.67    | 0.68    | 0.65    | 0.64    | 0.62    | 0.67    | 0.59    | 0.55    | 0.57    | 0.52    | 0.55    | 0.80    | 0.77    | 0.75    | 0.86    |
euhedral to subhedral and is fine- to coarse-grained (600–1000 µm). It shows albite twinning and often has a sericitized core. The medium- to very coarse-grained K-feldspar (500–4000 µm) is microcline occurring as tartan twins. Exsolution textures are commonly represented by perthite and equilibrium quartz-feldspar intergrowth (i.e. myrmekite) occurring mainly in amphibole-bearing samples. Myrmekite texture is developed at the margins of plagioclase and microcline. Worm- or finger-like droplets of quartz are enclosed in plagioclase. Biotite appears as discrete subhedral to anhedral grains, as well as grain aggregates interstitial to feldspars and quartz (Fig. 2B). It is often altered to chlorite, opaque minerals and hematite. Accessory phases are apatite, zircon, monazite, ilmenite, magnetite and allanite. Allanite generally occurs as euhedral to subhedral crystals interstitial to the major minerals (Fig. 2C). It is strongly pleochroic (from red to brownish red). Zircon (Fig. 2B) and apatite are usually euhedral and are enclosed in biotite and feldspars. Monazite is euhedral and incorporated in biotite or feldspars (Fig. 2D). Magnetite occurs as euhedral crystals and enclosed in amphiboles. Allanite solely appears in granites which contain amphibole, whereas monazite appears only in amphibole-free varieties.

4.2. Mineral chemistry

4.2.1. Biotite

Representative microprobe analyses of biotites are given in Table 1. They are annite according to DEER et al. (1992). There are two types of biotites in the studied granites, one with higher mg# of 0.42–0.46 (Mg/Fe=0.75–0.86) and the other one with lower mg# of 0.34–0.40 (Mg/Fe=0.52–0.68). The high Ti content (TiO₂=1.4–3.4 wt. %) is a characteristic feature of biotites, and is similar to the biotites of other rocks from the Ditrău Alkaline Massif (PÁL-MOLNÁR, 2000; MOROGAN et al., 2000; BATKI et al., 2014). Mg contents slightly decrease with increasing AlVI (Table 1).

4.2.2. Amphibole

Representative chemical compositions of amphiboles are shown in Table 2. According to LEAKE et al. (1997) they are calcic amphiboles and compositionally vary between ferro-edenite and ferrohornblende (Fig. 3). BCa contents vary from 1.68 to 1.82, while BNa content varies between 0.17–0.31. BNa content shows positive correlation with mg#, while BCa content increases with decreasing mg#.

All amphiboles similar to biotites are iron rich. They have high Fe/(Fe+Mg) ratios of 0.69–0.75. Their AlVI content ranges from 1.20 to 1.35 and the AlVI values vary between 0.00–0.18.

4.2.3. Feldspars

Representative analyses of feldspars are listed in Table 3. Feldspars represent both plagioclase and K-feldspar. Plagioclase is mainly albite with composition of Ab₉₆An₁₃Or₁ to Ab₈₆An₁₃Or₁ (Fig. 4). Zoned plagioclase can not be observed. K-feldspar is orthoclase (Or₉₄-97).
5. MAJOR AND TRACE ELEMENT GEOCHEMISTRY

Geochemical data for the major element composition of the DAM granites (two samples) were previously published by Streckeisen (1954). Morogan et al. (2000) also gave major, trace and REE data of three samples (Table 4). In this study, new analyses for major, trace and REEs include eleven granites.

The DAM granites are high in SiO₂, Al₂O₃, FeOt (up to 3.1 wt. %) and alkalis (Na₂O+K₂O=8.2–11.5 wt. %), and generally low in CaO (Table 4). The MgO content of amphibole-bearing granites varies from 0.47 to 0.84. Mg values are very low for all of the samples. The studied granites are mostly alkaline (Fig. 5A) and strongly peraluminous in composition (ASI>1; Fig. 5B). FeOt, MgO, CaO and TiO₂ decrease with increasing SiO₂ suggesting the fractionation of amphibole and ilmenite (not shown, Table 4). They may also account for the high Nb concentrations (up to 429 ppm). Decreasing amounts of Al₂O₃, P₂O₅ and Zr with increasing SiO₂ could be controlled by the early extraction of allanite, apatite and zircon, respectively. K₂O values are constant with increasing SiO₂ as a
| Sample | VrG6835 | VrG6838 | VrG6839 | VrG6835 | VrG6838 | VrG6839 | VrG6835 | VrG6838 | VrG6839 | VrG6835 | VrG6838 | VrG6839 | VrG6835 | VrG6838 | VrG6839 | VrG6835 | VrG6838 | VrG6839 |
|--------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|
| Mineral | pl1     | pl2     | pl3     | pl4     | pl5     | mc1     | mc2     | mc3     | mc4     | mc5     | mc6     | mc7     | mc8     | mc9     | mc10    | mc11    | mc12    |
| SiO₂   | 68.40   | 67.08   | 65.50   | 66.42   | 64.69   | 64.76   | 65.12   | 64.79   | 65.24   | 65.01   | 65.45   | 65.10   | 65.90   | 65.50   | 65.38   | 66.12   | 66.71   |
| TiO₂   | 0.05    | 0.17    | 0.01    | 0.01    | 0.13    | 0.01    | 0.13    | 0.01    | 0.10    | 0.10    | 0.02    | 0.02    | 0.06    | 0.08    | 0.20    | 0.20    | 0.08    |
| Al₂O₃  | 3.03    | 2.03    | 0.36    | 0.11    | 0.03    | 0.28    | 0.28    | 0.28    | 0.28    | 0.28    | 0.28    | 0.28    | 0.28    | 0.28    | 0.28    | 0.28    | 0.28    |
| FeO    | 0.03    | 0.11    | 0.12    | 0.12    | 0.01    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    |
| MgO    | 0.13    | 0.08    | 0.16    | 0.11    | 0.10    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    | 0.18    |
| CaO    | 11.07   | 12.39   | 11.30   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   | 12.34   |
| K₂O    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    |
| Total  | 99.30   | 99.91   | 100.57  | 99.62   | 99.89   | 99.92   | 99.09   | 99.39   | 99.65   | 99.81   | 99.81   | 99.81   | 99.81   | 99.81   | 99.81   | 99.81   | 99.81   |
| O₂     | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       | 8       |
| Si     | 2.94    | 3.01    | 2.97    | 3.02    | 2.99    | 2.87    | 2.90    | 2.91    | 2.92    | 2.92    | 2.92    | 2.92    | 2.92    | 2.92    | 2.92    | 2.92    | 2.92    |
| Ti     | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    |
| Al     | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    |
| Fe     | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    |
| Mg     | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    |
| Ca     | 0.01    | 0.03    | 0.02    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    |
| K      | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    | 0.01    |
| Total  | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    | 5.01    |

| Oxygens | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  | 8  |
|---------|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|----|
| pl: plagioclase; mc: microcline
Table 4: Whole-rock analyses of granites from the northern part of the Ditrău Alkaline Massif, Romania.

| Location | Sample | Location | Sample | Location | Sample | Location | Sample | Location | Sample |
|----------|--------|----------|--------|----------|--------|----------|--------|----------|--------|
| Laposbükk Creek | Vrg 6835 | Laposbükk Creek | Vrg 6839 | Laposbükk Creek | Vrg 6883/A | Laposbükk Creek | Vrg 7459 | Laposbükk Creek | Vrg 7460 |
| Creangă Mare Creek | Vrg 6847 | Creangă Mare Creek | Vrg 6856 | Creangă Mare Creek | Vrg 6726 | Creangă Mare Creek | Vrg 6842 | Creangă Mare Creek | Vrg 7425/A |
| Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 |
| Location | Sample | Location | Sample | Location | Sample | Location | Sample | Location | Sample |
| Hologag Creek | Vrg 6840 | Magasbükk Creek | Vrg 6842 | Hologag Creek | Vrg 6726 | Hologag Creek | Vrg 6842 | Hologag Creek | Vrg 7425/A |
| MoroGAN et al., 2000 average of 148 samples | STRECKEISEN, 1954 1 | Hologag Creek | Vrg 6840 | Jolotca Creek | Vrg 6842 | Jolotca Creek | Vrg 6726 | Jolotca Creek | Vrg 6842 |
| — | — | — | — | — | — | — | — | — | — |

| Location | Sample | Location | Sample | Location | Sample | Location | Sample | Location | Sample |
|----------|--------|----------|--------|----------|--------|----------|--------|----------|--------|
| Creangă Mare Creek | Vrg 6835 | Creangă Mare Creek | Vrg 6839 | Creangă Mare Creek | Vrg 6883/A | Creangă Mare Creek | Vrg 7459 | Creangă Mare Creek | Vrg 7460 |
| Turcului Creek | Vrg 6856 | Turcului Creek | Vrg 6726 | Turcului Creek | Vrg 6842 | Turcului Creek | Vrg 7425/A | Turcului Creek | Vrg 7458 |
| Creangă Mare Creek | Vrg 6847 | Creangă Mare Creek | Vrg 6856 | Creangă Mare Creek | Vrg 6726 | Creangă Mare Creek | Vrg 6842 | Creangă Mare Creek | Vrg 7425/A |
| Turcului Creek | Vrg 7459 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 |
| Location | Sample | Location | Sample | Location | Sample | Location | Sample | Location | Sample |
| Hologag Creek | Vrg 6856 | Hologag Creek | Vrg 6726 | Hologag Creek | Vrg 6842 | Hologag Creek | Vrg 7425/A | Hologag Creek | Vrg 7458 |
| MoroGAN et al., 2000 average of 148 samples | STRECKEISEN, 1954 2 | Hologag Creek | Vrg 6856 | Jolotca Creek | Vrg 6856 | Jolotca Creek | Vrg 6726 | Jolotca Creek | Vrg 6856 |
| — | — | — | — | — | — | — | — | — | — |

| Location | Sample | Location | Sample | Location | Sample | Location | Sample | Location | Sample |
|----------|--------|----------|--------|----------|--------|----------|--------|----------|--------|
| Creangă Mare Creek | Vrg 6835 | Creangă Mare Creek | Vrg 6839 | Creangă Mare Creek | Vrg 6883/A | Creangă Mare Creek | Vrg 7459 | Creangă Mare Creek | Vrg 7460 |
| Turcului Creek | Vrg 6856 | Turcului Creek | Vrg 6726 | Turcului Creek | Vrg 6842 | Turcului Creek | Vrg 7425/A | Turcului Creek | Vrg 7458 |
| Creangă Mare Creek | Vrg 6847 | Creangă Mare Creek | Vrg 6856 | Creangă Mare Creek | Vrg 6726 | Creangă Mare Creek | Vrg 6842 | Creangă Mare Creek | Vrg 7425/A |
| Turcului Creek | Vrg 7459 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 | Turcului Creek | Vrg 7458 |
| Location | Sample | Location | Sample | Location | Sample | Location | Sample | Location | Sample |
| Hologag Creek | Vrg 6856 | Hologag Creek | Vrg 6726 | Hologag Creek | Vrg 6842 | Hologag Creek | Vrg 7425/A | Hologag Creek | Vrg 7458 |
| MoroGAN et al., 2000 average of 148 samples | STRECKEISEN, 1954 2 | Hologag Creek | Vrg 6856 | Jolotca Creek | Vrg 6856 | Jolotca Creek | Vrg 6726 | Jolotca Creek | Vrg 6856 |
| — | — | — | — | — | — | — | — | — | — |

Mg# (100(MgO/MgO+FeO)); A/NK: Al2O3/(Na2O+K2O); NK/A: (Na2O+K2O)/Al2O3; A/CNK: Al/Ca + Na + K
| Location                      | Laposbükk Creek | Creangă Mare Creek | Turcului Creek | Jolotca Creek |
|-------------------------------|------------------|--------------------|----------------|---------------|
| Sample                        | VrG 6835         | VrG 6839           | VrG 6838/A     | VrG 7459      |
| Whalen et al., 1987           |                  |                    |                |               |
| VrG 6835                     |                  |                    |                |               |
| Location                      | VrG 6836         | VrG 6837           | VrG 7458       |                |
| Laposbükk Creek              |                  |                    |                |               |
| Creangă Mare Creek           |                  |                    |                |               |
| Jolotca Creek                |                  |                    |                |               |
| Weight                        |                  |                    |                |               |
| Values                        |                  |                    |                |               |
| Br                            |                  |                    |                |               |
| Sr                            |                  |                    |                |               |
| Y                             |                  |                    |                |               |
| La                            |                  |                    |                |               |
| Ce                            |                  |                    |                |               |
| Nd                            |                  |                    |                |               |
| Sm                            |                  |                    |                |               |
| Eu                            |                  |                    |                |               |
| Gd                            |                  |                    |                |               |
| Tb                            |                  |                    |                |               |
| Ho                            |                  |                    |                |               |
| Er                            |                  |                    |                |               |
| Tm                            |                  |                    |                |               |
| Dy                            |                  |                    |                |               |
| Yb                            |                  |                    |                |               |
| Lu                            |                  |                    |                |               |
| ΣREE                          |                  |                    |                |               |

*Table 4. (continued)*
result of continuous K-feldspar fractionation during differentiation, but Na$_2$O has a slight negative correlation with SiO$_2$ from 70 w. %, consistent with the late stage of albite crystallization (not shown, Table 4). Ce increases up to an SiO$_2$ content of about 70 w. % and then decreases in the granites, suggesting later fractionation of monazite. V shows the same pattern as TiO$_2$ due to ilmenite fractionation (not shown, Table 4). Amphibole-bearing granites own higher Sr (up to 544ppm) and Ba (up to 816 ppm) contents than the amphibole-free varieties (Sr < 103ppm, Ba < 222ppm).

Primitive mantle-normalized granites display strong negative Ba, Sr and Eu anomalies (Eu/Eu* = 0.15–0.66) (Fig. 6A). The primitive mantle-normalized REE (Fig. 6B) patterns show strong enrichment in LREE and significant fractionation of HREE with an (La/Yb)$_N$ ratio of 13–38 (Table 4). The (La/Yb)$_N$ ratios are influenced by the enrichment of HREE. The rocks have high MREE content, especially Sm and Gd. Ba, Sr, Zr, and Sc decrease with increasing SiO$_2$ (not shown, Table 4). The Y/Nb ratios are up to 0.88.

6. DISCUSSION

6.1. Mantle source and geodynamic implications

Many types of granitoid classification have been proposed in the literature. These classifications divide granitoids mostly on the basis of their petrography (e.g. PUPIN, 1980; LAMEYRE & BOWDEN, 1982; NACHIT et al., 1985; TISCHENDORF & PALCHEN, 1985), geochemistry (CHAPELL & WHITE, 1974; PUPIN, 1980; COLLINS et al., 1982; PEARCE et al., 1984; WHALEN et al., 1987) and tectonic environment (PITCHER, 1983). Essentially all of these classifications broadly correspond to the three possible sources of granitoids: crustal, mantle-derived and mixed (BARBARIN, 1990). The petrogenetic model developed for granitoid rocks by LOISELLE & WONES (1979) and WHITE & CHAPPELL (1983) contain four types of granitoids (S, I-, M-, and A-type) which also overlap with the crustal, mantle-derived and mixed origin of granitoids. The A-type granitoids occur along rift zones (LOISELLE &
According to WHALEN et al. (1987) A-type granites can be separated from I- and S-type granites based on their Ga/Al ratios and the fractionation of accessory phases. The average composition of A-type granites (WHALEN et al., 1987) is also presented in Table 4.

The DAM granites exhibit high Zr+Nb+Ce+Y content (>350 ppm) and Ga/Al ratios indicating A-type characteristics of the rocks (Fig. 7). Furthermore, the DAM granites fulfill the compositional criteria (LOISELLE & WONES, 1979; WHALEN et al., 1987) for identifying them as A-type granites with high concentrations of SiO₂, Na₂O+K₂O, Zr, Nb, Ga, Y, Ce and Fe/Mg ratios, and low CaO, Sr and Ba contents (Table 4).

As suggested by several authors A-type granites can be formed in different ways (COLLINS et al., 1982; WHALEN et al., 1987; EBY, 1990; FROST & FROST, 1997; BONIN, 2007). Generally, models for the petrogenesis of A-type magmas are as follows: (1) Fractionation of mantle-derived basaltic magmas (EBY, 1990; BONIN, 2007); (2) mantle-derived magmas may assimilate crustal material during their ascent to produce a syenitic derivative that fractionates to a granitic composition (POITRASSON et al., 1995; BARTH, 1945); (3) this syenitic magma can also form granite with a mixed source characteristic due to further assimilation (BARKER et al., 1975); (4) melting of crustal material, namely low degrees of partial melting of granite that were depleted in incompatible elements by previous melt extraction (WHALEN et al., 1987; COLLINS et al., 1982).

EBY (1992) subdivided anorogenic A-type granitoids into two groups: A1-type granites (Y/Nb<1.2) representing a mantle differentiate of continental rift-related and other intraplate environments, whilst A2-type granites (Y/Nb>1.2) derived from melting of underplated continental crust emplaced in a variety of tectonic environments (e.g. back-arc, collision, post-collision and transcurrent faults etc.). The studied granites have low Y/Nb ratio of 0.04-0.88 suggesting within-plate A1-type characteristics (Fig. 8A) and emplacement in an extensional environment (Fig. 8B). This also implies that they are mantle differentiates with sources corresponding to OIB (Fig. 9).
The studied rocks are predominantly ferroan granites (Table 4). According to FROST & LINDSLEY (1991) reduced basaltic rocks from extensional environments could undergo Fe-enrichment or reduction through differentiation, whereas the relatively oxidized arc basaltic rocks could not. This implies that the DAM granites originated from a reduced basaltic compositional source. According to FROST & FROST (2011) ferroan granitoids are principally peraluminous and metaluminous, however there are also ferroan granites with a peraluminous nature. The ferroan alkali granitoids with a peraluminous component, like the studied rocks, can occur owing to the greater amount of felsic crust assimilation compared to metaluminous granitoids (FROST & FROST, 2011).

### 6.2. Geobarometry

Amphibole composition in plutonic rocks is commonly applied to determine the pressure of the crystallization and solidification of the pluton that can be used to determine the depth of the magma emplacement (e.g. VYHNAL & MCSWEEEN, 1990; ZHANG et al. 2006; STEIN & DIETL, 2001; PAPOUTSA & PE-PIPER, 2014). After empirical notification of the Al in amphibole barometer by HAMMARSTROM & ZEN (1986) and HOLLISTER et al. (1987), experimental studies (e.g. JOHNSON & RUTHERFORD, 1989a, b; THOMAS & ERNST, 1990; SCHMIDT, 1992) also confirmed that the Al content of the amphibole can indicate the pressure of its crystallization. These studies found that the Al content of the amphibole can increase with increasing pressure due to the Tschermak substitution $\text{Si} + 2\text{Al}^{3+} = \text{Al}^3 + 3\text{Al}^{VI}$. However, it is important to note that the composition of amphibole is sensitive for several parameters including the intensive parameters (p, T, fO2, fH2O), the crystallizing phases and the equilibrium melt composition. Thus, amphibole can be used as a barometer if the thermodynamic degrees of freedom can be reduced. Several criteria have to be met for accurate barometric calculations. The empirical studies indicate that a requisite mineral assemblage is necessary for hornblende barometry including quartz, plagioclase, K-feldspar, biotite, hornblende, Fe-Ti oxide and titanite, to buffer the amphibole composition (HOLLISTER et al., 1987; HAMMARSTROM & ZEN, 1986; ANDERSON & SMITH, 1995). The barometer has been calibrated experimentally for the assemblage quartz + plagioclase + K-feldspar + biotite + hornblende + Fe-Ti oxide + titanite + melt + fluid (JOHNSON & RUTHERFORD, 1989a, b; THOMAS & ERNST, 1990; SCHMIDT, 1992). Excluding titanite, all of these minerals can be found in the studied granites. The role of titanite is unknown but the study of ANDERSON & SMITH (1995) did not find any correlation between the presence or absence of this phase and the calculated pressure. They concluded that either the activity of titanite was at a level sufficiently close to saturation so as to buffer Al content in hornblende or not significantly affect the equilibr. Titanite is sensitive for the oxygen-fugacity and was stable in high fO2 experiments of JOHNSON & RUTHERFORD (1989a). The high Fe/(Fe+Mg) and low Fe3+/ (Fe3++Fe2+) ratio of the studied amphiboles suggest an intermediate/low fO2 for the crystallization condition of their host granite using the criteria of ANDERSON & SMITH (1995). Maybe the oxygen-fugacity was too low to stabilize titanite during the crystallization of the studied granites.

ANDERSON & SMITH (1995) found that amphiboles with an Fe/(Fe+Mg) ratio higher than 0.6 yield anomalously higher pressures for proterozoic anorogenic granites than the former barometric constraints when the Al-in-amphibole barometry was applied. According to the experiments of SCHMIDT (1993) Tschermak substitution preferentially removes Mg from the M2 site instead of Fe. ANDERSON & SMITH (1995) suggested that the low fO2 during crystallization of these anorogenic granite magmas lowered the Mg content of the amphiboles relative to Fe2+ and forced Tschermak substitution. This also accompanied Fe3+ - Al3+ exchange due to the low Fe3+ relative to total Fe. Thus the higher Fe/Mg and low Fe3+/ (Fe3++Fe2+) ratio of the amphiboles may be related to the low fO2 during crystallization of those anorogenic granites which resulted in the elevated Al content and the overestimation of the crystallization pressure. The amphiboles of the present study are characterized typically by low Fe3+/ (Fe3++Fe2+) (0.17±0.07) and high Fe/(Fe+Mg) (0.71±0.02) suggesting that the pressure values ob-
tained from Al in amphibole barometry can be viewed as a maximum.

Temperature can also affect the calculated pressure value through the edenite exchange that also increases the Al content of the amphibole. To eliminate the influence of temperature, the recalibration of ANDERSON & SMITH (1995) has been used which enables pressure calculations for igneous amphiboles that have been affected by the edenite substitution.

HOLLISTER et al. (1987) suggested that the composition of the coexisting plagioclase should be in the range of An25-35 because the tschermak substitution is governed by a reaction involving 2 quartz + 2 anorthite + biotit = tschermakite + orthoclase. According to this reaction HOLLISTER et al. (1987) suggested that low anorthite content of the plagioclase lowers the Al content of the amphibole. On the other hand HUMPHREYS et al. (2006) suggested that the plagioclase composition has a reverse effect on the Al content of amphibole due to the edenite + albite = richterite + anortite reaction (HOLLAND & BLUNDY, 1994). Thus, a higher An content of the plagioclase lowers the Al content of amphibole. The effect of the plagioclase composition on the Al content of amphibole is unclear, but it should be taken into account because in the studied samples it falls out of the optimal range (An = 11 mol%).

For the pressure calculation we used the spreadsheet of ANDERSON et al. (2008) (RiM69,ch04_hbd_plagthermo-jla.xls). This spreadsheet calculates pressure based on the Al in amphibole barometer using the ANDERSON & SMITH (1995) equation and temperature based on the amphibole-plagioclase thermometer (HOLLAND & BLUNDY, 1994; BLUNDY & HOLLAND, 1990). We used the "preferred" pressure values based on thermometer B of HOLLAND & BLUNDY (1994), taking into account the study of ANDERSON (1996). The calculated pressure value is 370 ± 40 MPa that corresponds to 14.1 ± 1.5 km depth using 2700 g/cm³ value for crustal density. As previously mentioned, above this pressure and depth, values can be interpreted as a maximum due to the higher Fe/Mg ratio of the studied amphiboles, however PAPOUTSA & PE-PIPER (2014) indicated that amphiboles that have a higher Fe/Mg ratio than the recommended value may also give reasonable pressure values. The other factor that may lower the accuracy of our result comes from the Ab-rich composition of the plagioclase, but the effect of this is not easy to quantitatively determine because of the opposite view of the effect of plagioclase composition on the Al content of amphibole.

The calcic amphiboles are not stable below ~100 MPa pressure according to experimental studies (e.g. RUTHERFORD & DEVINE, 2003, 2008). We conclude that the studied granites were emplaced at middle or upper crustal levels at ~14 km depth or shallower, but deeper than the ~4 km stability depth of calcic amphiboles.

### 6.3. Fractional crystallization

Geochemical characteristics of the DAM granites indicate that they have formed as a result of strong fractional crystallization. The distribution of whole-rock major and trace elements (Fig. 6, Table 4) together with the petrographic observations suggest the fractionation of calcic amphibole, ilmenite, allanite, apatite and zircon in the early stage of evolution of the studied granites. During this stage, Ce increases and starts to decrease from SiO2 70 w%, which indicates crystallization of monazite. While K-feldspar crystallized continuously during differentiation, albite is a late stage product of the evolution.

The fact that LREE are enriched compared to HREE indicates a fractionation between these elements. The presence of concave-up curvature in the pattern of HREE may reflect either HREE-rich fluid interaction or accumulation of zircon crystals (EBY et al., 1998). Since the most common accessory constituent is zircon, the most likely explanation for the upward HREE patterns is the zircon accumulation. The studied rocks show significant negative Ba, Sr and Eu anomalies (Fig. 6A). Plagioclase and amphibole fractionation produce negative Sr and Eu anomalies, whereas K-feldspar precipitation is responsible for the negative Ba anomaly (WU et al., 2002). According to ARSLAN & ASLAN (2006) the negative Ba anomaly may be considered as evidence of high temperature feldspar fractionation rather than biotite precipitation. Figure 6 shows that amphibole-bearing granite is distinct for Eu, Ba, and Sr due to its smaller negative anomaly. The high content of Sm and Gd may reflect the early precipitation of perthitic feldspar (BOWDEN & WHITELY, 1974), or crystallization of hornblende (KOCHHAR, 2000), while the Th anomaly is due to the presence of monazite and allanite.

### 7. CONCLUSIONS

Beside a series of alkaline igneous rocks, a granite body also crops out in the DAM. New whole-rock and mineral chemical data on the granites, together with the petrographic observations suggest the following implications.

(1) DAM granites are mostly alkaline and strongly peraluminous in composition. They are A-type granites with intra-plate A1-type characteristics indicated by their high Zr, Nb, Ga, Ce and Y content and Ga/Al and Fe/Mg ratios, and by their low CaO, Sr and Ba contents and Y/Nb ratios.

(2) The A1-type nature implies that the studied granites are mantle differentiates with sources corresponding to OIB and extensional environment.

(3) The studied granites were probably emplaced at middle or upper crustal levels at about 14 km depth or shallower, but deeper than the ~4 km stability depth of calcic amphiboles.

(4) Magmatic evolution of granites was mainly controlled by fractionation of calcic amphibole, ilmenite, allanite, apatite and zircon in the early stage and monazite and albite in a later stage of evolution. K-feldspar crystallized continuously during differentiation.

(5) Amphibole-bearing granites differ from the amphibole-free varieties in their lower Sr, Eu and Ba anomalies which suggests their less fractionated character.
These observations confirm the mantle origin of the DAM granites from a parental mafic magma together with fractional crystallization processes as assumed previously by MOROGAN et al. (2000) and PÁL-MOLNÁR (2000). However, the peraluminous nature of the studied granites suggests the possibility of further assimilation and crustal contamination.

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