Campanian U–Pb ages of volcaniclastic deposits of the Hațeg Basin (Southern Carpathians): Implications for future intrabasinal lithostratigraphic correlations

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(Manuscript received July 25, 2023; accepted in revised form November 17, 2023; Associate Editor: Igor Broska)

Abstract: Here we report new LA-ICP-MS zircon U–Pb ages of the Upper Cretaceous volcaniclastic deposits of the Hațeg Basin (Southern Carpathians, Romania). These deposits crop out around the area of Densuş and Râchitova localities in NW Hațeg and were previously included in the Maastrichtian continental Densuş–Ciula Formation. They are interpreted as eruptive sequences generated by repeated explosive volcanic eruptions, related to the Apuseni–Banat–Timok–Srednogoria (ABTS) magmatic activity, that manifested in Eastern Europe during the Late Cretaceous. The three samples selected for U–Pb age determination were collected from the Densuş volcaniclastic sequences; they are primary andesitic–dacitic lithic clasts resulted directly from volcanic eruptions. The new zircon U–Pb ages of 80.44±0.14, 80.22±0.25, and 81.88±0.17 (Early to earliest-Late Campanian), as well as geochemical analyses of the three sampled volcaniclastics, overlap with previously reported data for Banatite rock samples from the Banat and Apuseni segments of the ABTS belt. Age and composition similarities strongly suggest an affinity between the Late Cretaceous Neotethyan subduction-initiated magmatism and the volcaniclastic deposits of the Hațeg Basin. The Densuş–Ciula Formation unconformably overlies the Răchitova Formation – the youngest marine beds in this area, divided into Upper and Lower members. Recently, the age of the Upper Member was biostratigraphically restricted to the Early to earliest-Late Campanian, about the same period of time during which, according to the new U–Pb ages, subaerial volcanism took place. Considering their Early to earliest-Late Campanian age, the volcaniclastic deposits may represent a different lithostratigraphic unit from the Maastrichtian Densuş–Ciula Formation, to which they were formerly associated with. On these grounds, we suggest that the association of the volcaniclastic products with this formation, along with their stratigraphic and tectonic relationships, is revised.

Keywords: zircon U–Pb dating, volcaniclastic, Campanian, ABTS belt, Banatite, Hațeg Basin, Densuş–Ciula Formation

Introduction

Located in the western part of the Southern Carpathians (Fig. 1), the Hațeg Basin (Fig. 2) is widely known primarily for its latest Cretaceous vertebrate fauna dominated by dwarf insular dinosaurs, inhabitants of the so-called Hațeg Island of the Late Cretaceous European Archipelago (e.g., Nopcsa 1923; Weishampel et al. 1991; Benton et al. 2010; Csiki-Sava et al. 2015). A rather unique occurrence in Europe, the continental deposits of the Hațeg Basin containing these fossil accumulations are locally closely associated with an assemblage of volcaniclastic beds (e.g., Anastasiu & Csobuka 1989; Grigorescu & Anastasiu 1990; Anastasiu 1991), presumed, until now, to be of about the same age as the peculiar dwarf dinosaurs.

These volcaniclastic deposits are restricted to the northwestern part of the Hațeg Basin and crop out mainly in the area of Densuş and Râchitova localities (Fig. 2). They are often considered to belong to the same lithostratigraphic unit, namely Densuş–Ciula Formation, together with some of the youngest, continental beds that were deposited in this region during the Late Cretaceous and host dwarf dinosaur remains (e.g., Nopcsa 1905; Laufler 1925; Grigorescu 1992; Grigorescu & Csiki 2002). Nevertheless, the volcaniclastic deposits have a distinct spatial distribution within this unit, being restricted mainly to the Râchitova–Ciula Mică–Densuş–Ștei quadrangle. Studies of fossil vertebrates found in the upper, continental sediments of the Densuş–Ciula Formation, as well as biostratigraphic constraints from the underlying marine deposits (Figs. 3, 4), suggest that this unit is of Late Cretaceous (Maastrichtian) age (e.g., Grigorescu et al. 1990; Grigorescu 2010; see below).

The eruptive events that produced these volcaniclastic beds, are widely associated with the regional-scale magmatic activity that developed during the Late Cretaceous along a narrow band, called the Apuseni–Banat–Timok–Srednogoria (ABTS) belt, which extends from the Apuseni Mountains to the Banat region in Romania (Fig. 1), to Timok in Serbia and Srednogoria in Bulgaria, and finally further east to the Pontides in Turkey. Also known as the Banatitic Magmatic and Metallogenic Belt (e.g., Berza et al. 1998; Vlad & Berza 2003), this
**Fig. 1.** Simplified tectonic map of the Romanian Carpathians, emphasizing the distribution of the Late Cretaceous Banatitic magmatism (modified from Săndulescu 1984, 1988).

**Abbreviations:** TF-Trotuş fault; IMF-Intramoesian fault; PCF-Peceneaga-Camena fault; HB-Haţeg Basin

**Fig. 2.** Simplified geological map of the Haţeg Basin (completed after Csiki-Sava et al. 2016 and Melinte-Dobrinescu 2010). Legend: 1 – surrounding crystalline basement; 2 – pre-Quaternary sedimentary infill of the Haţeg Basin; 3 – marine Răchitova Formation (lower Santonian to lower Upper Campanian); 4 – continental Densuş-Ciula Formation (mainly Maastrichtian), with v – volcaniclastic sequences (Lower Member – Lower to lower Upper Campanian; this paper); 5 – continental Sînpetru Formation and correlative units (Maastrichtian); 6 – Quaternary deposits; black stars (7) mark the positions of the two volcanic tuffs sampled by Bojar et al. (2011) at Răchitova (a) and Ciula Mică (b), while the red star (8) marks the position of the Densuş volcaniclastic deposits.

*GEOLOGICA CARPATHICA*, 2023, 74, 5, 407–422
magmatic structure is part of the larger Alpine–Carpathian–Balkan–Himalayan belt, which spreads from Europe to Asia (e.g., Janković 1997) and mostly consists of plutons and dikes, sometimes associated with volcano-plutonic structures and volcanic formations (e.g., Berza et al. 1998; Ciobanu et al. 2002; Vlad & Berza 2003; Iancu et al. 2005). The ABTS magmatism developed within the framework of the Late Cretaceous northward subduction of the Neotethys or Vardar oceanic crust under the European margin and is usually attributed to the subduction process itself (e.g., Lips 2002; Neubauer 2002; Kamenov et al. 2003; von Quadt et al. 2005; Chambefort & Moritz 2006; Zimmerman et al. 2008; Gallhofer et al. 2015). Alternatively, some authors have put forward a different model, according to which magmatic activity along the belt occurred in response to processes related to lithospheric extension caused by orogenic collapse, after subduction of the oceanic crust ended (e.g., Berza et al. 1998; Bojar et al. 1998; Willingshofer et al. 1999; Ciobanu et al. 2002; Iancu et al. 2005). The subduction process took place in the broader context of the Late Cretaceous convergence between the European (Dacia and Tisza mega units) and African plates (Adria unit) (e.g., Boccaletti et al. 1974; Schmid et al. 2008, 2011; Maţenco et al. 2010). During the early part of the Mesozoic, the area corresponding to the present-day ABTS belt was affected by a series of rifting events, followed by collisional processes associated to the Austrian orogeny at the end of the Early Cretaceous, the Laramide or Early Alpine Phase during the Late Cretaceous and finally the Alpine Phase in the Cenozoic (e.g., Zimmerman et al. 2008 and references therein). Continental collision generated complex post-orogenic tectonic movements, causing significant deformation and bending of the belt during the Cenozoic (e.g., Ratschbacher et al. 1993; Fügenschuh & Schmid 2005).

Radiometric K–Ar and zircon U–Pb age data have shown that magmatic activity along the ABTS belt lasted from about 92 to 67 Ma (e.g., von Quadt et al. 2002; Stoykov et al. 2004; von Quadt & Peytcheva 2005; Marchev et al. 2006; Chambefort et al. 2007; Kolb 2011). In Romania, in the Apuseni and Banat segments, magmatic activity occurred roughly between 84 and 71 Ma (Gallhofer et al. 2015 and references therein).

Unlike the main segments of the ABTS belt, for which geochronology data are well enough represented, available ages of the Upper Cretaceous volcaniclastic deposits of the Hațeg Basin are limited. No age measurements of the Densuș volcaniclastic deposits were carried out prior to this study, however, some lower-resolution K–Ar ages of the Răchitova outcrop were previously published.

Bojar et al. (2011) used the K–Ar radiometric method to date a tuff layer (RA-12) that belongs to the Răchitova volcaniclastic deposits (45°36′5″ N; 22°45′5″ E) and a second tuff layer (AVROM-204) that crops out further to the east, near Ciula Mică, along the Get Valley (45°36′37″ N; 22°46′22″ E; Fig. 2). For sample RA-12 they reported an age of 69.8±1.3 Ma and a roughly comparable age of 71.3±1.6 Ma for sample AVROM-204. Both ages fall around the Campanian–Maastrichtian boundary or into the earliest Maastrichtian, in good accordance, as noted by Bojar et al. (2011), with the biostatigraphically constrained early Maastrichtian age of the Lower Member of the Densuș–Ciula Formation. Dating of a second sample RA-1 (andesitic volcaniclast) collected from the Răchitova outcrop, yielded a significantly older age of 82.7±1.5 Ma (Early Campanian). This age was interpreted by Bojar et al. (2011) as indicative of reworked Campanian-aged volcanic products, however, without mentioning a possible source of the volcanism.

We employed U–Pb dating on igneous zircon crystals in order to determine the age of three rock samples collected from the Densuș volcaniclastic deposits (Fig. 2), assumed until now to belong to the Lower Member of the Densuș–Ciula Formation (Fig. 4b). This study aims to: (1) provide the first absolute ages of the Densuș volcaniclastic sequences, allowing a comparison between their age and the age of the Răchitova volcaniclastic deposits, previously dated by similar means; (2) verify the temporal correlation between the volcaniclastic deposits of the Hațeg Basin and the Banatitic magmatism, to which they are assumedly genetically linked and (3) establish further constraints on the temporal relationship between the volcaniclastic successions and the surrounding Upper Cretaceous marine and continental deposits from northwest Hațeg.

**Geological setting**

Hațeg Basin (Fig. 2) is an intramontane postorogenic sedimentary basin, located in the western part of the Southern Carpathians, Romania. Its sedimentary succession suggests that it was formed during the Late Cretaceous, as a consequence of syn- and post-collisional extensional tectonics, after the collision between the African and European plates and later evolved as a so-called ‘Gosau-type’ basin on top of the Getic unit (Willingshofer et al. 1999, 2001).
The uppermost Cretaceous continental deposits of the Haţeg Basin (e.g., Grigorescu & Anastasiu 1990; Grigorescu et al. 1990) mark the end of widespread shallow to deep marine deposition for the most part of this epoch (Pop 1990), following the main thrusting and nappe emplacement phase that created the present-day large-scale structures of the Southern Carpathians (Sândulescu 1984). These continental deposits were historically divided into two major lithostratigraphic

Fig. 4. Lithostratigraphic column of the Upper Cretaceous sediments from NW Haţeg. a — Succession of the turbiditic marine beds of the Răchitova Formation, from Melinte-Dobrinescu & Grigorescu (2014); b — Succession of the continental beds of the Densuș-Ciula Formation, from Csiki-Sava et al. (2016). Lithology legend: 1 – volcano-sedimentary rocks, 2 – volcanic tuffs, 3 – tuffites, 4 – conglomerates, 5 – sandstones, 6 – siltstones, 7 – mudstones/shales, 8 – limestones, 13 – unconformity, 14 – contact unknown/not exposed; age constraints: 15 – palynology, 16 – radiometric ages, 17 – nannoplankton, 18 – foraminifera.
units: Densuş–Ciula Formation, in the northwestern part of the basin and Sînpetru Formation, in the central-eastern part of the basin (Fig. 2; e.g., Grigorescu 1992), albeit their lithostratigraphic make-up may be somewhat more complicated than this two-fold division (e.g., Csiki-Sava et al. 2016).

Deposits of the Densuş–Ciula Formation are known to rest unconformably atop the turbidites of the Râchitova Formation (Laufer 1925; Figs. 3, 4). Râchitova Formation represents the youngest marine deposits in the northwestern part of the Haţeg Basin and consists of a Lower and an Upper Member, separated on the basis of their relative stratigraphic position, lithology, and paleontological content (e.g., Grigorescu & Melinte 2001; Melinte-Dobrineșcu 2010). The unconformable contact between the two formations is clearly visible around Densus, where it can locally be of tectonic nature (Fig. 3; e.g., Nopcsa 1905; Bârzoï & Şeclăman 2010). In other areas (e.g., Râchitova), according to bed geometry and dip details, it is assumed to be an angular unconformity at the base of the Lower Member, (e.g., Grigorescu & Melinte 2001). The Upper Member of the Râchitova Formation (Figs. 3, 4a) was formerly placed in the Upper Campanian owing to two identified nannofossil events, the first occurrence (FO) of the calcareous nanoplankton taxa Uniplanarius sissinghii and U. trifidus (Melinte-Dobrineșcu 2010). However, recent palynostratigraphic studies indicate that this member is older, with a revised age of Early to Late Campanian (Ţabâră & Slimani, 2019).

Densuş–Ciula Formation contains three separate and successively younger members – Lower, Middle, and Upper, each characterized by different lithology and paleontological content (Fig. 4b; e.g., Grigorescu et al. 1990; Grigorescu 1992, 2010).

The Lower Member is described as being dominated by volcaniclastic material, associated with shales, conglomerates, and sandstones (Fig. 4b; e.g., Anastasiu & Cso Bukka 1989; Grigorescu 1992). Both Densuş and Râchitova volcaniclastic deposits were recognized as the Lower Member by previous authors due to the continental nature of the volcanic eruptions and also because of the stratigraphic position of the volcaniclastics relative to the other Upper Cretaceous deposits of this formation, as they are altogether emplaced on top of the marine Râchitova Formation (e.g., Laufer 1925; Grigorescu & Anastasiu 1990; Grigorescu 1992). Recent age estimation based on different criteria such as its stratigraphic superposition, biostratigraphy, and the above mentioned K–Ar age of the Râchitova tuff layer, placed the Lower Member in the lower Maastrichtian, with an approximate age of about 68–72 Ma (Fig. 4b; e.g., Grigorescu 2010; Bojar et al. 2011; Csiki-Sava et al. 2016), although its age assessment varied significantly over the years (see below). Melinte-Dobrineșcu (2010) and Melinte-Dobrineșcu & Grigorescu (2014) assigned a latest Campanian age to the lowermost part of the Lower Member and a Maastrichtian age to the remaining part of this member, as well as to the Middle and Upper members (Fig. 4a).

The dominantly siliciclastic Middle Member is made up of repetitive sequences of sandstones, mudstones, and matrix-supported conglomerates (Fig. 4b); the conglomerates may occasionally contain variable amounts of reworked volcanogenic clasts (e.g., Grigorescu 1992; Bojar et al. 2005; Grigorescu et al. 2010; Vasile et al. 2011a; Botfalvai et al. 2017, 2021). The presence of such volcaniclastic fragments appears, however, to be restricted to certain lithotypes and stratigraphic intervals within this member (e.g., Grigorescu et al. 2010; Vasile et al. 2011a; Botfalvai et al. 2021). Deposits of the Middle Member yielded the largest number of vertebrate fossils unearthed from the Densuş–Ciula Formation (e.g., Grigorescu 1992; Csiki-Sava et al. 2016). Latest studies have shown that the age of this member falls within the early to early-late Maastrichtian (e.g. Csiki-Sava et al. 2016; Botfalvai et al. 2021).

The Upper Member mostly consists of coarse siliciclastic sediments, especially red sandstones and conglomerates, siltites, and reddish mudstones, supposedly devoid of vertebrate remains and reworked volcanic material (Fig. 4b; e.g., Grigorescu & Anastasiu 1990; Weishampel et al. 1991; Grigorescu 1992). Albeit its age was supposed to possibly extend into the Paleocene (e.g., Weishampel et al. 1991; Grigorescu 1992), recent discoveries of vertebrate fossil occurrences in deposits belonging to this member, around General Berthelot and Crăguş, place it in the upper Maastrichtian (Vasile et al. 2011a,b).

The Densuş volcaniclastic deposits extend between Densus and Ștei localities, along the Densuş river valley, and they are interpreted as two different andesitic-dacitic eruptive sequences (e.g., Bârzoï & Şeclăman 2010; Popa & Seghedi 2015). For differentiation purposes, we named the two outcrops sequence no. 1 and sequence no. 2. The two sequences are geographically found in close proximity of each other, being separated by a distance of only a few tens of m, with sequence no. 2 (GPS coordinates: 45°34′53″ N/22°45′9″ E) positioned east of sequence no. 1. (GPS coordinates: 45°34′51″ N/22°45′3″ E), towards Densus. Both sequences were built up by the accumulation of products of multiple Late Cretaceous subaerial explosive volcanic eruptions, interrupted by periods of inactivity, produced by two different volcanic centers (Popa & Seghedi 2015). They consist of a succession of primary volcanic deposits resulted directly from volcanic eruptions, like deposits from pyroclastic density currents and tephra fall, and secondary deposits such as lahars, with subordinate epiclastic beds (Popa & Seghedi 2015). Lithology of the two Densuş volcaniclastic sequences indicate that they actually are fragments of former volcanic edifices. In the case of sequence no. 1, the deposits constituted the medial volcanic facies, while the deposits that make up sequence no. 2, from where the rock samples analysed here were collected, represented the medial and distal facies of the former volcano. All the volcanic material contained by these deposits can be traced down to a single volcanic source, which is different for each of the two sequences. Consequently, we appreciate that the U–Pb ages of the sampled lithic fragments reflect the ages
of the volcanic eruptions that produced depositional sequence no. 2.

Similarly, the more northerly Răchitova outcrop consists of a succession of primary and secondary volcaniclastic deposits of andesitic, dacitic, and rhyolitic composition, also produced by cyclic explosive volcanic eruptions, in continental settings (e.g., Bărzoi & Şeclăman 2010).

**Previous assessments of the stratigraphic position and age of the Răchitova–Densuș volcaniclastic beds**

The Răchitova–Densuș volcaniclastic beds have a somewhat convoluted history of stratigraphic interpretations and these previous assessments will be briefly reviewed here, in order to better emphasize the significance of our new U–Pb geochronology data. The petrography, geochemistry, and genetic-sedimentological characteristics of these rocks had been addressed in some detail previously by Anastasiu & Csobuka (1989), Grigorescu & Anastasiu (1990), Anastasiu (1991), and Bărzoi & Şeclăman (2010), and will not be discussed here further unless necessary; their updated petrographical–petrological and geochemical overview is currently in progress.

The occurrence of volcaniclastic beds in the Densuș area had been first noted by Nopcsa (1905), who correlated them with the vertebrate-bearing continental deposits cropping out more to the southeast, near Sânpetru (Fig. 2). He noted that they grade into dark-coloured fossiliferous siliciclastics, reminiscent of those near Sânpetru. He also remarked that these volcaniclastic beds (his ‘tuffaceous facies’) are wedged between continental ones to the east and flyschoid Upper Cretaceous marine deposits to the west, thus assigning them a ‘Danian’ (*sensu* latest Cretaceous) age. Both Nopcsa (1905) and Schafarzik (1909) reported the presence of a tectonic contact along subvertical faults between the marine and volcaniclastic sequences west of Densuș (Fig. 3), whereas more to the west, near Ștei, the volcaniclastics are described as following with a slight angular unconformity on top of the turbidites. Subsequently, the interpretation of Laufer (1925) progresses beyond that of Nopcsa in establishing that the ‘tuffaceous facies’ represents the stratigraphically lower term of the Danian, resting mainly conformably, albeit transgressively, on top of the Upper Cretaceous flyschoid deposits. The ‘tuffaceous facies’ succession is considered to be covered conformably by the continental deposits of the ‘fluvial-lacustrine facies’ (Laufer 1925).

In an important contribution concerning the stratigraphy and age of the Densuș volcanoclastics and of their neighboring units, Antonescu et al. (1983) referred to the ‘tuffaceous facies’ of Nopcsa, resting atop Upper Cretaceous flysch, to the volcaniclastic Maastrichtian Densuș Formation, and the overlying reddish ‘lacustrine facies’, to the upper Maastrichtian Ciula Formation. He acknowledged that the upper, unfossiliferous part of this unit may represent either the upper Maastrichtian or the Paleogene.

Later, Grigorescu (1992) opined that Densuș and Ciula formations of Antonescu et al. (1983) should be considered in fact two, temporally successive members of the same major lithostratigraphic unit, the Densuș–Ciula Formation of late Maastrichtian age (see also Weishampel et al. 1991). Meanwhile, Grigorescu & Anastasiu (1990) divided the same Densuș–Ciula Formation into three different, superposed members, with the Upper Member, extending eastward from west-Fărcădăin, of a possibly Paleogene age. These authors also noted that the contact between the upper Maastrichtian Lower Member of the Densuș–Ciula Formation (=volcaniclastic successions) and the underlying flyschoid Campanian–lower Maastrichtian is represented by an angular unconformity, visible in the field at Ștei. Subsequently, these Upper Cretaceous marine beds were formalized as the Răchitova Formation by Grigorescu & Melinte (2001), with a reassessed age of latest Santonian to Late Campanian based on calcareous nannoplankton, later refined by Melinte-Dobrinescu (2010) to the early Santonian to late Late Campanian interval. The latter author also noted that the youngest identified bioevent demonstrates that marine deposition ceased by the latest Campanian, with the start of continental deposition (marked by the deposition of the volcaniclastic Lower Member of the Densuș–Ciula Formation that unconformably covers the Upper Member of the Răchitova Formation over an erosional base; see also Bărzoi & Şeclăman 2010; Bojar et al. 2011) around the Campanian–Maastrichtian boundary. The age of the upper Răchitova Formation had been further revised to the Early to early-Late Campanian by Țabără & Slimani (2019), using marine and continental palynomorph assemblages, suggesting an even earlier conclusion to the marine sedimentation in the northwestern Hațeg Basin.

Finally, based on the petrography and geochemistry of the volcaniclastic rocks from Densuș and Răchitova, Bărzoi & Şeclăman (2010) identified their origin as a (most probably continental) island arc emplaced on top of a thinned continental crust, and suggested that these rocks represent an exotic tectonic unit welded together with other units of the Hațeg Basin alongside mainly local strike-slip faults. Accordingly, the dominantly volcaniclastic Răchitova–Ștei unit is seen as tectonically set apart from other surrounding units, even from the siliciclastics-dominated Middle and Upper members of the Densuș–Ciula Formation (separated by these authors as the Tuștea unit). Nevertheless, they acknowledge that, locally, the volcaniclastics rest unconformably on top of Upper Cretaceous turbidite marine beds.

In conclusion, in spite of several research studies conducted on the volcaniclastic deposits from the Răchitova–Densuș area, there are still lingering uncertainties regarding their nature, position, and age, as well as their spatial and temporal relationships with other surrounding geological units. Probably the most generally accepted opinion concerns their age, assessed to be latest Cretaceous, whether as ‘Danian’ (e.g., Nopcsa 1905; Laufer 1925), Maastrichtian (e.g., Grigorescu 1992), early Maastrichtian (Bojar et al. 2011; Csiki-Sava et al. 2016), or even starting with the latest Campanian (Melinte-Dobrinescu 2010).
Materials and methods

Three samples, D2-1, D2-2, and D2-3, were collected from sequence no. 2, of the two volcaniclastic sequences that crops out in the Densuş area (Fig. 5a,b; sample location GPS coordinates: 45°34′54″ N/22°45′11″ E, alt 441.2 m), for U–Pb radiometric age determination.

The samples are primary andesite and dacite lithic clasts embedded in an approximately 4 m thick volcanic mudflow or type lahar deposit of mixed sediment composition, belonging to the succession that forms sequence no. 2. From a lithological point of view, the lahar layer can be defined as a conglomerate deposit and consists of a high concentration of predominantly sub-angular and sub-rounded coarse-grained volcanic (mainly andesites and dacites) and metamorphic (mostly quartzites, amphibolites, and schists) lithic clasts, with sizes up to 30 cm, supported in a matrix of fine-grained volcanic ash, with grain size less than 1 mm. Most probably, volcanic rock fragments deposited in the area close to the volcanic vent or on the flanks of the volcano (the central and proximal facies) were entrained into the mudflow, whose formation was likely triggered by an eruption episode. Lahars are generated when a large quantity of external water mixes with unconsolidated, loose, fragmented material, such as tephra or other sediments deposited on the flanks of volcanos, forming gravity-driven mudflows which move down the slopes of a volcano at great speeds (e.g., Smith & Fritz 1989; Vallance 2000; Major 2022).

Approximately one kg of each bulk sample was crushed and then sieved in order to separate the fraction smaller than 300 μm. Heavy mineral mass fraction <300 μm was separated with a high-density liquid (diiodomethane with a density of 3.3 g/cm³), followed by a further magnetic separation using a Frantz Magnetic Barrier Separator. The separated zircons were mounted in epoxy resin alongside other zircon specimens of known age.

U–Pb isotopic analyses were performed on polished individual grains at the Arizona LaserChron Center, using an Element2 high resolution single collector inductively coupled plasma-mass spectrometer (LA-ICP-MS) equipped with a Teledyne G193 nm Excimer laser with a beam diameter of 30 μm (Gehrels et al. 2008). The selection of 100 crystals ensured an analyzed sample representative of the entire zircon population (Gehrels 2014). Every 5 crystals of unknown age were bracketed by a known standard; our primary standards for this study were the Sri Lanka and R33 zircons, which have ages similar to most of our unknowns (Gehrels & Pecha 2014; Roban et al. 2023). Ages that are less than 90 % concordant (discordance between $^{206}\text{Pb}/^{238}\text{U}$ and $^{207}\text{Pb}/^{235}\text{U}$ ages) were discarded from further analysis. For ages less than 1.4 Ga, the 6/8 age was reported and used. For the purpose of this paper, $^{206}\text{Pb}/^{238}\text{U}$ age results were plotted and average ages were calculated by the ISOPLOT-R package (Vermeesch 2018). Data reduction was performed using the Iolite software.

Whole-rock major and trace element compositions were determined at ALS Ireland (http://www.alsglobal.com/geochemistry). Powdered samples were submitted to lithium borate fusion followed by acid dissolution, and then analyzed using inductively-coupled plasma–atomic emission spectrometry (ICP-AES) for major elements (ALS code ME-ICP06) and inductively-coupled plasma–mass spectrometry (ICP-MS) for trace elements including rare-earth elements (ALS codes ME-MS42, ME-MS61, ME-MS81). Weight loss on ignition (LOI) was determined gravimetrically after heating the rock powders to ~1000 °C using a thermogravimetric analyzer (TGA; ALS code OA-GRA05). The results are summarized in Table 1. Limit of detection (LOD) is given in the table.

Fig. 5. a — Overview of volcaniclastic sequence no. 2 that crops out in the Densuş area, Haţeg Basin, from where the three samples were collected; b — The exact location of the rock samples, a small valley that cuts through some of the volcaniclastic deposits of this sequence (GPS coordinates: 45°34′54″ N/22°45′11″ E). Author: Violeta Vornicu.
Petrographic analysis of the rock samples was performed using an Olympus BX41 TF microscope.

Results

Petrography

The three volcaniclastic rock samples have porphyritic texture, typical for andesite/dacite rocks, with phenocrysts embedded in a fine-grained groundmass composed of microcrystals and volcanic glass (Fig. 6a, b, c). The majority of crystals are idiomorphic and inequigranular and they range in size from 0.1 to 2 mm, although sometimes larger crystals can be seen.

Sample D2-1 is mostly composed of phenocrysts of brown hornblende and plagioclase feldspar. The groundmass largely consists of plagioclase feldspar and volcanic glass (Fig. 6a). Sample D2-2 has a roughly similar composition, with phenocrysts of plagioclase feldspar, amphibole, and clinopyroxene, set in a fine-grained groundmass primarily made up of plagioclase feldspar and glass particles (Fig. 6b). Equally, the mineralogical composition of sample D2-3 is dominated by plagioclase feldspar, hornblende and clinopyroxene (Fig. 6c). Apatite, zircon, and opaques are common accessory minerals in all three samples. Many of the plagioclase crystals show concentric chemical zonation, from calcium-rich cores to more sodic rims and the hornblende is usually found as glomerocrysts. Sometime, the plagioclase feldspar is impregnated with iron oxides or replaced by sericite. Mafic minerals are, at times, altered to chlorite or epidote. Frequently, hornblende crystals exhibit partial or total opacitization due to replacement by magnetite and/or pyroxene.

Geochemistry

Major and trace element geochemical results are in good agreement with the geochemical data previously reported by Bărzoi & Şeclăman (2010), for volcaniclastic rock samples collected from Densuş and Râchitova; for comparison, their results are provided as supplementary material (Table S1). The initial chemical composition of the three samples could have been slightly modified by diagenetic processes, given their old Late Cretaceous age. Loss on ignition (LOI) values (Table 1) indicate low to moderate alteration.

On the TAS (total alkali vs. silica) diagram (Le Bas et al. 1986) samples D2-1 and D2-3 are classified as dacites, while sample D2-2 falls into the field of andesites (Fig. 7). According to the AFM diagram (Irvine & Baragar 1971; Fig. 8a), the three samples fall into the field of calc-alkaline magmas, with the samples from by Bărzoi & Şeclăman (2010) following the same chemical trend.

On the primitive mantle-normalized trace element diagram (Sun & McDonough 1989; Fig. 9), the Densuş samples, plotted together with those from Bărzoi & Şeclăman (2010), display enrichment in large-ion lithophile elements (LILEs), like Ba, Pb, K, and Sr and depletion in high field strength elements (HFSEs), such as Nb, Zr, and Ti.

U–Pb geochronology

Zircon U–Pb analyses of the three samples yielded crystallization ages between 80 and 82 Ma, in accord with the age of the Late Cretaceous Neotethyan subduction-related magmatism (92–67 Ma; Gallhofer et al. 2015 and references therein).

Table 1: Bulk chemical composition of the three Densuş volcaniclastic rock samples.

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<tr>
<th>Sample ID</th>
<th>D2-1</th>
<th>D2-2</th>
<th>D2-3</th>
<th>LOD/%</th>
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</table>

GEOLOGICA CARPATHICA, 2023, 74, 5, 407–422
Individual mean crystallization ages are $80.44 \pm 0.14$ Ma for sample D2-1 (Fig. 10), $80.22 \pm 0.25$ Ma for sample D2-2 (Fig. 11), and $81.88 \pm 0.17$ Ma for sample D2-3 (Fig. 12), showing that all three sampled specimens were formed during the Early to earliest-Late Campanian. Detailed U–Pb geochronological data of the three samples (Tables S2, S3, S4), as well as U–Pb concordia diagrams (Figs. S1, S2, S3), are added as supplementary files.

It is worth mentioning that inherited zircon crystals have also been found in two of the three rock samples. The ages of inherited zircons identified in sample D2-1 are 143.68 Ma, 150.59 Ma and 157.42 Ma respectively (Late Jurassic–earliest Cretaceous). Compared to the host rock, inherited zircon grains in sample D2-2 display much older U–Pb ages, between 2.54 and 2.8 Ga (Neo-Archean).

**Discussion**

*Temporal correlation between the volcaniclastic deposits of the Haţeg Basin and the Late Cretaceous Banatitic magmatism*

The new zircon U–Pb ages and geochemical results are in good agreement with previously reported data for Banatite rock samples from the Banat and Apuseni segments of the ABTS belt. These data are made available in the Carpathian–Pannonian Magmatism Database and can be accessed online at [https://osf.io/23kdg/](https://osf.io/23kdg/) (Vlăsceanu et al. 2021). As far as the Late Cretaceous ABTS magmatism is concerned, this compilation contains data gathered from the analysis of 127 whole-rock samples by Dupont et al. (2002), Gallhofer et al. (2015), and Vander Auwera et al. (2016).

In Romania, in the Apuseni and Banat segments of the ABTS belt (Fig. 1), geochronology data revealed that magmatic activity took place roughly between 84 and 71 Ma (Gallhofer et al. 2015 and references therein) and the U–Pb ages of the three Densuş volcaniclastic rock samples fall well
within this time interval (Fig. 13). Fifty-four igneous rock samples collected from Banat and Apuseni were dated by Gallhofer et al. (2015), their study covering relatively large areas of both segments. Zircon U–Pb dating of samples from the Banat segment (see Fig. 1) gave age results in the range 70.2–83.98 Ma. Dating of two extrusive rock samples – DG002 (dacite) and DG042 (andesite) – from the Banat Mountains yielded ages of 80.8 Ma and 79.5 Ma respectively, in accord with our age data. Age values of rock samples from the Apuseni segment (see Fig. 1) range from 75.5 to 80.8 Ma. 

Dating of extrusive rocks, such as dacites and rhyolites, produced ages between 77.85 and 80.8 Ma.

It should be mentioned that Rusca Montană Basin, which belongs to the Poiana Ruscă area, part of the Banat segment of the ABTS belt, is the closest location to the Haţeg Basin where Banatitic magmatic activity occurred during the Late Cretaceous. U–Pb dating of four intrusive rock samples collected from the Poiana Ruscă area, mostly granodiorites, produced age values between 75 and 78 Ma, lower than the ages of the Densuş volcaniclastic samples but still comparable. Dated samples were DG054 that yielded an age of 78 Ma, DG045 with the age of 75.6 Ma, DG044 which is 76.6 Ma old and finally sample DG049 with the age of 77.15 Ma. Even though extrusive rocks from Poiana Ruscă have not been dated as yet, the age of andesite and dacite samples from this location was estimated at 80 Ma by Gallhofer et al. (2015), based on the stratigraphic position of the rocks in relation to the igneous intrusions that have been dated. In any case, since this is an estimated age, it should only be used as a reference value.

In terms of chemical composition, our major and trace element results overlap with the geochemical data previously published by Dupont et al. (2002), Gallhofer et al. (2015) and Vander Auwera et al. (2016) for several intrusive/extrusive rock samples collected from the Banat and Apuseni segments of the ABTS belt (Fig. 8b). As discussed above, the three Densuş samples belong to the calc-alkaline magma series (Fig. 8a,b), like almost all Late Cretaceous igneous rock specimens from the ABTS belt, except for those from East Srednogorie, which fall into the field of tholeiitic magmas due to their high Fe and Mg content (Gallhofer et al. 2015 and references therein). Most of the Banatite rock samples from
Apuseni belong to the high-K calc-alkaline rock suit and have SiO₂ contents greater than 60 wt %, being classified as dacites and rhyolites (Gallhofer et al. 2015; Vander Auwera et al. 2016), while the majority of Banat samples plot into the calc-alkaline and high-K calc-alkaline fields and have intermediate to acidic compositions (Dupont et al. 2002; Gallhofer et al. 2015).

Although there is no direct link between the volcaniclastic deposits of the Haţeg Basin and the Late Cretaceous magmatic products of the ABTS belt, the former being positioned way outside the belt area, good temporal correlation, as argued above, together with similar geochemical characteristics, strongly suggest that magmatic/volcanic activity from the two locations could have developed within the same geotectonic framework. A point of importance that should be noted here is that, apart from the Banatitic magmatism, there is no other magmatic event known to have taken place in the area corresponding to the present-time Banat region, that may be regarded as a potential source of the volcaniclastic products that crop out in the Haţeg Basin. Thereby, it is reasonable to bring into question a possible genetic relationship between the studied deposits and the ABTS magmatism.

**Temporal relationship between the volcaniclastic deposits and the surrounding Upper Cretaceous marine and continental beds from the northwestern part of the Haţeg Basin**

The newly determined zircon U–Pb ages of the Densuş volcaniclastic sequences are consistent with the K–Ar age of a Răchitova volcaniclastic rock sample (RA-1) of 82.7±1.5 Ma, previously dated by Bojar et al. (2011) and they are indicative of subaerial volcanic eruptions during the Early to earliest-Late Campanian. However, the U–Pb ages are significantly older than the ages of the two tuff layers already mentioned, RA-12 and AVROM-204 (69.8±1.3 and 71.3±1.6 Ma), also sampled by Bojar et al. (2011). With respect to the two samples collected by Bojar et al. (2011) from the Răchitova volcaniclastic sequence, the rhyolitic tuff layer (RA-12) is found at the very base of the outcrop, while the andesitic unit (RA-1) is stratigraphically positioned above the tuff layer, high up in the sequence, so the tuff layer should normally be older than the andesitic products. Surprisingly, as per the K–Ar age results,
As previously mentioned, the oldest continental sediments of the Densuş–Ciula Formation, namely the volcanioclastic deposits recognized as its Lower Member, overlay the youngest marine deposits of the Râchitova Formation (Figs. 3, 4), formed during the Early Campanian (Ţabără & Slimani 2019). The U–Pb ages of the Densuş volcanioclastic rock samples, of about 80–82 Ma, are very close to the age of the marine beds and suggest that continental volcanism took place around the same time as that of the last stages of marine sedimentation. This interesting temporal relation between the volcanioclastic beds and the marine deposits of the Râchitova Formation was also noticed by Melinte-Dobrinescu & Grigorescu (2014).

The precise geotectonic context that triggered the volcanic eruptions is currently unclear, however evidence from multiple lines of research suggests that, probably, the volcanic activity that produced these deposits does not originate within the present-day Hâşteş Basin (Bârzoi & Şeclăman 2010). In this regard, an important aspect is that, unlike the segments of the ABTS belt, no intrusive bodies like plutons or dykes have been found in the Hâşteş Basin. Previous authors (e.g., Laufer 1925; Pătraşcu et al. 1993) mentioned the presence of dykes in the area and their discovery would definitely prove that magmatic activity occurred in the Hâşteş Basin but the occurrence of such intrusions has not yet been confirmed by our field observations. The hypothesis that the actual volcanism originated outside the Hâşteş area is further strengthened by the absence of volcanic vents, features that can be directly linked to the generation of the volcanioclastic deposits. Evidence in favour of this idea also comes from deposit thickness. All volcanioclastic successions of the Hâşteş Basin are only a few tens of meters thick so, most probably, they do not represent the entire depositional sequences resulted from volcanic eruptions. According to literature, deposits produced by explosive eruptions of composite volcanoes, the same type of volcanoes that, as our studies have shown, were responsible for erupting the volcanic material, are normally much thicker and spread over wider areas around the volcanic vent (e.g., Vessell & Davis 1981; Smith 1988, 1991; Davidson & De Silva 2000; Kano & Takarada 2007). The most plausible explanation for their reduced size is that they are fragments cut off from the original eruptive sequences, which likely covered much larger areas.

As discussed above, Densuş–Ciula Formation is believed to be of Maastrichtian age, as suggested by the K–Ar ages of the two aforementioned tuff layers and by its stratigraphic position, being deposited atop the biostratigraphically dated marine beds. Prior to the U–Pb dating results, the volcanioclastic successions, which make up the Lower Member, were also thought to largely represent the Maastrichtian (e.g., Grigorescu 2010; Bojar et al. 2011; Csiki-Sava et al. 2016).

Since the volcanioclastic deposits have been dated to Early to earliest Late Campanian, it becomes apparent that there is a notable age difference between the volcanioclastic Lower Member and the Middle and Upper members of the Densuş–Ciula Formation, believed to be of early to late Maastrichtian age (Fig. 4b). This age gap may be interpreted as an indicator that the volcanioclastic successions and the Upper Cretaceous non-volcanic continental sediments from northwest Hâşteş might have been formed within different geotectonic frameworks. Further refinement of the age of the Middle and Upper members of the Densuş–Ciula Formation could possibly shed light on this matter.

Accepting the new U–Pb ages as the timeline of the Late Cretaceous volcanic eruptions, we suggest that the association of the Lower Campanian volcanioclastic products with the Maastrichtian continental sedimentary succession of the Densuş–Ciula Formation, and subsequently their previously established lithostratigraphic relation, is revised, as they may represent different lithostratigraphic units. Accordingly, we propose a reinterpretation of the stratigraphic relationships between the different Upper Cretaceous units from the northwestern part of the Hâşteş Basin (Fig. 14). Such a reinterpretation would largely resurrect the bipartite lithostratigraphic scheme proposed for these uppermost Cretaceous continental deposits by Antonescu et al. (1983), who grouped the volcanioclastic successions into Densuş Formation, and the overlying, dominantly red-coloured, and fossiliferous continental beds into Ciula Formation.

Conclusions

In this paper we report new laser ablation ICP-MS zircon U–Pb ages of three primary andesitic/dacitic volcanioclastic
Zircon U–Pb dating yielded age values between 80 and 82 Ma (Early to earliest-Late Campanian), falling within the assessed time span of the Late Cretaceous ABTS subduction-related magmatism. Furthermore, the ages of the three volcaniclastic samples are in close agreement with those of Banatite rock samples from the Banat and Apuseni segments of the ABTS belt, dated by the same radiometric method.

The ages of the volcaniclastic deposits, as well as the age of the underlying Upper Member of the marine Răchitova Formation, fall into the Early to early-Late Campanian, suggesting that subaerial volcanic eruptions occurred around the time when sedimentation of the youngest marine deposits began. This interesting temporal relationship between the two Late Cretaceous events is worth exploring further.

Although the volcaniclastic deposits were regarded by previous authors as the Lower Member of the Densuş–Ciula Formation, the new U–Pb ages of 80–82 Ma are inconsistent with the estimated Maastrichtian age of the continental sedimentary succession of this formation, that makes up its Middle and Upper members. If the inferred Maastrichtian age of these two members is confirmed, we suggest that the association of the volcaniclastic deposits with the Densuş–Ciula Formation is re-evaluated and the volcaniclastic products are treated as a separate lithostratigraphic unit.

Nevertheless, for the time being we refrain from formalizing such a proposal as we reckon that in order to definitively establish the precise nature of the relationships between the marine, volcaniclastic, and continental successions, additional studies are still required, particularly of their age, conditions of genesis, and tectonic evolution.

Acknowledgment: We thank Editor Silvia Antolíková for her valuable guidance and support; we also thank two anonymous reviewers for the time and effort invested in reading and carefully revising the manuscript. We appreciate their thoughtful and constructive review that helped improve the manuscript content. This research was supported by the Romanian Executive Agency for Higher Education, Research, Development, and Innovation; Funding projects PN-III-P4-ID-PCE-2016-0014 and PN-III-P4-ID-PCE-2020-2570.

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Electronic supplementary material is available online: Tables S1–S4 and Figs. S1–S3 at http://geologicacarpathica.com/data/files/supplements/GC-74-5-Vornicu_Supplements.docx

GEOLOGICA CARPATHICA, 2023, 74, 5, 407–422