

# Contributions to Mineralogy and Petrology

## Petrogenesis and amphibole–melt trace element partitioning of the 156 ka Haramul Mic crystal-rich dacite, Ciomadul, Romania --Manuscript Draft--

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| Abstract:                                     | <p>The 156 ka Haramul Mic crystal-rich (ca. 40 vol%) dacitic lava dome represents eruption of a crystal mush. It is the first product of the Ciomadul Volcanic Complex, eastern-central Europe, following at least 100 kyr dormancy. The mineral phases (plagioclase, amphibole, biotite, apatite, titanite and zircon) and the interstitial glass have relatively homogeneous major and trace element composition suggesting near-equilibrium pre-eruptive state. Geothermobarometric and hygrometric calculations indicate a near-eutectic, low-temperature (730 oC), low-pressure (200–400 MPa), wet (6.5–6.7 wt% melt H<sub>2</sub>O content) and oxidized silicic magmatic system. A wide set of amphibole-melt trace element partition coefficient data is provided, which can be used for such low temperature silicic volcanic and plutonic systems. Additionally, mineral-melt trace element partition coefficients are also calculated for titanite, zircon, plagioclase and biotite phases. Quality-check was performed by Onuma-plots and the crystal lattice parameters were determined which fit well with published results. In contrast to the subsequent eruptions, there is no sign for mafic magma recharge as</p> |                     |

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|                               | <p>eruption initiation. Thus, volatiles could have played a role such as 'second boiling' or volatile oversaturation due to degassing from underplated mafic magma. The lack of reaction corona around amphibole macrocrysts suggests relatively fast magma ascent, although this can also be due to the still relatively high water content (&gt;4 wt%) of the melt, where amphibole remained stable. The Ciomadul provides an example where various processes can contribute to eruption initiation reflected by the compositional characteristics of the crystal cargo even though the composition of the erupted magma is similar.</p> |
| <b>Response to Reviewers:</b> | <p>The number of figures was reduced (now there is 10 figures in the main text, all the others are in the Electronic Supplementary Material); tables containing the mean values of trace elements in glass and mineral phases and the calculated partition coefficients are added; figures and tables are separated from the main text.</p>                                                                                                                                                                                                                                                                                                |



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28<sup>th</sup> February 2025

Dear Professor Müntener,

We are pleased to submit our manuscript entitled

**Petrogenesis and amphibole–melt trace element partitioning of the 156 ka Haramul Mic crystal-rich dacite, Ciomadul, Romania**

by Emese Pánczél, Szabolcs Harangi, Maurizio Petrelli, Răzvan–Gabriel Popa, Attila Virág, Ioan Seghedi, Olivier Bachmann and Réka Lukács

as an original research article for consideration of a publication in the Contributions to Mineralogy and Petrology.

In this paper, we describe the petrology and geochemistry of a crystal-rich dacitic lava dome, which represents an erupted crystal mush. The Haramul Mic lava dome extrusion took place at 156 ka after long, at least 100 kyr dormancy and this was the onset of the development of the Ciomadul volcanic complex, the youngest volcano of eastern-central Europe. Ciomadul is a typical long-dormant PAMS volcano (erupted last time more than 10 ka but there are indications that a subvolcanic Potentially Active Magma Storage can still exist) and this manuscript along with our former publications in this journal (Kiss et al. 2014; Cserép et al. 2023) can highlight the attention to such apparently inactive volcanic systems which pose underrated hazard and risk.

The mineral assemblage (plagioclase, amphibole, biotite, apatite, titanite and zircon) and the fresh interstitial glass have relatively homogeneous major and trace element composition suggesting near-equilibrium pre-eruptive state. This enabled us to determine a wide set of amphibole-melt trace element partition coefficient data as well as the mineral-melt partition coefficients for titanite, zircon, plagioclase and biotite. These can be used for low temperature (< 750 °C), near-eutectic silicic volcanic and plutonic systems. Such an extensive  $K_d$  database for similar magmatic systems is still quite rare in the literature.

In contrast to the subsequent volcanism of Ciomadul, there is no sign for mafic magma recharge as eruption initiation. Thus, volatiles could have played a role such as 'second boiling' or volatile oversaturation due to degassing from underplated mafic magma. The

1 Ciomadul provides an example where various processes can contribute to eruption initiation  
2 reflected by the compositional characteristics of the crystal cargo even though the  
3 composition of the erupted magma is similar.  
4

5 We believe that our results are relevant and novel, and that the conclusions could be of wide  
6 interest to the scientific community.  
7

8  
9 All authors contributed to various parts of this study from the conceptualization, through field  
10 works, lab-works and petrological and geochemical analyses, data procession and  
11 interpretation, discussion of the results as well as writing the manuscript.  
12  
13

14 All co-authors have seen and agree with the content of the manuscript and there is no financial  
15 interest to report.  
16

17 We confirm that this work is original and has not been published elsewhere, nor is it currently  
18 under consideration for publication elsewhere.  
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22  
23 Sincerely Yours,  
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# **Petrogenesis and amphibole–melt trace element partitioning of the 156 ka Haramul Mic crystal-rich dacite, Ciomadul, Romania**

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## Abstract

The 156 ka Haramul Mic crystal-rich (ca. 40 vol%) dacitic lava dome represents eruption of a crystal mush. It is the first product of the Ciomadul Volcanic Complex, eastern-central Europe, following at least 100 kyr dormancy. The mineral phases (plagioclase, amphibole, biotite, apatite, titanite and zircon) and the interstitial glass have relatively homogeneous major and trace element composition suggesting near-equilibrium pre-eruptive state. Geothermobarometric and hygrometric calculations indicate a near-eutectic, low-temperature (730 °C), low-pressure (200–400 MPa), wet (6.5–6.7 wt% melt H<sub>2</sub>O content) and oxidized silicic magmatic system. A wide set of amphibole-melt trace element partition coefficient data is provided, which can be used for such low temperature silicic volcanic and plutonic systems. Additionally, mineral-melt trace element partition coefficients are also calculated for titanite, zircon, plagioclase and biotite phases. Quality-check was performed by Onuma-plots and the crystal lattice parameters were determined which fit well with published results. In contrast to the subsequent eruptions, there is no sign for mafic magma recharge as eruption initiation. Thus, volatiles could have played a role such as 'second boiling' or volatile oversaturation due to degassing from underplated mafic magma. The lack of reaction corona around amphibole macrocrysts suggests relatively fast magma ascent, although this can also be due to the still relatively high water content (>4 wt%) of the melt, where amphibole remained stable. The Ciomadul provides an example where various processes can contribute to eruption initiation reflected by the compositional characteristics of the crystal cargo even though the composition of the erupted magma is similar.

## Keywords

Crystal-rich dacite, crystal mush, pre-eruptive magma condition, amphibole, partition coefficient, eruption initiation, hydrous magma, Ciomadul

## Introduction

Understanding magma dynamics that lead to volcanic eruptions is crucial for improving volcanic hazard assessment, risk mitigation, and eruption forecast (Saunders et al. 2012; Pankhurst et al. 2018; Re et al. 2021; Ostoreno et al. 2022; Kent et al. 2023; Hughes et al. 2025). Geophysical and geodetic monitoring instruments have been proven to be effective in detecting signals from crustal magma bodies, however, only petrological studies on volcanic rocks from past eruptions can provide insights into magmatic processes that produce these signals. This is particularly true for potentially active volcanoes, which have not exhibited historical eruptions and spend extended periods (sometimes thousands or even ten thousand years) in dormancy. Growing geophysical studies (e.g., Sparks et al. 2008; Hill et al. 2009; Comeau et al. 2015; Harangi et al. 2015b; Trasatti et al. 2018; Cordell et al. 2020; Paulatto et al. 2022; Pang et al. 2025) suggest that melt-bearing magma reservoirs may persist beneath these quiescent volcanoes, warning us that they can potentially be reactivated and erupt again. There are even seemingly inactive, long-dormant volcanoes where the last eruption took place more than 10 ka, yet there are indications for existence of reactivable subvolcanic magma reservoir. For them, Harangi et al. (2015a, b) suggested the term volcano with Potentially Active Magma Storage (PAMS) emphasizing the need of attention since they pose unknown hazard and risk.

Petrological studies can reveal past volcanic histories and possible pathways of reactivation. Integrated studies involving the various phases (e.g., different minerals and glass) within volcanic rocks can lead to the quantification of the magma storage conditions and the timescale of magmatic processes as well as constraints to the eruption initiation (Bachmann and Huber 2016; Putirka 2017; Ubide and Kamber 2018; Costa et al. 2020; Petrelli and Zellmer 2020; Kent et al. 2023). Among these approaches, the determination of trace element partitioning between mineral and melt phases in different volcanic systems is essential to conduct petrogenetic model calculations aimed at discerning the chemical evolution of magmas (Blundy and Wood 2003; Padilla and Gualda 2016; Shimizu et al. 2017).

Ciomadul, in Romania, is the youngest volcano in the eastern-central Europe, with its last eruption at 30 ka (Harangi et al. 2010, 2015a, 2020, Szakács et al. 2015; Karátson et al. 2019; Molnár et al. 2019). In spite of the long quiescence period, there are indications of a melt-bearing magma reservoir beneath it (Popa et al. 2012; Harangi et al. 2015b; Comeau et al. 2024) and Szakács and Seghedi (2013) have already highlighted the potential volcanic hazard of Ciomadul. Petrological studies (Kiss et al. 2014; Laumonier et al. 2019; Lukács et al. 2021; Cserép et al. 2023) have characterized the magma evolution at various stages and emphasized

the role of a long-standing low-temperature felsic crystal mush in the upper crust as well as the more mafic recharge magmas. However, a comprehensive study providing a complete set of mineral–melt partition coefficients directly from natural samples in a low-temperature silicic system to quantitatively support petrologic investigations of PAMS volcanoes is still missing. To fill this gap, the present paper aims to characterize the first volcanic products emplaced after more than 100 kyr dormancy and represent the onset of development of the Ciomadul Volcanic Complex. We show that crystal-rich dacitic magma of Haramul Mic, extruded at 156 ka, is a rare example of the evacuation of a near-eutectic crystal mush. The striking homogeneity of various mineral phases and the glass phase enables us to determine mineral–melt partition coefficients, with special emphasis on amphibole–melt trace element distribution in a low-temperature silicic magmatic system. Since amphibole plays an important role in the evolution of arc magmas and controls the composition of both erupted magmas and plutons (Davidson et al. 2007, 2013), the results of this study can also help quantitatively test petrogenetic processes and crustal evolution worldwide.

## Geological background

In the Carpathian–Pannonian Region (eastern-central Europe), a wide range of volcanic activity has occurred for the last 20 Ma (Szabó et al. 1992; Seghedi et al. 1998, 2004, 2005; Harangi 2001; Konečný et al. 2002; Harangi and Lenkey 2007; Lexa et al. 2010; Seghedi and Downes 2011; Lukács et al. 2018; Harangi et al. 2025). It was partly coeval with the formation of the Pannonian Basin by lithospheric thinning but continued even during the post-rift period and was post-collisional at the eastern area. The Ciomadul, located at the southeastern end of the 160 km long Călimani–Gurghiu–Harghita volcanic chain along the East Carpathians, is the youngest manifestation of this long-lasting volcanism (Szakács et al. 1993, 2015; Harangi et al. 2010, 2015a; 2020; Karátson et al. 2016; 2019; Molnár et al. 2018, 2019; Lukács et al. 2021, Seghedi et al. 2023a, b). Volcanic activity started at Călimani about 11.3 Ma, building up andesitic-dacitic volcanic complexes and then, it shifted gradually south-eastward producing younger and smaller volume volcanoes (Pécskay et al. 1995; Szakács and Seghedi 1995; Mason et al. 1996; Karátson and Tímár 2005; Seghedi et al. 2023b). Since most of the volcanism took place after the 10–12 Ma continent–continent collision at the East Carpathians (Maţenco et al. 2007, 2010), it can be considered as a post-collisional type (Seghedi et al. 2011, 2019). Volcanic activity in the South-Harghita started about 5.3 Ma (Pécskay et al. 1995; Seghedi et al. 2023a) and ended with the last eruption of Ciomadul about 30 ka (Vinkler et al. 2007; Harangi et al.

2010, 2015a, 2020; Karátson et al. 2016; Molnár et al. 2019). Composition of the erupted  
magmas in South Harghita changed at about 2 Ma and then at 1 Ma, becoming adakitic in  
character with high-Ba-Sr and low-HREE during the youngest period (Szakács et al. 1993;  
Seghedi et al. 1995, 2023a; Molnár et al. 2018, 2019).

Ciomadul is located near the geodynamically active Vrancea seismic zone where  
frequent intermediate-focus earthquakes indicate a decoupling between a near-vertical  
descending slab and the overlying crust, either by slab break-off or delamination (Sperner et al.  
2001; Fillerup et al. 2010; Ismail-Zadeh et al. 2012; Ducea et al. 2020). In addition to the  
Ciomadul volcanism, a temporarily partly overlapping volcanic activity took place about 50 km  
to distance west, forming an alkali basalt volcanic field (1.3–0.7 Ma; Downes et al. 1995;  
Harangi et al. 2013; Seghedi et al. 2016; Ducea et al. 2020). However, the relationship between  
the Vrancea slab descent and the two types of volcanism is still debated (e.g., Seghedi et al.  
2023a).

Ciomadul (Fig. 1a) consists of an older lava dome field and a younger central volcanic  
complex (Molnár et al. 2019; Lukács et al. 2021). Volcanic activity can be divided into two  
periods: the Old Ciomadul Eruptive Period (OCEP), lasted from ~1 Ma to ~350 ka (Molnár et  
al. 2018), during which sporadic, andesitic, dacitic and shoshonitic lava domes were formed.  
This period involved three epochs interrupted by more than 100 kyrs long dormant intervals.  
During the Young Ciomadul Eruptive Period (YCEP), that started about 160 ka, the central  
Ciomadul volcanic complex was formed fed by homogeneous dacitic magma. It is subdivided  
into two eruptive epochs: a 160–100 ka lava dome building activity (Karátson et al. 2013, 2016;  
Molnár et al. 2019; Lukács et al. 2021) followed by a dominantly explosive eruptive epoch with  
small lava dome extrusions about 57–30 ka (Vinkler et al. 2007; Harangi et al. 2015a; Karátson  
et al. 2016; Molnár et al. 2019; Lukács et al. 2021; Cserép et al. 2023). Initial eruptions of the  
YCEP formed an amalgamated, elongated lava dome complex, while the younger lava domes  
extruded south-eastward and between them two explosion craters were excavated.

The Haramul Mic lava dome, studied in this paper, is the oldest one in the YCEP (157  
ka; Molnár et al. 2018, 2019), located a little bit separated, 4 km northeast from the main lava-  
dome complex (Fig. 1a). This eruption took place after a long, almost 200 kyrs quiescence  
period (Lukács et al. 2018, 2019; Molnár et al. 2018, 2019) and initiated the most voluminous  
YCEP eruptions. Ages calculated from isotopic data measured in zircon crystal interiors and  
outer rims spread from the eruption age back to 450 ka. (Lukács et al. 2021). This indicates that

continuous crystallization occurred in the magma chamber even during the long-dormant interval, i.e. some melt fraction was always present in the magma reservoir.

## Samples and analytical methods

Haramul Mic is an 880.7 m asl high, flat lava dome (Fig. 1b) covering an area of ~1.3 km<sup>2</sup> and has 0.15 km<sup>3</sup> volume (Karátson et al. 2019). It is built up by homogenous, unaltered, crystal-rich dacite lava blocks. The lava dome rock contains abundant ~0.1–2 cm diameter felsic crystal clots delimited by a curved boundary line. Dark biotite–plagioclase enclaves (cm-sized) occasionally occur. Several representative samples were collected from the lava dome, which show homogeneous petrological and geochemical features (Electronic Supplementary Material 1; ESM 1).

After macroscopic investigation, 10 thin sections were made and examined under polarizing petrographic optical microscope and with an AMRAY 1830 I/T6 scanning electron microscope (SEM) at the Department of Petrology and Geochemistry of Eötvös Loránd University (Budapest, Hungary). Crystallinity (area %) was determined using the public domain software of ImageJ and CalcImage®. Based on the petrographic studies, two thin sections were selected for electron microprobe analysis and laser ablation ICP-MS measurement.

Major element chemical composition of the main mineral and glass phases were determined using JEOL JXA 8900 RL electron microprobe equipped with WDS at Geozentrum, Georg-August University of Göttingen (Germany). Analytical conditions involved 15 kV acceleration voltage, 15 nA beam current and 5–10 µm defocused beam diameter. Albite, sanidine, wollastonite, TiO<sub>2</sub>, rhodonite, hematite, olivine, anorthite, Cr<sub>2</sub>O<sub>3</sub>, NiO, NaCl and celastine standards were used for amphibole and biotite measurements. For plagioclase, albite, sanidine, hematite, olivine, anorthite, SrTiO<sub>3</sub> and celsian standards were applied. For glass phase albite, sanidine, anorthite, wollastonite, TiO<sub>2</sub>, rhodonite, hematite, NaCl, olivine, ScPO<sub>4</sub>, celsian, VG2 glass, topas and SrTiO<sub>3</sub> were used. The Phi-rho-Z correction is used for data processing. The Fe<sup>3+</sup> value in amphibole was estimated based on the IMA recommendation (Leake et al. 1997; Schumacher 1997), which is consistent with the method suggested by Holland and Blundy (1994). The full dataset with detection limits and uncertainties is given in the Electronic Supplementary Material (ESM 1).

A 22 mm x 22 mm area of the Haramul Mic thin section was scanned and measured by electron microprobe equipped with WDS at ETH Zurich. For the thin section map measurement

the following conditions were used: 15 keV acceleration voltage, 200 nA beam current, defocused beam at 25  $\mu\text{m}$  to match the pixel size, 40 ms per pixel (= dwell time), two passes with the following elements (all are K-alpha X-ray lines): Spectrometer 1: Si, Al on TAP; Spectrometer 2: Ca, Cr on PET; Spectrometer 3: K, Ti on PET-L; Spectrometer 4: Fe, Mn on LiF-H; Spectrometer 5: Na, Mg on TAP-H. The different mineral phases were selected with the help of major element concentration filtering in CalcImage® software. The concentration ranges used as filters were the following: for plagioclase  $\text{SiO}_2$  <80 wt%,  $\text{K}_2\text{O}$  <2 wt%,  $\text{TiO}_2$  <2 wt%,  $\text{MgO}$  <2 wt%, total oxides 90-110 wt%; for amphibole  $\text{Al}_2\text{O}_3$  <10 wt%,  $\text{MgO}$  min. 5 wt%, max. 16 wt% and total oxides 90-99 wt%; for titanite  $\text{SiO}_2$  min. 20 wt%, max. 158 wt%,  $\text{CaO}$  min. 15 wt%, max. 52.4 wt%,  $\text{TiO}_2$  min. 20 wt%, max. 62.8 wt% and total oxides 90–110 wt%; for oxides  $\text{FeO}$  min. 70 wt%, max. 84 wt% and total oxides min. 85 wt%, max 110 wt%. Amphibole and plagioclase major element concentration data were extracted from CalcImage®. In case of amphibole and plagioclase full data, the used pixel width was 10. For plagioclase phenocryst rims, the used pixel width was 2 during data extraction, and points were selected manually. Data was saved from 24646 pixels for amphibole, from 95060 pixels for plagioclase (whole crystals) and 706 pixels for plagioclase crystal rims. From the raw extracted data, cation numbers were calculated and filtered for outlier values. During amphibole data outlier filtering, it was examined whether the cation numbers are reliable and specific for amphibole. Those extracted data points were deleted from the database where cation numbers ( $\text{Fe}^3$ , Na, Al) had negative values. In case of plagioclase data, there were only a few outlier values where some cation numbers had extreme values. Coloured element maps were also created in CalcImage® software based on the relative concentration of major elements (oxides).

In situ trace element concentrations in the groundmass glass, plagioclase, amphibole, and biotite phenocrysts, plus accessory zircon and titanite crystals were measured by LA-ICP-MS. The Laser Ablation device consisted of a Teledyne / Photon Machine G2 coupled with a Thermo Fisher Scientific iCAPQ quadrupole-based ICP-MS at the Department of Physics and Geology, University of Perugia (Italy). Ablation was performed with a 193 nm ArF excimer laser beam under He atmosphere. All spots were ablated with a fluence of 3.5  $\text{J}/\text{cm}^2$  and the laser pulse repetition rate was 10 Hz. Argon and  $\text{N}_2$  were added to the helium plus ablated material stream before reaching the ICP unit to stabilize the plasma and increase the signal, respectively. The NIST 610 glass was used as reference material for the calibration of each experiment (Perkins et al. 1997; Petrelli et al. 2016). The quality control was the USGS BCR 2G reference material, analysed as unknown (Jochum et al. 2005). Due to varying crystal size

and quality, samples were ablated with crater diameters within the range of 15–30  $\mu\text{m}$ . A laser diameter of 15 and 20  $\mu\text{m}$  was used for glass, 30  $\mu\text{m}$  for plagioclase and amphibole, 20 and 25  $\mu\text{m}$  for biotite, titanite and zircon. Reference materials were ablated with the same crater diameter as the samples; hence, a new experiment was set up for each diameter size. Data reduction was implemented using the Iolite4 software. In case of the groundmass glass, plagioclase, biotite, and zircon, the utilized internal standard was Si, for amphibole we used Ca, both previously determined by EMPA. For zircon and titanite, the stoichiometric ratio was applied. In homogeneous or small crystals and the groundmass glass, discrete spots were selected. In mm-sized, euhedral, slightly zoned plagioclase and amphibole phenocrysts, core-to-rim transects were acquired to reveal concentration changes within crystals. To reduce the LA-ICP-MS data, “Trace\_Elements” Data Reduction Scheme of the Iolite4 software package (Paton et al. 2011) was used. The full dataset with detection limits and uncertainties is given in the Electronic Supplementary Material (ESM 2).

The mineral assemblage of the Haramul Mic dacite (Molnár et al. 2018) is suitable to perform various thermobarometry calculations. Amphibole–plagioclase thermobarometry calculations were conducted using Thermobar – Amphibole Thermobarometry and Chemometry module (Wieser et al. 2022). Both minerals show relatively homogeneous chemical compositions with little internal variance. As input data, all plagioclase measurement data were paired with all amphibole measurement data. Within Thermobar (Wieser et al. 2022), the Al-in-hornblende barometer experimentally calibrated by Schmidt (1992) was used to calculate the initial amphibole-only pressure and combined it with the amphibole–plagioclase thermometer, equation B by Holland and Blundy (1994) to get the initial temperature calculation. Then, the probability density estimations of pressure and temperature were computed by 100 iterations of the Anderson and Smith (1995) barometer and the Holland and Blundy (1994) thermometer, equation B, respectively. The temperature and pressure values were also calculated using the spreadsheet from Anderson et al. (2008), which applied equation B of the Holland and Blundy (1994) thermometer. The pressure was calculated iteratively based on the works of Schmidt (1992) as well as Anderson and Schmidt (1995). The obtained pressure values were compared with those obtained by the method of Mutch et al. (2016) with the notion that quartz and K-feldspars were not observed in the mineral assemblage.

Zirconium in titanite thermobarometry was calculated combining the pressure equation of Erdmann et al. (2019) with the thermometer by Hayden et al. (2008), using 0.6  $\text{TiO}_2$  activity.

Titanium in zircon thermometry by Ferry and Watson (2007) was applied calculating with 0.6 TiO<sub>2</sub> activity and 1 SiO<sub>2</sub> activity.

The homogeneous mineral and glass composition enabled us to determine mineral/melt trace element partition coefficients. Uncertainty estimation for the calculated mineral/melt trace element partition coefficients was performed with standard error estimate (SEE = standard deviation / square root of n, where n is the number of measurements) and uncertainty propagation. The calculated partition coefficients were plotted on Onuma-diagrams (Onuma et al. 1968), where the Simple Fit method by Dalou et al. (2018) was used with standard error estimate (SEE) as errors. Curve-fitting was based on the equation of Blundy and Wood (1994, 2003) with 730 °C (1003 °K) average temperature (derived from the thermometric calculations) that can be considered as being representative for the entire magmatic system of the Haramul Mic. For amphibole, titanite and zircon crystals, the cleared average trace element concentration data was used. Melt phase is represented by the average trace element concentration data of interstitial glass in the felsic clots. Onuma-plots with the Simple Fit method can be considered statistically acceptable if Nash > 0.95 and RMSE (root mean square error) is not >> 1 (Dalou et al. 2018). The full dataset with errors is given in the Electronic Supplementary Material (ESM 3).

## Results

### Petrography

The Haramul Mic lava dome host rock is crystal-rich with an average macro- and mesocryst (as defined by Zellmer 2021) content of 27–30% and an additional significant amount (10–16%) of crystal clots (Fig. S1a in ESM4). The macro- and mesocryst assemblage consists of ~75–80% plagioclase, ~10–15% amphibole, and ~10% biotite with <1%, while accessory minerals are represented by apatite, titanite, zircon and Fe-Ti oxide minerals. Petrographic features were studied using high-resolution element mapping of an entire thin section of a representative lava dome rock (Fig. 2a and Fig. S1b–d in ESM4). Plagioclase is the most ubiquitous mineral phase occurring often as a group of crystals. They are up to a few millimetre-sized, mostly euhedral and usually show slight oscillatory zoning. There are distinct crystal size groups with strong contrasts. Approximately one-third of them (mostly the >1 mm size crystal population) comprises more calcic core surrounded by a spongy zone followed by oscillatory rim and flat, sharp rims of crystal facets without resorption features (Fig. 2b and Fig.

S1b in ESM4). Some of them involve amphibole or biotite inclusions. Amphibole macro- and mesocrysts are also euhedral and subhedral having flat, sharp margins (Fig. 2d,e). They do not show clear compositional zonation except for slight patchy structure in their interior (Fig. 2e). They occasionally involve biotite and apatite inclusions. Biotite mesocrysts are also subhedral to euhedral (Fig. 2e and Fig. S1d in ESM4) and often contain plagioclase inclusions. The groundmass is glassy and unaltered consisting of abundant sheared vesicles and minor plagioclase microlites (Fig. 2b,d). Therefore, the texture of the rock is hyalopilitic.

Light coloured, irregular, often round-shaped crystal clots with a few mm-to-cm size commonly occur in the host rock (Fig. S1a in ESM4). These felsic crystal clots contain ~0.5–2 mm euhedral–subhedral tabular plagioclase crystals (>75%), which show moderate patchy and oscillatory zoning. Plagioclase crystals are often grown on each other. Additional crystals are ~0.1–0.5 mm euhedral–subhedral amphibole and mostly subhedral biotite in various amounts (Fig. 2c). In a few cases amphibole also forms crystal clots within the larger felsic clots. Interstitial glass phase between crystals covers about 10%. It is microlite-free and contains rounded vesicles (Fig. 2c). Hence, the texture of the crystal clots is dikytaxitic or glomerophytic.

## Geochemistry

### *Bulk rock composition*

Bulk rock composition of two samples from different parts of the petrologically homogeneous lava dome was presented by Molnár et al. (2018). They have identical compositions (ESM1), akin to most of the other lava dome rocks and fall into the more silicic compositional group of Ciomadul. They are potassic trachydacite having  $K_2O=3.7$  wt%,  $K_2O/Na_2O$  ratio of 0.82–0.87 at  $SiO_2$  of 67.2–67.4. They have similar trace element patterns as the other lava dome rocks (Fig. 3a; Molnár et al. 2018 and Fig. S2a in ESM4). The most remarkable features are the elevated Ba and Sr concentration values (Ba=1192–1208 ppm; Sr=1046–1078 ppm; Fig. 3a), which are a little bit lower than those of the pumices (Ba=1100–1800 ppm; Sr=1200–1800 ppm; Molnár et al. 2019) and the depleted heavy rare-earth element (HREE;  $La/Yb_{cn}=26.8$ ;  $Yb_{cn}=5.6$ ; Fig. 3a and Fig. S2a in ESM4).

### *Groundmass glass and interstitial glass*

Glassy groundmass both in the host rock and within the felsic clots has high-Si potassic rhyolitic composition ( $SiO_2 = 77.0–80.2$ wt%;  $K_2O = 5.0–5.7$  wt% in volatile-free 100% data).

The most evolved composition is shown by the glass in the host rock consistent with the occurrence of small amount of plagioclase microlites, while glass in the felsic clots is less silicic ( $\text{SiO}_2 = 77.0\text{--}79.0$  wt%). The calculated volatile content (deviation of the total of major element oxides from 100 wt%) of the glass is 3.6–5.1wt% in the felsic crystal clots, whereas 2.5–3.1wt% in the groundmass of the host rock. The trace element concentration characteristics are presented by primitive mantle (McDonough and Sun 1995) normalized multielement plots (Fig 3a and Fig. S3a,b in ESM4) and chondrite (McDonough and Sun 1995) normalized rare-earth element plots (Fig. S2a in ESM4). The groundmass glass has elevated LIL elements, Th-U values as well as positive Pb anomaly, while some of the HFS elements (e.g., Nb, Ta, Ti) and the heavy REE are depleted. Negative Eu anomaly cannot be noticed. All transitional metals (Ni, Cr, Co, V) have low concentrations, while Sc is between 13 and 16 ppm (Table 1). There are only subtle differences in the trace element properties of the glass in the groundmass and the felsic clots (Fig. 3a and Fig. S2a and S3a,b in ESM4). In the latter one, glass composition is more homogeneous (Fig. S3b in ESM4), whereas in the groundmass glass Sr shows some variation, presumably because of the plagioclase microlites (Fig. S3a in ESM4). In the felsic clot glass, Ba content is 1200–1450 ppm, while Sr is much lower compared to the bulk rock (Fig. 3a), i.e., it ranges between 120 and 190 ppm. In contrast, in the groundmass glass, they are 900–1520 ppm and 160–390 ppm, respectively. Noteworthy, no differences in other trace element content can be observed between the glass phases, except for Li and in a lesser extent in Sc. Most of the Li concentration is between 55 and 70 ppm in the glass of the felsic clots (Table 1), whereas it is significantly less in the groundmass glass of the host rock (Li=35–45 ppm).

### ***Plagioclase***

Plagioclase composition is in the range between 25 and 50 mol% anorthite (An) content, i.e., all plagioclase macrocrysts are andesine–oligoclase (Fig. 4a). The most dominant composition is between 25 and 35 mol% An content, irrespective whether the plagioclase is in the host rock or the felsic clots. The more calcic core of the large macrocrysts has 45–50 mol% An content (Fig. 4b). The FeO concentration usually remains in a restricted interval, between 0.1 and 0.2 wt%, irrespective of the An content.

Trace element concentration was measured in plagioclase macrocrysts both in the host rock and the felsic clots. In addition, trace element profiles were determined along core-to-rim transects for 4 plagioclase macrocrysts. In general, Sr content is between 2000 and 4400 ppm (Table 1), indicating that plagioclase is responsible for the high Sr content of the bulk rock. It

shows a positive correlation with the An (Ca) content. The Ba concentration is in the range of 250–970 ppm irrespective of the An content. The light REE and Eu show also a positive correlation with Sr and An contents. No difference in trace element composition can be observed between the plagioclases in the host rock and the felsic clots. There are two exceptions, the first one is Li (it is in the range between 10 and 30 ppm), which generally is lower in the plagioclase macrocrysts of felsic clots. The Sc content is, however, systematically higher (Sc=6–11 ppm) in the plagioclase of felsic clots compared to the host rock plagioclase (Sc=4.5–7.5 ppm). The Li correlates positively with Sr and An contents. Along the profiles, Li drops at the outermost margin, whereas it is relatively homogeneous along most of the macrocrysts. Trace element values do not change from core to rim (Sr=2000–3000 ppm), except for a few, strongly zoned macrocrysts (Fig. S4 in ESM4). In the latter ones, the Sr profiles show marked decrease in concentration at the major resorption surfaces. The high-Ca core has the highest Sr content (3500–4000 ppm), whereas in the margin, it is in the range of 2000–2500 ppm, similar to Sr values in other macrocrysts. Here, the core is enriched in light REE (La–Nd; Fig. S4 in ESM4), whereas no variation is observed in the middle REE (Sm–Eu).

### ***Amphibole***

Amphibole is the predominant mafic mineral in the dacite. It occurs as macro- and mesocrysts in the dacitic lava dome rock and also in the felsic crystal clots. They are also Ca-amphibole, classified mostly as magnesio-hornblende and some of them are pargasite based on Hawthorne et al. (2012; Fig. 5a).

The amphibole compositions are fairly homogeneous compared to the amphibole occurring in other lava dome rocks (Kiss et al. 2014) and pumices (Cserép et al. 2023) and do not show differences in the host dacite and the felsic clots (Fig. 5ad–f). The slight compositional variation is related primarily to the patchy zoned macrocrysts (Fig. 2e). They have low Al<sub>2</sub>O<sub>3</sub> content ranging between 6 and 10 wt%, similar to the magnesio-hornblende group of the Ciomadul Mic lava dome rocks (Kiss et al. 2014) and the Group-1 amphibole in the younger pumices (Cserép et al. 2023), while SiO<sub>2</sub> is 44–49 wt%, TiO<sub>2</sub> is ~0.5–1.5 wt% and MgO is 12–14.5 wt% in most of the measured spots. In all cases, concentration of Al<sub>2</sub>O<sub>3</sub> has a positive correlation with FeO and TiO<sub>2</sub> content, while a negative correlation is found with SiO<sub>2</sub> and MgO concentration (Fig. 5c,d). There is a linear correlation between tetrahedral Al (Al<sup>IV</sup>) and Ti, (Na+K)<sup>A</sup> and Mg (Fig. 5d–f), but no significant correlation was observed with the octahedral Al (Al<sup>VI</sup>) (Fig. 5b). These linear relationships imply dominant, temperature-dependant Si<sup>IV</sup> +

$()^A \rightarrow \text{Al}^{\text{IV}} + (\text{Na}+\text{K})^A$  (edenite) and  $\text{Si}^{\text{IV}} + \text{Mg}^{\text{VI}} \rightarrow \text{Al}^{\text{IV}} + \text{Ti}^{\text{VI}}$  (Ti-tschermakite) substitution mechanisms within crystal lattice (Bachmann and Dungan 2002).

Trace element composition of amphibole is also rather homogeneous and similar in the host rock and in the felsic crystal clot (Fig. 3b and Fig. S2b and Fig. S5a,b in ESM4). Chondrite normalized (McDonough and Sun 1995) rare-earth element (REE) patterns show slight light REE enrichment and slight negative Eu-anomaly (Fig. S2b). A positive correlation can be observed between Ba, Sr, La and Zr, where the crystal rims are characterized by the lowest values (Fig. S6 in ESM4). Both Ba and Sr have relatively low concentration, ranging between 50 and 200 ppm similarly as the magnesio-hornblende in the Ciomadul Mic dacite (Kiss et al. 2014). The ~100–150 ppm fluctuations cannot refer to analytical errors only, as the uncertainty of detection (2 standard errors) for Sr and Ba are 4–17 ppm and 3.5–18 ppm, respectively. Yttrium is mostly between 25 and 50 ppm, whereas Sc is relatively high, ranging from 56 and 80 ppm.

#### ***Biotite***

All the measured biotite crystals have  $\text{Mg}/\text{Fe}^{\text{tot}}$  ratios around 1.4, Mg values of 3.1 and low,  $<0.12 \text{ Al}^{\text{vi}}$  value. Consequently, all biotites and can be separated from phlogopite according to the nomenclature of Speer (1984). The  $\text{TiO}_2$  content is ~3.4–3.9 wt%, while MgO and  $\text{Al}_2\text{O}_3$  concentrations are ~13–14 wt% and 13.5–14.5 wt%, respectively. As for the trace elements, Cr content is ~120–190 ppm, Sc is ~13.5–16 ppm and Ba is mostly between 3000 and 6000 ppm.

#### ***Accessory minerals***

Titanite and zircon are common accessory minerals in addition to apatite. Both mineral phases have homogeneous trace element compositions (Fig. 3c,d and Fig. S2c,d, S7a,b in ESM4). In the titanite, Hf concentration is ~22–35 ppm, Zr concentration is 300–460 ppm and have similar, slightly light REE-enriched patterns with only a slight negative Eu-anomaly. The Hf concentration is mainly ~9000–11,000 ppm and Ti is between ~1.8 and ~8.2 ppm. The Th/U ratio is ~0.4–1.4, the Yb/Dy ratio is ~4–8. They show moderate negative Eu anomaly ( $\text{Eu}/\text{Eu}^*=0.5\text{--}0.85$ ), and positive Ce anomaly ( $\text{Ce}/\text{Ce}^*=43\text{--}255$  based on Loader et al. 2017). With increasing Hf concentration, Yb/Dy is increasing, but Th/U and Ti are decreasing.

## Discussion

### Pre-eruption magma storage conditions

The Haramul Mic dacite lava dome represents one of the initial eruptions of the Ciomadul volcanic complex after a long (>100 kyr) quiescence period (Molnár et al. 2018, 2019). Eruption took place offset of the main volcanic massif (Fig. 1a) at 157 ka, when 0.15 km<sup>3</sup> coherent lava body extruded and formed a flat, pancake-like lava dome (Fig. 1b). The low-aspect ratio dome morphology is typical of silicic magmas (Myers et al. 2024). Remarkably, it is a highly crystalline dacite with >40 vol% crystal content, in which 10–16 vol% is felsic crystal clot sitting in rhyolitic glassy matrix. Mineral chemical composition, both for major and trace elements, is fairly homogeneous, particularly of plagioclase and amphibole. Crystals have sharp margins; they are euhedral to subhedral in shape. These petrological features suggest that the erupted magma could represent evacuation of a portion of felsic crystal mush. The mineral assemblage enables us to describe the pre-eruption conditions and determine the trace element partition coefficients. This will be followed by discussion of possible eruption initiation mechanisms.

The Haramul Mic dacite contains plagioclase, amphibole and biotite macrocrysts in addition to accessory minerals of titanite, apatite, Fe–Ti oxides and zircon. Amphibole and plagioclase major element point measurements were compared to the scanned quantitative EMPA data set. Sampling in this case can be considered statistically as more objective than the manually selected spot measurements, although with less accurate results. Distribution of the scanned EMPA data is fairly similar to the distribution of the point measurement data (Fig. S8 and Fig. S9 in ESM 4) proving that our spot data are representative. There is no significant difference between crystal core and rim composition. Plagioclase has An content mostly between 25 and 35 mol%, dominantly 30–35 mol%, which is very similar to that found for the plagioclase composition of Pinatubo dacite (Pallister et al. 1996; Tamura and Nakagawa 2023). Amphibole is also compositionally relatively homogeneous, having Al<sub>2</sub>O<sub>3</sub> content between 6.5 and 8.5 wt% and Mg-value ( $\text{Mg}^{2+}/\text{Mg}^{2+}+\text{Fe}^{\text{tot}}$ ) ranging between 0.58 and 0.66 (Fig. 6). This is identical with the low-Al amphibole group recognised in the Ciomadul volcanic rocks (Kiss et al. 2014; Laumonier et al. 2019; Cserép et al. 2023). However, so far only the Haramul Mic dacite contains solely this amphibole population, whereas in the other volcanic rocks, a high-Al and high-Mg amphibole group also occurs representing the various recharge magmas. The existence of the low-Al and low-Mg amphibole throughout the 156–30 ka Ciomadul dacites confirms a long-living low-temperature felsic crystal mush body in the upper crust as implied

by zircon studies (Harangi et al. 2015a; Lukács et al. 2021). Noteworthy, this crystal mush-type amphibole composition was observed in other dacitic volcanic rocks worldwide, such as in the Fish Canyon Tuff (Bachmann and Dungan 2002), Soufrière Hills, Montserrat (Murphy et al. 2000; Rutherford and Devine 2003; Buckley et al. 2006; Humphreys et al. 2009), Pinatubo (Pallister et al. 1996; Tamura and Nakagawa 2023), Methana, Greece (Popa et al. 2020) and in various dacites from the Andes such Taápaca (Rout et al. 2021) and Cerro La Torta (Prat et al. 2023) (Fig. 6).

The linear correlation between tetrahedral Al ( $Al^{IV}$ ) and Ti,  $(Na+K)^A$  and Mg in the Haramul Mic amphibole suggests that the substitution mechanisms (edenite and Ti-tschermakite exchange) in crystal lattice are controlled by temperature without significant pressure variation (Fig. 5d–f). Quartz and K-feldspar do not occur; however, the groundmass is Si-rich potassic rhyolite ( $SiO_2=74\text{--}77$  wt%;  $K_2O=4.5\text{--}5.5$  wt%;  $K_2O/Na_2O=1.8\text{--}2.2$ ), therefore the melt could have been  $SiO_2$  saturated during crystallisation. However, quartz and K-feldspar crystals are occasionally found in the 135 ka Ciomadul Mic lava dome (Kiss et al. 2014) in addition to the other macrocrysts, therefore it can be assumed that they were present in the crystal mush but entered the melt phase during the mush reactivation. Thus, the Al-in amphibole geobarometer with the latest calibration of Mutch et al. (2016) was also applied to constrain the crystallization pressure, i.e., the depth of magma reservoir in addition to the application of the amphibole–plagioclase thermobarometry (Schmidt 1992; Holland and Blundy 1994; Anderson and Schmidt 1995). This was completed by Zr-in-titanite (Hayden et al. 2008; Erdmann et al. 2019) and Ti-in-zircon (Ferry and Watson 2007) thermobarometry calculations with  $TiO_2$  activity of 0.6 and  $SiO_2$  activity of 1.

The estimated crystallization pressure range for the Haramul Mic dacitic magma is 200–400 MPa for the felsic clots and 100–500 MPa regarding the host dacite, but with much smaller range of 260–310 MPa at 95% probability (Fig. 7). The pressure values (200–400 MPa range and 276 MPa median value) calculated by the Mutch et al. (2016) equation fit perfectly this result as well as that given by Anderson et al. (2008), whereas the titanite barometry gives the lower end of this pressure range (200–215 MPa). Considering an average upper crustal density of  $2700\text{ kg/m}^3$ , the depth of the magma reservoir from where the erupted magma was withdrawn could have been at 8–15 km. The calculated crystallization temperature values are 675–775 °C for the amphibole–plagioclase pairs of the felsic clots and 660–800 °C for the host dacite with 710–735 °C temperature range at 95% probability for both cases (Fig. 7). A narrower temperature range (700–760 °C) was obtained by the Holland and Blundy (1994) plagioclase-

amphibole thermometer with the average plagioclase composition of An=33 mol%. This temperature range is supported by the Ti-in-zircon and Zr-in-titanite thermometry results (685–725 °C and 690–713 °C, respectively). The crystallization temperature range for the Haramul Mic magma is smaller than what was given for the Fish Canyon Tuff (706–814 °C; Bachmann and Dungan 2002), although there is a good agreement with the mean temperature of 715 °C given for low-Al amphibole population. Although the dimensions of these magma reservoirs are very different, yet very similar magma storage conditions characterise both. This represents a near-solidus, near-eutectic highly crystalline silicic magma, close to the ‘viscous death’ state, which appears to be common for dacitic volcanic systems, worldwide. The lack of coexistent Fe–Ti oxides prevents the accurate determination of the oxidation state of the magma, but the lack of a pronounced negative Eu anomaly in the bulk rock and glass rare-earth element patterns implies relatively oxidized condition as shown also for the younger Ciomadul dacitic magmas by Cserép et al. (2023).

The common felsic crystal clots in the Haramul Mic dacite can represent fragments of this crystal mush (Fig. 2c). They contain mostly plagioclase crystals in various sizes and euhedral amphibole and biotite, often as inclusions. The inter-crystal glass is fresh, microlite-free, but contains relatively large, rounded vesicles. The outer crystal margins are mostly intact, only subtle rounding can be observed. As accessories, titanite, apatite and zircon occasionally are found, mostly within the plagioclase crystals. Noteworthy, no compositional difference in plagioclase and amphibole composition is found between the felsic clots and the host dacite, while the groundmass glass in the host dacite is more silicic, presumably due to the microlite content. Thus, the felsic crystal clots are interpreted as intact crystal mush fragments with less than 10 vol% melt fractions. They were swept out by the evacuated magma from the magma reservoir, while most of the crystal phases of the crystal mush were incorporated into the melt-dominated uprising magma body. The abundance of the rounded vesicles in the felsic clots and the sheared vesicles in the glassy groundmass of the dacite (Fig. 2b–d) indicate relatively high water content of the melt and decompression-driven vesiculation en-route to the surface. The ubiquitous occurrence of amphibole and biotite as mafic minerals in the Haramul Mic dacite also suggests relatively high-water content of the magma. This is estimated using the plagioclase-liquid hygrometer-thermometer calibrated by Waters and Lange (2015). For this calculation, the homogeneous inter-crystal glass composition and the plagioclase Ab–An content in the felsic clot were used. For the temperature, what is the most sensitive parameter in this calculation, the average 720 °C was used, while for the pressure, 250 MPa. As a result,

6.5–6.7 wt% H<sub>2</sub>O in the melt was calculated. Considering the crystallinity (ca. 40 vol%) and the dissolved water content of the Haramul Mic dacite, the lava dome extrusion type eruption is consistent with the model given by Popa et al. (2021). Efficient outgassing in the ascending viscous magma could have been a driver factor to deliver such highly crystalline magma to the surface. The high-water content enhanced also the relatively fast ascent rate. The amphibole crystals in the Haramul Mic magma are all intact, they have no reaction rim as observed many times in amphibole-bearing dacites such as in Soufrière Hills, Montserrat (Devine et al. 1998; Rutherford and Devine 2003; Buckley et al. 2006; De Angelis et al. 2015) and also in the younger, 140–95 ka lava dome rocks of Ciomadul (Kiss et al. 2014) that is consistent with the rapid magma ascent.

### **Mineral–melt trace element partition coefficients**

Trace element partition coefficients (McIntire 1963; Gast 1968) are crucial in quantitative petrogenetic modelling to better understand magmatic evolution (e.g., Allégre et al. 1977; Hanson 1978; Bea et al. 1994; Shaw 2006; Deering and Bachmann 2010 and many others). Therefore, there has been a great effort to determine these values at various pressure, temperature, redox, and melt composition conditions. Intermediate to silicic magmatic systems play a key role in crustal evolution and within them, amphibole has a particular importance (Davidson et al. 2007, 2013). Amphibole can crystallize from a wide magmatic condition and its composition is highly sensitive to the crystallization environment. Amphibole–melt distribution coefficients were calculated from experimental studies, where the crystallization conditions were controlled (e.g., Klein et al. 1997; Hilyard et al. 2000; Adam and Green 2006; Tiepolo et al. 2007; Nandedkar et al. 2016), whereas others have used natural systems and provided mineral–melt distribution coefficients from compositional analysis of equilibrium phases (e.g., Nagasawa 1970; Bacon and Druitt 1988; Sisson 1994; Bachmann et al. 2005; Padilla and Gualda 2016). Among them, most of the studies focused on basaltic systems and there are only rare data for dacitic to rhyolitic conditions.

Trace element partition can be interpreted based on the crystal lattice strain model described by Brice (1975), Blundy and Wood (1994, 2003) and Wood and Blundy (1997). This implies crystallographic control on the trace element partition in a way that for isovalent trace elements there is a parabolic relationship between the logarithm of the partition coefficient and the ionic radius illustrated by the Onuma diagrams (Onuma et al. 1968; Fig. 8). The best-fit curve in the Onuma plot allows to test the quality of the calculated partition coefficients. The

lattice strain parameters ( $D_0$ , the maximum mineral–melt distribution coefficient;  $r_0$ , cation radius at the highest  $D_0$  and  $E$ , the Young’s modulus) are obtained from such diagram and can be compared to the results coming from experimental studies. Nandedkar et al. (2016) pointed out that the amphibole–liquid partition coefficients increase significantly with decreasing temperature and increasing magma evolution trends, where the host melt becomes more silicic, while the determined crystal lattice parameters ( $r_0$  and  $E$ ) remain nearly constant. However,  $r_0$  has a negative correlation with the ferromagnesian content of the amphibole (Shimizu et al. 2017). A collection of the published amphibole–liquid partition coefficient data (Tiepolo et al. 2007; Shimizu et al. 2017) shows that most of them were derived from mafic to intermediate melt composition and temperature range from near liquidus stage (around 1000–1100 °C) to 780 °C. In contrast, only limited data are available for more silicic (dacitic to rhyolitic) magmatic systems at low (<750 °C) temperature, close to the cotectic or eutectic minimum point, appropriate for many plutonic formations (Barnes et al. 2016). In such condition, accessory minerals crystallizing along with amphibole can also affect the occupation of trace elements in the amphibole crystal lattice. The Haramul Mic dacite provides an ideal opportunity to determine a wide range of trace element partition coefficients for the coexisting mineral phases and silicic liquid, since thermometry calculations suggest stable low temperature (around  $730 \pm 30$  °C) crystallization of amphibole, plagioclase, biotite, titanite and zircon. The mineral phases have relatively homogeneous chemical compositions and euhedral crystal shape, while the groundmass is glassy (rhyolitic in composition;  $\text{SiO}_2$  is around 75 wt%). The textural and geochemical data imply that they represent a nearly invariant eutectic condition. Although microlites exist in the groundmass glass of the host dacite, clear glass phase is found in the felsic crystal clots. This enabled us to calculate the trace element partition coefficients based on trace element concentration of various mineral phases and the melt phase (represented by the unaltered, pure glass) in equilibrium. Average of in-situ trace element compositional data was used, determined by LA-ICP-MS analysis both for the mineral and glass phases (Table 1).

Mineral–melt trace element partition coefficients were calculated for all detectable elements by LA-ICP-MS in five mineral phases (plagioclase, amphibole, biotite, titanite and zircon; Table 2; Fig. 9) of the Haramul Mic dacite that represent a natural database for low-T, near-eutectic silicic magmatic system. Comparably extensive  $K_d$  database for similar systems is still quite rare in literature except for a few publications such as Bachmann et al. (2005), Padilla and Gualda (2016). We were able to determine mineral–melt partition coefficient for a range of trace elements in case of amphibole, titanite and zircon, however, a limited dataset was

obtained for biotite and plagioclase due to the low concentration of many elements and the abundance of mineral and melt inclusions in biotite (Fig. 9). In order to avoid errors resulted from contamination, the biotite data was carefully filtered before  $K_d$  calculations, but interpretation and use of the reported biotite partition coefficient values require caution. In plagioclase, most trace elements are incompatible, especially REE, thus their concentrations are near the LA-ICP-MS detection limits, which cause higher uncertainty. Taking all these into account, in the following only amphibole, titanite and zircon trace element partitioning behaviour is discussed in detail.

### *Amphibole*

Amphibole/melt REE and Y partition coefficient values are higher for the Haramul Mic dacite compared to literature data compiled by Tiepolo et al. (2007) and Shimizu et al. (2017), which are mainly related to mafic to intermediate magmatic systems. They are a little bit higher than those determined by Nandedkar et al. (2016) experimentally for the more silicic liquid ( $\text{SiO}_2=65.9$  wt%) and the lowest temperature (780 °C). This is consistent with the rhyolitic composition ( $\text{SiO}_2=75$  wt%) and the lower crystallization temperature (around 730 °C) for the Haramul Mic. This condition as well as the amphibole compositions are more similar to that of the Fish Canyon Tuff (Bachman et al. 2005), the Deer Mountain (Sisson 1994) and the Peach Spring Tuff high-silica rhyolite (Padilla and Gualda 2016). In these cases, the amphibole-liquid trace element distribution values are in the same range within uncertainty, the best similarity is found with the Peach Spring Tuff (Fig. 10a). Melt composition significantly affects the REE partitioning behaviour (positive correlation between melt  $\text{SiO}_2$  and  $K_d$  values), as well as magma temperature, that generally correlates negatively with the  $K_d$  values (Wood and Blundy 2002; Blundy and Wood 2003; Tiepolo et al. 2007; Nandedkar et al. 2016).

We got a statistically acceptable parabolic fit [Nash max=0.983079 and min=0.973591 ( $1\sigma$ ), while RMSE max=7.1990 and min=5.8538 ( $1\sigma$ )] for the obtained trivalent trace element partition coefficients of the Haramul Mic dacite in the Onuma plot using the Simple Fit method by Dalou et al. (2018) and this confirms the reliability of our result (Fig. 8a). The highest  $K_d$  values (close to 10) for amphibole were obtained for Gd, Tb and Dy. Cerium also fits the curve implying that it exists mostly as  $\text{Ce}^{3+}$ . The only deviation is shown by Y and Gd, the lower values are similar as obtained by Padilla and Gualda (2016) for the Peach Spring Tuff rhyolite and this can also be observed in the experimentally determined data set (Shimizu et al. 2017). The reason for that is not known and needs further studies to explain such behaviour. Another anomalous partition coefficient value is shown by Sc ( $4.62 \pm 0.16$ ), departing significantly from

the trivalent parabola, although with a lesser extent compared to that published for the Fish Canyon Tuff (Bachmann et al. 2005) and the Peach Spring Tuff (Padilla and Gualda 2016) as well as to that experimentally determined by Nandedkar et al. (2016) for the more evolved liquid composition (RN15V2). This is due to the higher Sc concentration of the Haramul Mic glass (13–16 ppm) and the lower Sc content in the amphibole (57–80 ppm). Partition coefficients for the high-field strength elements (HFSE) as well as U and Th show similarity with the Fish Canyon and Peach Spring Tuff values but are systematically lower compared to the experimentally obtained values of Nandedkar et al. (2016). This can be due to the coexistence and co-crystallisation with zircon and titanite, which have higher  $K_d$  values for these elements.

### *Zircon*

The calculated zircon/melt trace element partitioning coefficients are generally similar, although a little bit higher than the published data for various rhyolitic and dacitic systems (Sano et al. 2002; Bachmann et al. 2005; Colombini et al. 2011; Padilla and Gualda 2016; Claiborne et al. 2018) (Fig. 10b). The reliability of the  $K_d$  values is proved by the excellent fit (Nash max=0.996511 and min=0.993613 ( $1\sigma$ ), while RMSE max=7.1326 and min=5.2782 ( $1\sigma$ ); Dalou et al. 2018) in the trivalent cation parabolic curve on the Onuma diagram (Fig. 8b). The middle and heavy REE, Y, Hf, U and Th are strongly compatible ( $K_d > 10$ ), Nb is slightly incompatible. Cerium shows a positive shift from the curve, whereas Eu is just a little bit beneath it, which is consistent with published data. Both Eu and Ce anomalies (Fig. 10b) reflect the redox conditions of the magmatic system during crystal growth.  $Ce^{4+}$  and  $Eu^{2+}$  are more compatible than their trivalent versions, thus the anomalies in these trace element partition values refer to the oxidative conditions (e.g., Colombini et al. 2011; Trail et al. 2012; Clairborne et al. 2018).

### *Titanite*

The titanite/melt partition coefficients are higher than those determined experimentally (e.g., Tiepolo et al. 2002; Prowatke and Klemme 2005) and akin to the values calculated for silicic volcanic rocks (Bachmann et al. 2005; Colombini et al. 2011; Padilla and Gualda 2016). The  $K_d$  values are generally high ( $>100$ ), particularly for the middle REE and Y (Fig. 10c). In the Onuma diagram, a good fit (Nash max=0.971056 and min=0.966498 ( $1\sigma$ ), while RMSE max=3.8763 and min=3.5775 ( $1\sigma$ ); Dalou et al. 2018) was obtained for the trivalent parabola, although the values for Y and Gd are a little bit lower (Fig. 8c), similarly to amphibole (Fig. 8a). Noteworthy, the  $K_d$  value for Sc is less than 1, i.e. behaves slightly incompatible and this

value is significantly lower than published by Bachmann et al. (2005), Colombini et al. (2011) and Padilla and Gualda (2016).

#### *Crystal lattice parameters*

The obtained partition coefficients can be interpreted in terms of the crystal lattice model (Brice 1975; Blundy and Wood 1994, 2003). The lattice parameters for the trivalent cations were determined by the best-fit parabola by the fitting method of Dalou et al. (2018) in the Onuma diagram and presented in Table 3. The  $D_0$  is inversely correlated with the crystallization temperature (Klein et al. 1997; Tiepolo et al. 2007; Nandedkar et al. 2016) and our data are consistent with it. The amphibole  $D_0$  value ( $9.2 \pm 0.18$ ) is comparable to what Nandedkar et al. (2016) obtained for the most evolved, low temperature magmatic system, and we got high values also for titanite and zircon, similarly as Padilla and Gualda (2016). In contrast, the Young's modulus ( $E$ ) does not change with temperature, our data for amphibole ( $363 \pm 10$ ) fits well with those determined by Nandedkar et al. (2016). The lattice parameters for titanite and zircon also show a good fit with the values published by Sano et al. (2002), Bachmann et al. (2005), Colombini et al. (2011) and Padilla and Gualda (2016). Thus, the mineral/melt partition coefficients calculated for the Haramul Mic dacite can be used for silicic volcanic and plutonic systems characterised by low temperature ( $<750$  °C), near-eutectic magmatic conditions. These data broaden the available database about  $K_d$  values to the low-T, low-p, wet, oxidative, silicic magmatic system and can serve as important key input parameters for petrogenetic model calculations. Modern in-situ trace element concentration analysis with LA-ICP-MS can avoid inclusions and provide data only from crystal rims, hence measurement data is more reliable compared to bulk analysis techniques (Petrelli et al. 2007). Determining partition coefficients from natural samples with precise and accurate analytical methods and calculations can be even more realistic than experiments, especially in the case of the presence of accessory minerals like zircon and titanite (Padilla and Gualda 2016).

#### **Eruption initiation**

The Ciomadul is a typical long-dormant volcano as shown by the detailed zircon geochronology (Harangi et al. 2015a, 2020; Molnár et al. 2018, 2019). During the Old Ciomadul eruptive period (1000–300 ka), the active phases were separated even more than 100 kyr quiescence, whereas during the Young Ciomadul eruptive period, several 10 thousand years repose time was recognised. Thus, although the last eruption took place at 30 ka (Vinkler et al.

2007; Harangi et al. 2010; Harangi et al. 2015a, 2020; Karátson et al. 2019), eruption rejuvenation cannot be unambiguously excluded. Zircon U-Th-Pb dating in combination with thermal modelling suggested that a long-living crystal mush magma reservoir has been stored in the crust for more than 1 Myr (Laumonier et al. 2019; Lukács et al. 2021) and geophysical studies (Popa et al. 2012; Harangi et al. 2015b; Comeau et al. 2024) revealed that such highly crystalline magma may still exist in a depth range of 8–20 km. Constraints on eruption initiation after long dormancy are, therefore, important to better understand the behaviour of this volcano as well as similar volcanic systems, worldwide. Due to the long quiescence period, monitoring data are not available, only the volcanic formations of the past eruptions can tell us the processes, which occurred in the magma reservoir.

Kent et al. (2023) reviewed the possible eruption initiation mechanisms for various volcanic systems using large number of published cases and concluded that intrusion of mafic magma into a shallow level mushy magma reservoir (mafic recharge or mafic rejuvenation) is far the most common processes to reactivate a portion of the physically un-eruptible crystal-rich magma body and form eruptible magma batch. Mafic recharge played an important role in the volcanism of Ciomadul as shown for the dominantly lava dome extrusion epoch by Kiss et al. (2014) and the youngest eruptive epoch by Cserép et al. (2023). Nevertheless, some differences in the composition of the recharge magma can be recognised. During the 135 ka Ciomadul Mic lava dome eruption, arrival of relatively primitive hot mafic magma into the felsic crystal mush caused a strong reheating ( $>200$  °C; Kiss et al. 2014), whereas in the mostly explosive eruptive epoch, the recharge magmas were more hydrous and delivered amphibole crystals with slightly different composition (Cserép et al. 2023).

The Haramul Mic lava dome extrusion took place after more than 100 kyr repose time as shown by the zircon (U–Th)/He ages and represents the onset of the Young Ciomadul eruptive period (Lukács et al. 2021). The high crystallinity of the erupted dacitic magma and the relative chemical homogeneity of the main mineral phases are unique among the Ciomadul rocks. The calculated pre-eruptive magma conditions point to a low-temperature (ca. 730 °C), near eutectic stage. Mafic magma recharge can be excluded as eruption initiation since no sign of such event can be detected from core to rim compositional variation in amphibole and plagioclase. On the other hand, the melt contained elevated water, 6.5–6.7 wt% calculated using the plagioclase-liquid hygrometer of Waters and Lange (2015). Remarkably, Li drops at the outermost margin of the plagioclase and often in amphibole macrocrysts and this indicates the appearance of gas phase in the melt (Berlo et al. 2004; Kent et al. 2007). The inter-crystal glass

in the felsic crystal clots contains large, spherical vesicles (Fig. 2c), whereas sheared vesicles are frequent in the groundmass glass (Fig. 2b,d), which all suggest vesiculation in the magma. Magma ascent was relatively fast since no reaction corona was formed around amphibole macrocrysts, that is typical of decompression driven decomposition. Nevertheless, this can also be due to the still relatively high-water content ( $>4$  wt%) of the melt, where amphibole remained stable. Thus, we can envisage that evacuation of a portion of the crystal mush could be attributed to volatile accumulation, oversaturation and exsolution process. This can be consistent with the ‘second boiling’ process during progressive crystallization (Sisson and Bacon 1999) although we cannot exclude mafic magma underplating beneath the crystal mush resulting in intense degassing and as a result leading to volatile oversaturation in the crystal mush (Andersen et al. 2018). In addition to water,  $\text{CO}_2$  flush can also cause rejuvenation (Caricchi et al. 2018). Carbon dioxide emission with significant magmatic component from a deep mafic magma reservoir is going on at Ciomadul (Kis et al. 2019) that can support this scenario. The increased volatile content could decrease the melting point and cause also partial melting in the crystal mush, mostly involving biotite, quartz, and possibly K-feldspar, which increased the melt fraction and formed eruptible magma fraction. Moreover, during magma ascent and decompression, gas phase separates reducing the magma density and elevates the buoyancy. The glass, particularly the inter-crystal phase in the felsic crystal clots, contains high Ba ( $>1000$  ppm), high Li ( $>50$  ppm) and high Sc ( $>13$  ppm) which may have preserved this condition. Details of the crystal mush rejuvenation process from the perspective of the volatiles need further research, for example through the investigation of Li-diffusion in phenocrysts and fluid inclusion studies (e.g., Neukampf et al. 2022).

Volatiles played an important role also in the rejuvenation of the large silicic mushy magma reservoir during the Fish Canyon Tuff eruption (Bachmann et al. 2005). There are striking petrological similarities between the Haramul Mic and Fish Canyon Tuff dacites, and it seems that also in the eruption initiation, even though there is a large difference in the volume of the magmas. Noteworthy, no other Ciomadul volcanic rock shows this feature, therefore, the Haramul Mic eruption was unique. However, as the magma genetic reconstructions of the Ciomadul volcanism (Kiss et al. 2014; Cserép et al. 2023 and this study) show, various processes can play a role in eruption initiation even in a single volcano. The most common cause of rejuvenation is magma recharge as Kent et al. (2023) pointed out, but this can involve distinct types of magmas and other mechanisms, such as volatile accumulation, can also lead to eruption.

## Conclusions

- (1) Haramul Mic is a crystal-rich (~35–40 vol%) dacitic lava dome that represents the first eruption product of the Ciomadul volcanic complex, a typical long-dormant, PAMS volcano, after more than 100 kyr repose time. The petrographic features and the geochemical characteristics of the bulk rocks as well as the glassy groundmass and the mineral phases (amphibole, plagioclase, biotite, titanite, zircon) along with geothermobarometry and hygrometry calculations suggest a near-eutectic, near-solidus, low-temperature (730 °C), low-pressure (200–400 MPa), wet and oxidized silicic magmatic system.
- (2) Determination of trace element partition coefficients are crucial in quantitative petrogenetic modelling to better understand magmatic evolution. The Haramul Mic dacite represents a near-equilibrium crystal mush with various mineral phases and fresh interstitial glass showing homogeneous major and trace element composition. Therefore, it is a suitable natural sample for determining mineral–melt trace element partition coefficients. In this paper, a wide set of amphibole–melt trace element partition coefficient data is provided, which can be used for silicic volcanic and plutonic systems characterised by low temperature (<750 °C), near-eutectic magmatic conditions. Such an extensive  $K_d$  database for similar systems is still quite rare in the literature. Additionally, mineral–melt trace element partition coefficients are also calculated for titanite, zircon, plagioclase and biotite phases. Quality-check of the obtained data was performed by Onuma-plots. The crystal lattice parameters were determined which fit well with published results.
- (3) The high crystallinity of the Haramul Mic dacite and the relative chemical homogeneity of the main mineral phases are unique among the Ciomadul rocks, but such erupted crystal mush material is also rare worldwide. There is no sign for mafic magma recharge as eruption initiation, which characterized the subsequent eruptions. Instead, volatiles could have played a role ('second boiling' or volatile oversaturation due to degassing from underplated mafic magma). The lack of reaction corona around amphibole macrocrysts suggests relatively fast magma ascent, although this can also be due to the still relatively high water content (>4 wt%) of the melt, where amphibole remained stable. However, details of the crystal mush rejuvenation process from the perspective of the volatiles need further research.

**Supplementary Information** The online version contains supplementary material.

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## Statements and Declarations

**Data availability** All data required are made available.

**Conflict of interest** The authors have no relevant financial or non-financial interests to disclose.

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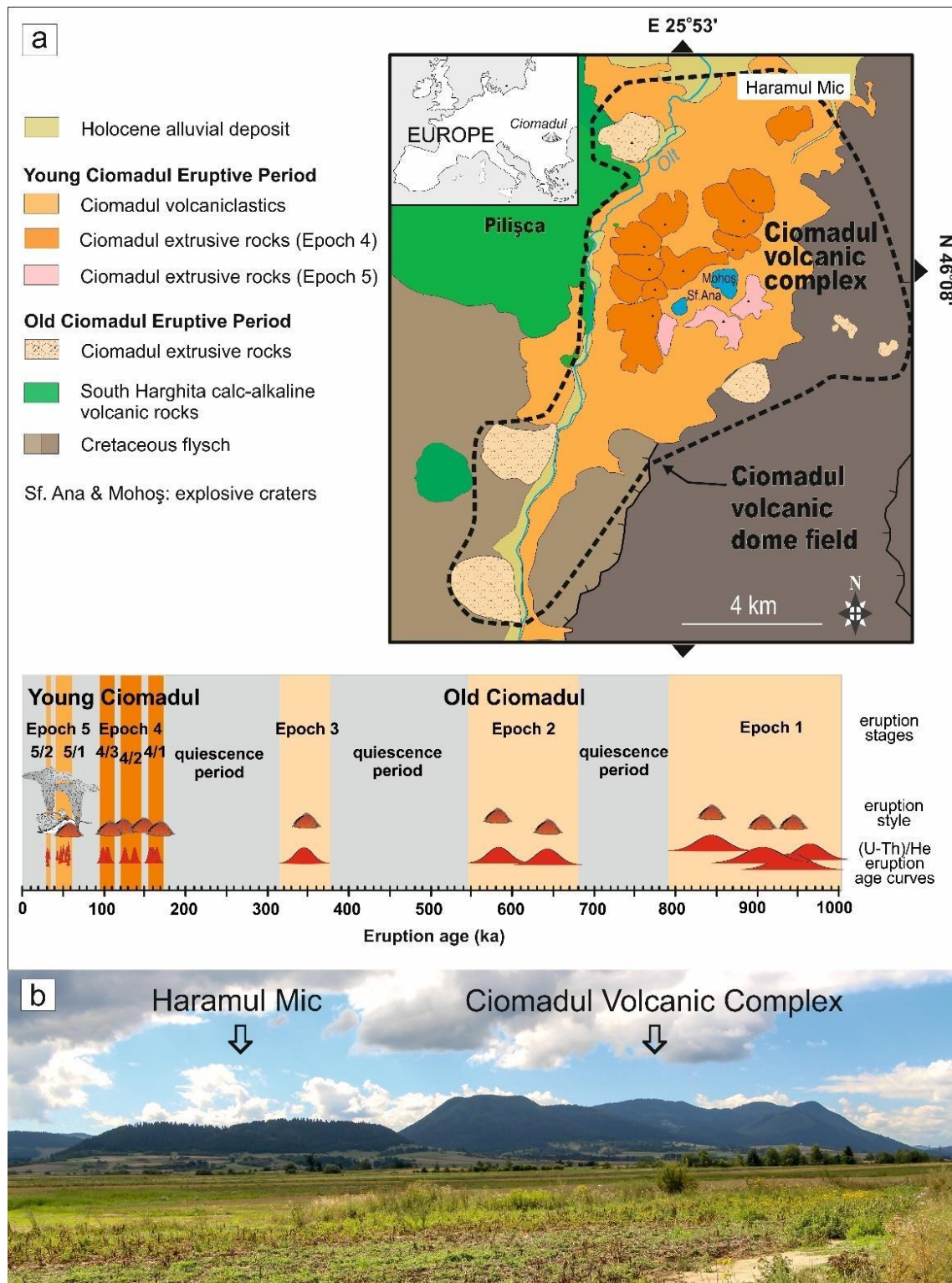
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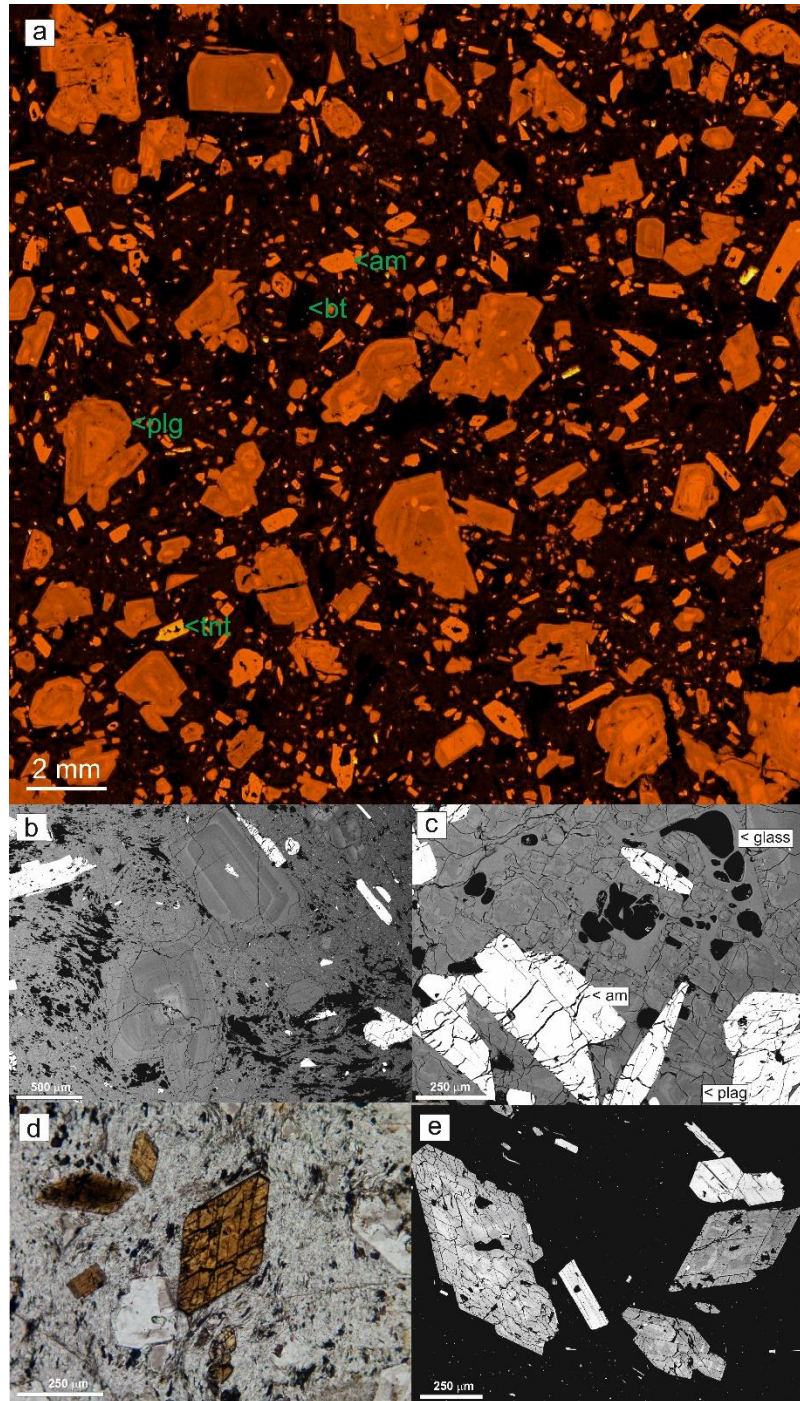
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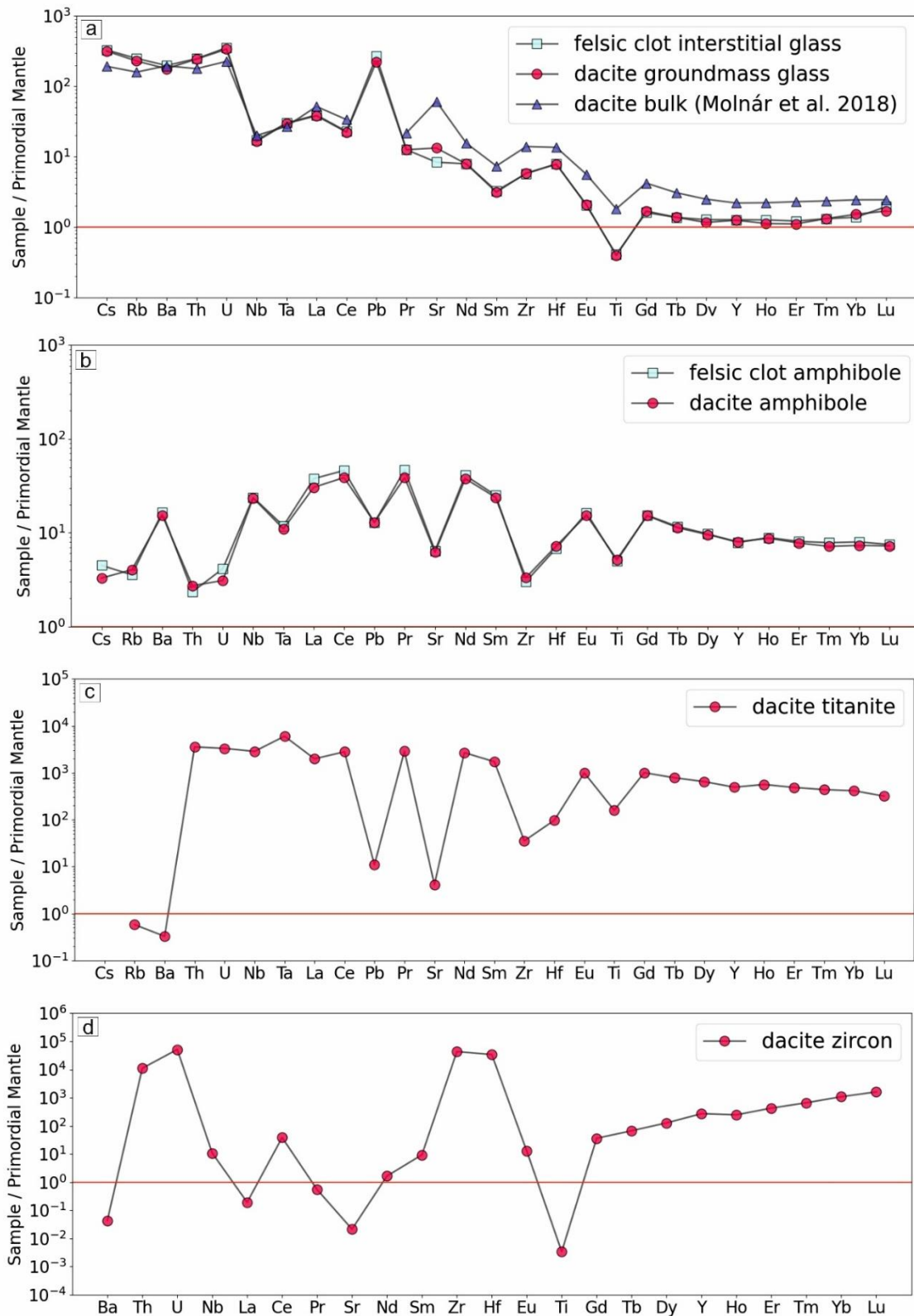
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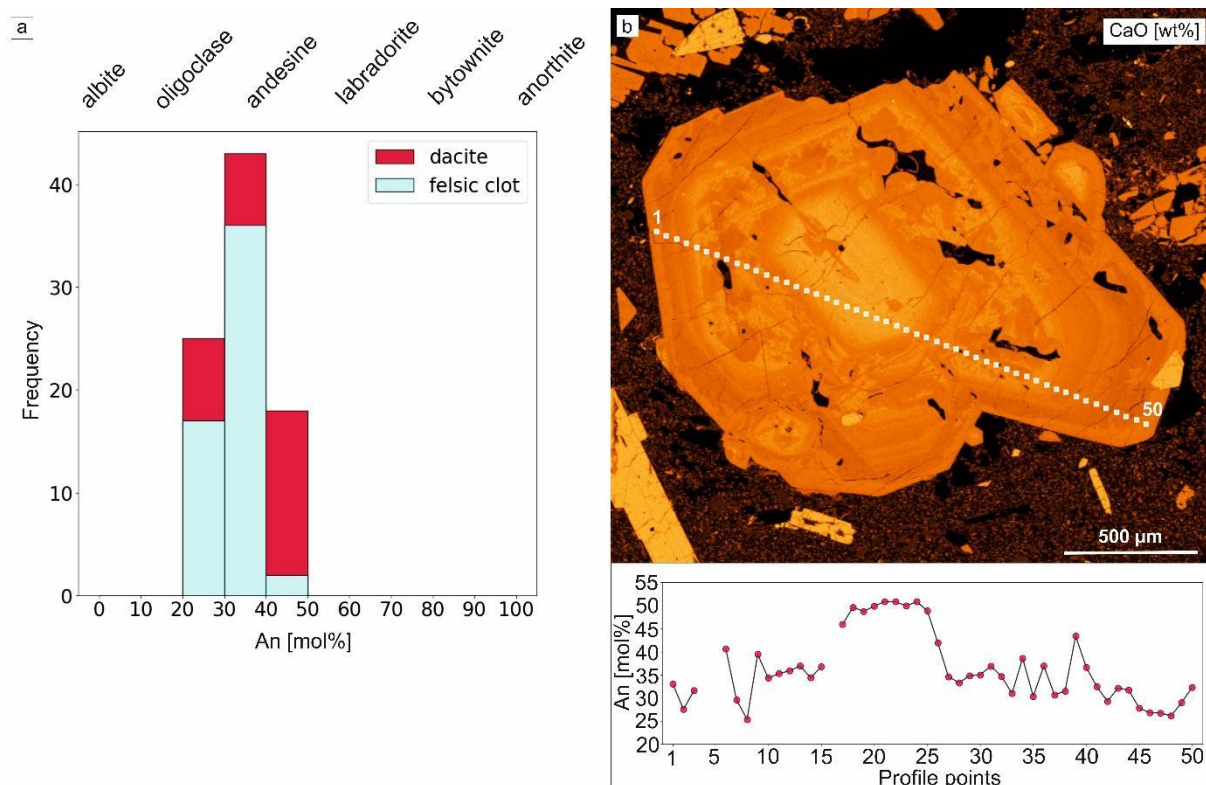
**Fig. 1 a** Simplified geological map (based on Seghedi et al. 1987 and Cserép et al. 2023) and eruptive history (Molnár et al., 2019) of the Ciomadul volcano, showing the location of Haramul Mic lava dome. **b** View of the Haramul Mic flat lava dome and the Ciomadul Volcanic Complex from north.



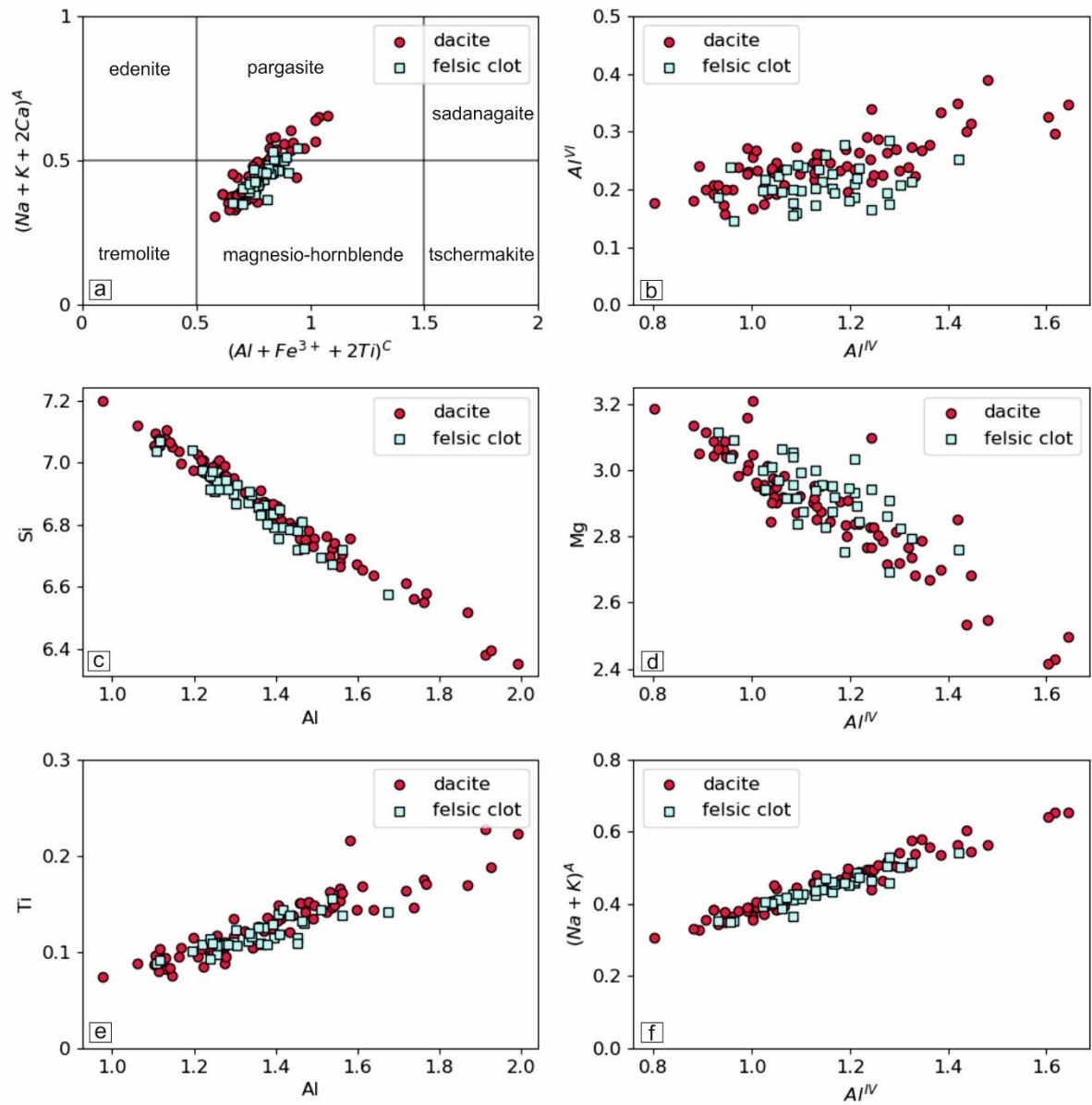
**Fig. 2** **a** High-resolution CaO element-oxide map of the Haramul Mic lava dome dacite: biotite (bt) crystals are black, zoned plagioclase (plg) macrocrysts are dark orange, amphibole (am) macro- and mesocrysts are light orange and accessory titanite (tnt) is yellow. The lighter colour shades indicate relatively higher concentration values. Back-scattered electron (BSE) and microscopic images of Haramul Mic dacite: oscillatory zoned plagioclase macrocrysts and amphibole and biotite meso- and microcrysts in the host dacite (**b**), felsic crystal clots of plagioclase and amphibole with intercrystal vesicular glass (**c**). Euhedral amphibole macro- and mesocrysts and biotite mesocrysts in the dacite (**d**, **e**). Note that the glassy groundmass of the host dacite contains abundant sheared vesicles (**b**, **d**), whereas spherical vesicles are in the intercrystal glass of the felsic clots (**c**).



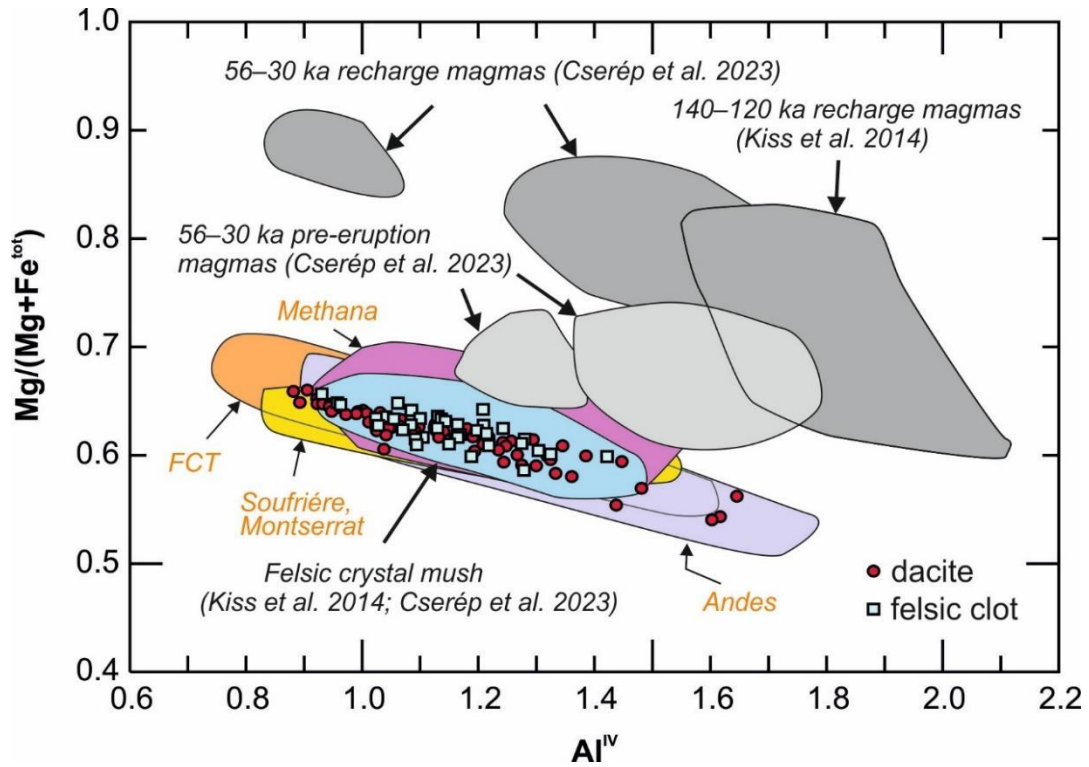
**Fig. 3** Primitive mantle (McDonough and Sun 1995) normalised trace element patterns (average values; see Supplementary Material ESM1) of the (a) interstitial glass of the host rock groundmass and the felsic clot compared with the host rock bulk composition (Molnár et al., 2018; Supplementary Material ESM 2); (b) amphibole macrocrysts; (c) titanite and (d) zircon.



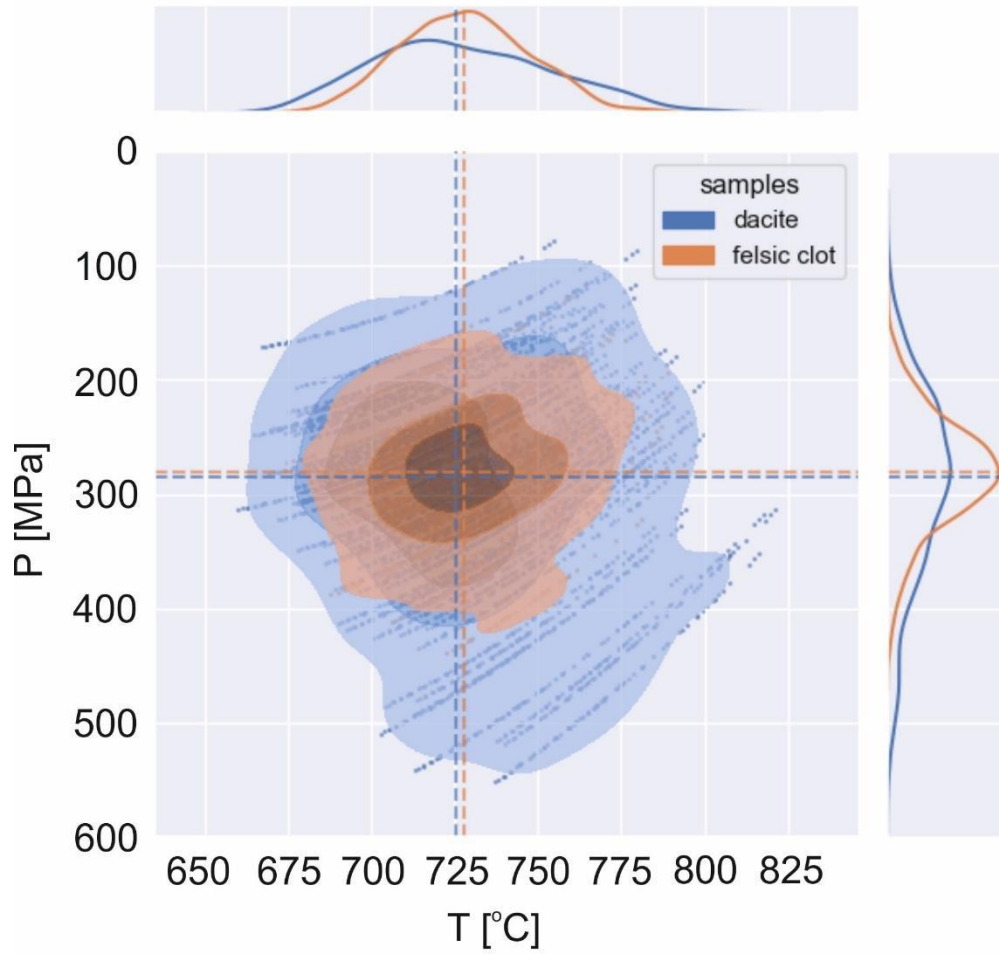
**Fig. 4 a** Frequency distribution of anorthite (An in mol%) content in the plagioclase macrocrysts of the Haramul Mic dacite and the felsic clots. **b** Variation of An content within a representative plagioclase macrocryst (traverse is shown in the CaO element-oxide map of the studied crystal).



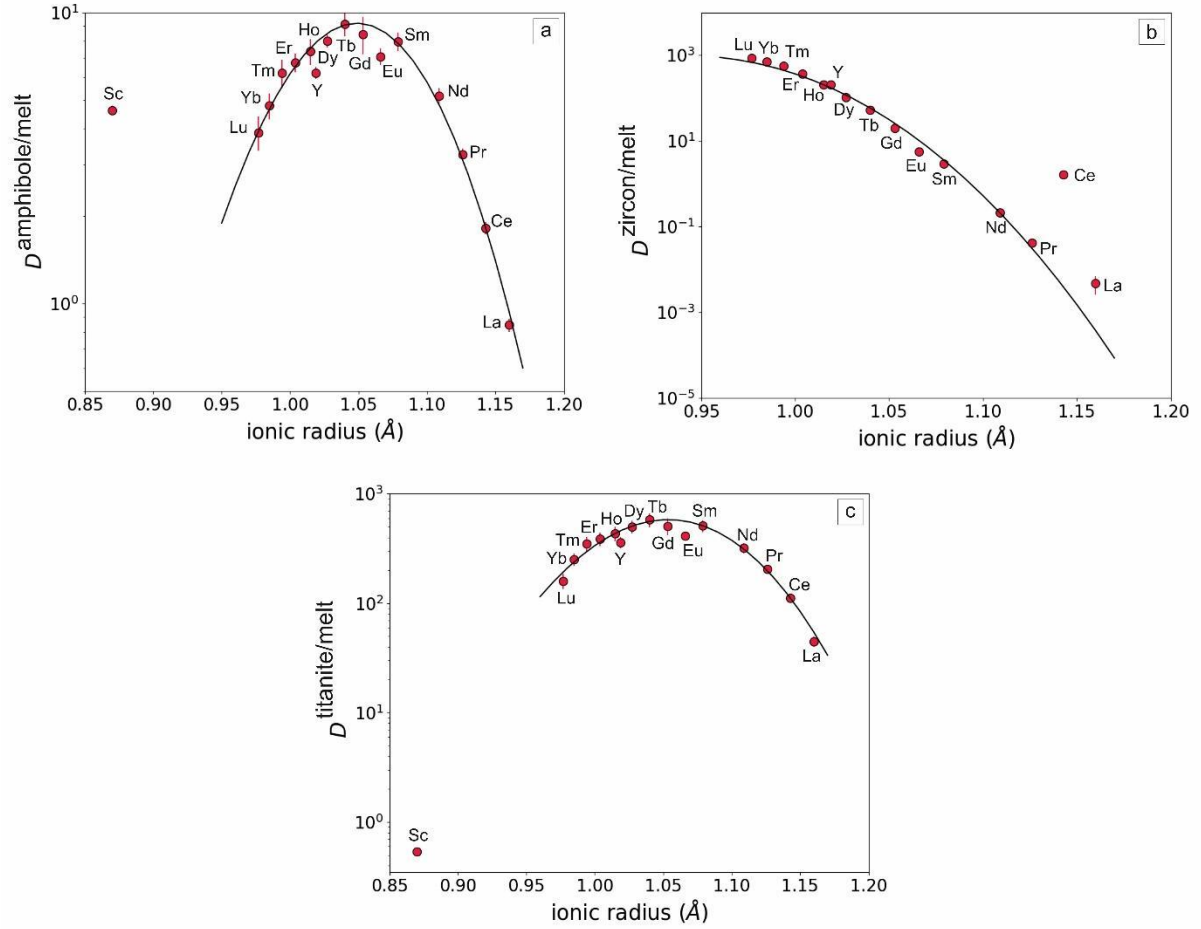
**Fig. 5** a Classification (based on Hawthorne et al. 2012) and (b–f) compositional variation of amphibole crystals. Correlation between various cation numbers illustrates the dominant, temperature-controlled substitution mechanisms (edenite and Ti-tschermakite) in the Haramul Mic amphiboles.



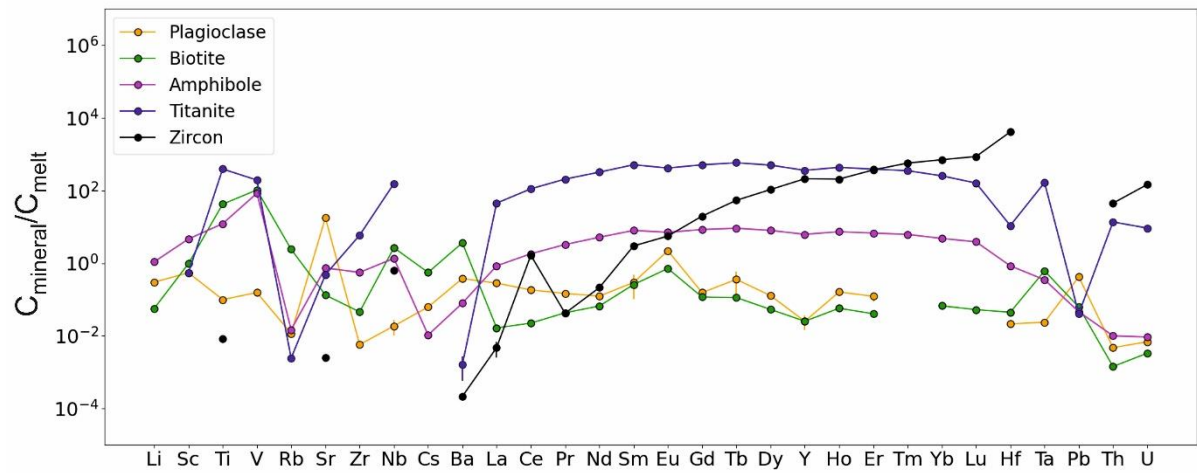
**Fig. 6** Tetrahedral Al ( $Al^{IV}$ ) vs. Mg-value ( $Mg^{2+}/Mg^{2+}+Fe^{tot}$ ) compositions of Haramul Mic amphiboles comparing to the literature amphibole data from the Ciomadul Volcanic Complex (Kiss et al. 2014; Cserép et al. 2023) and similar low-T, silicic volcanic systems (Murphy et al. 2000; Bachmann and Dungan 2002; Rutherford and Devine 2003; Buckley et al. 2006; Humphreys et al. 2009; Popa et al. 2020; Rout et al. 2021; Prat et al. 2023). Note the similarity of the amphibole macrocrysts from the dacite and the felsic clot amphibole. All Haramul Mic amphibole data is identical with the low-Al amphibole group representing a felsic crystal mush in the Ciomadul subvolcanic magma reservoir system (Kiss et al. 2014; Cserép et al. 2023) and overlaps the amphibole composition of Fish Canyon Tuff (FCT), Methana, Soufrière Hills and dacites from the Andes.



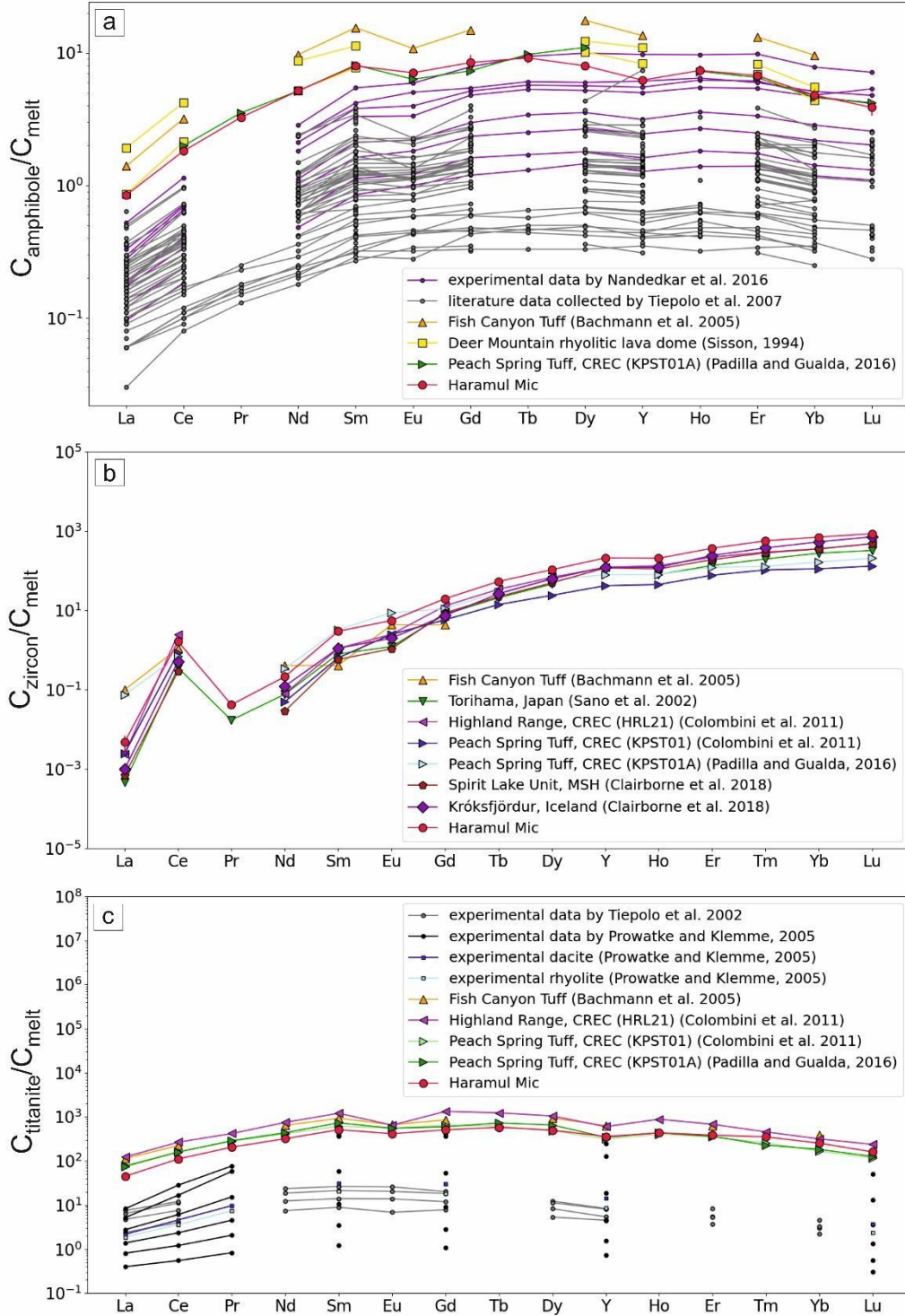
**Fig. 7** Kernel density estimate plot of the amphibole–plagioclase thermobarometry results for Haramul Mic host dacite and felsic clots. The pressure and temperature values were calculated using Thermobar by Wieser et al. (2022). The darkest coloured areas stand for the 95% probability of data occurrence, the mid-coloured areas stand for the 75% probability of data occurrence, and the brighter coloured areas stand for the 25% probability of data occurrence. Median values are shown with dashed lines.



**Fig. 8** Onuma plots (Onuma et al. 1968) showing REE partitioning between amphibole and melt (a) zircon and melt (b) and titanite and melt (c) phases of Haramul Mic dacite. The best-fit curves (Dalou et al. 2018) represent the ideal partitioning of trivalent trace elements in case of equilibrium between melt and mineral phase. Note that Sc does not fit the curves. Moderate Eu-anomaly (a–c) and for  $D^{\text{zircon/melt}}$  also Ce-anomaly (b) can be observed.



**Fig. 9** Calculated mineral–melt partitioning coefficients of all detectable trace elements between five mineral phases (plagioclase, amphibole, biotite, titanite and zircon) and melt.



**Fig. 10 a** Calculated amphibole–melt trace element (REE) partition coefficients for Haramul Mic dacite compared to literature data of experimental (Tiepolo et al. 2007; Nandedkar et al. 2016) and natural (Sisson et al. 1994; Bachmann et al. 2005; Padilla and Gualda 2016) systems. **b** Calculated zircon–melt trace element (REE) partition coefficients for Haramul Mic dacite compared to literature data from similar low-T, silicic volcanic systems (Sano et al. 2002; Bachmann et al. 2005; Colombini et al. 2011; Clairborne et al. 2018; Padilla and Gualda 2016). **c** Calculated titanite–melt trace element (REE) partition coefficients for Haramul Mic dacite compared to literature data of experimental (Tiepolo et al. 2002; Prowatke and Klemme 2005) and natural (Bachmann et al. 2005; Colombini et al. 2011; Padilla and Gualda 2016) systems.

| Mineral and glass trace element mean values |                     |           |           |       |           |           |           |       |             |           |           |       |
|---------------------------------------------|---------------------|-----------|-----------|-------|-----------|-----------|-----------|-------|-------------|-----------|-----------|-------|
|                                             | glass (felsic clot) |           |           |       | amphibole |           |           |       | plagioclase |           |           |       |
|                                             | mean                | min value | max value | SEE   | mean      | min value | max value | SEE   | mean        | min value | max value | SEE   |
| Li                                          | 60.3                | 55.2      | 67.9      | 1.846 | 65.7      | 14.2      | 112.0     | 4.337 | 18.1        | 7.7       | 27.2      | 0.297 |
| Sc                                          | 14.6                | 13.0      | 15.8      | 0.451 | 67.6      | 57.1      | 82.8      | 1.039 | 8.0         | 4.8       | 10.8      | 0.085 |
| V                                           | 3.59                | 2.5       | 4.85      | 0.354 | 301       | 222       | 531       | 7.749 |             |           |           |       |
| Rb                                          | 151                 | 142       | 160       | 2.404 | 2.23      | 1.35      | 4.74      | 0.107 | 1.76        | 0.40      | 6.70      | 0.421 |
| Sr                                          | 165                 | 127       | 189       | 8.597 | 122       | 69        | 204       | 5.528 | 2950        | 2040      | 4420      | 32    |
| Y                                           | 5.53                | 4.58      | 5.77      | 0.163 | 34.4      | 25.4      | 59.2      | 1.299 | 0.14        | 0.03      | 0.54      | 0.061 |
| Zr                                          | 60                  | 58        | 62        | 0.737 | 33.7      | 18.2      | 62.4      | 1.531 | 0.35        | 0.03      | 0.71      | 0.064 |
| Nb                                          | 11.3                | 10.6      | 12.1      | 0.184 | 15.4      | 10.8      | 20.1      | 0.348 | 0.21        | 0.11      | 0.31      | 0.098 |
| Ba                                          | 1304                | 1236      | 1420      | 24.3  | 104       | 36        | 178       | 5.669 | 493         | 248       | 975       | 8.280 |
| La                                          | 26                  | 24        | 27        | 0.443 | 22.0      | 13.5      | 34.5      | 1.102 | 7.37        | 1.72      | 15.90     | 0.210 |
| Ce                                          | 39                  | 37        | 41        | 0.567 | 70.8      | 46.2      | 113.4     | 3.117 | 7.17        | 1.77      | 16.10     | 0.212 |
| Pr                                          | 3.29                | 3.02      | 3.69      | 0.083 | 10.7      | 7.1       | 17.1      | 0.448 | 0.484       | 0.172     | 1.200     | 0.015 |
| Nd                                          | 9.66                | 8.10      | 11.00     | 0.437 | 50.0      | 32.9      | 86.5      | 2.212 | 1.201       | 0.240     | 4.200     | 0.045 |
| Sm                                          | 1.264               | 1.050     | 1.610     | 0.074 | 10.1      | 6.6       | 17.0      | 0.450 | 0.373       | 0.120     | 1.100     | 0.242 |
| Eu                                          | 0.350               | 0.270     | 0.420     | 0.019 | 2.48      | 1.64      | 4.11      | 0.091 | 0.772       | 0.360     | 1.970     | 0.017 |
| Gd                                          | 0.994               | 0.500     | 1.580     | 0.140 | 8.40      | 5.27      | 15.26     | 0.377 | 0.154       | 0.051     | 0.440     | 0.022 |
| Tb                                          | 0.123               | 0.080     | 0.163     | 0.010 | 1.13      | 0.71      | 2.05      | 0.051 | 0.044       | 0.015     | 0.100     | 0.028 |
| Dy                                          | 0.806               | 0.720     | 0.850     | 0.019 | 6.44      | 4.30      | 11.35     | 0.285 | 0.103       | 0.075     | 0.140     | 0.012 |
| Ho                                          | 0.178               | 0.129     | 0.250     | 0.016 | 1.31      | 0.88      | 2.35      | 0.058 | 0.029       | 0.016     | 0.048     | 0.003 |
| Er                                          | 0.509               | 0.360     | 0.630     | 0.034 | 3.43      | 2.44      | 5.43      | 0.123 | 0.063       | 0.044     | 0.081     | 0.019 |
| Tm                                          | 0.079               | 0.059     | 0.100     | 0.009 | 0.49      | 0.35      | 0.77      | 0.017 |             |           |           |       |
| Yb                                          | 0.679               | 0.490     | 0.960     | 0.066 | 3.25      | 2.13      | 5.06      | 0.108 |             |           |           |       |
| Lu                                          | 0.126               | 0.064     | 0.197     | 0.017 | 0.49      | 0.36      | 0.73      | 0.014 |             |           |           |       |
| Hf                                          | 2.361               | 2.000     | 2.860     | 0.127 | 2.00      | 1.16      | 3.62      | 0.074 | 0.051       | 0.048     | 0.053     | 0.003 |
| Ta                                          | 1.149               | 1.020     | 1.380     | 0.051 | 0.40      | 0.22      | 0.73      | 0.017 | 0.027       | 0.019     | 0.041     | 0.004 |
| Pb                                          | 39.5                | 28.6      | 52.7      | 2.937 | 1.83      | 1.06      | 2.71      | 0.054 | 16.6        | 11.3      | 20.7      | 0.113 |
| Th                                          | 19.7                | 18.4      | 20.8      | 0.338 | 0.20      | 0.09      | 0.70      | 0.022 | 0.094       | 0.026     | 0.210     | 0.016 |
| U                                           | 7.19                | 6.83      | 7.62      | 0.103 | 0.07      | 0.01      | 0.23      | 0.009 | 0.050       | 0.024     | 0.094     | 0.006 |
|                                             | biotite             |           |           |       | titanite  |           |           |       | zircon      |           |           |       |
|                                             | mean                | min value | max value | SEE   | mean      | min value | max value | SEE   | mean        | min value | max value | SEE   |
| Li                                          | 3.42                | 1.23      | 11.99     | 0.412 |           |           |           |       |             |           |           |       |
| Sc                                          | 14.20               | 13.17     | 15.04     | 0.102 | 7.79      | 6.48      | 9.7       | 0.466 |             |           |           |       |
| V                                           | 372                 | 363       | 387       | 1.326 | 696       | 613       | 783       | 28.5  |             |           |           |       |
| Rb                                          | 363                 | 330       | 378       | 2.172 | 0.37      | 0.35      | 0.39      | 0.012 |             |           |           |       |
| Sr                                          | 22                  | 14        | 35        | 0.802 | 80        | 53        | 98        | 6.523 | 0.4219      | 0.27      | 0.96      | 0.043 |
| Y                                           | 0.14                | 0.05      | 0.33      | 0.014 | 1980      | 1354      | 2590      | 227   | 1163        | 402       | 1920      | 97    |
| Zr                                          | 2.77                | 2.48      | 3.02      | 0.031 | 356       | 274       | 456       | 24    |             |           |           |       |
| Nb                                          | 30                  | 29        | 34        | 0.219 | 1741      | 1432      | 2061      | 109   | 7.11        | 2.36      | 13.90     | 0.833 |
| Ba                                          | 4752                | 3117      | 6760      | 183   | 2.15      | 0.48      | 4.90      | 1.384 | 0.28        | 0.10      | 0.81      | 0.041 |
| La                                          | 0.432               | 0.070     | 1.840     | 0.090 | 1165      | 607       | 1470      | 124.1 | 0.12        | 0.01      | 0.83      | 0.058 |
| Ce                                          | 0.876               | 0.014     | 3.720     | 0.191 | 4330      | 2640      | 5230      | 377.1 | 64          | 15        | 157       | 8.90  |
| Pr                                          | 0.142               | 0.024     | 0.510     | 0.027 | 677       | 443       | 881       | 59.4  | 0.14        | 0.03      | 0.30      | 0.018 |
| Nd                                          | 0.643               | 0.064     | 2.370     | 0.126 | 3090      | 2110      | 4280      | 305.8 | 2.06        | 0.35      | 5.03      | 0.279 |
| Sm                                          | 0.327               | 0.240     | 0.590     | 0.054 | 645       | 478       | 925       | 72.7  | 3.78        | 0.72      | 7.83      | 0.455 |
| Eu                                          | 0.251               | 0.160     | 0.361     | 0.010 | 145       | 121       | 181       | 9.1   | 1.95        | 0.51      | 3.90      | 0.211 |
| Gd                                          | 0.117               | 0.033     | 0.320     | 0.018 | 505       | 347       | 698       | 58.4  | 19.6        | 4.8       | 36.9      | 2.13  |
| Tb                                          | 0.014               | 0.005     | 0.029     | 0.003 | 72        | 47        | 100       | 8.86  | 6.6         | 1.8       | 11.8      | 0.675 |
| Dy                                          | 0.043               | 0.010     | 0.148     | 0.008 | 402       | 259       | 565       | 52.6  | 85.7        | 26.3      | 151.2     | 8.29  |
| Ho                                          | 0.010               | 0.004     | 0.019     | 0.002 | 77        | 52        | 106       | 9.81  | 36.6        | 12.5      | 64.0      | 3.22  |
| Er                                          | 0.021               | 0.012     | 0.029     | 0.002 | 196       | 133       | 257       | 23.2  | 187         | 67        | 315       | 15.32 |
| Tm                                          |                     |           |           |       | 27.8      | 19.9      | 35.0      | 3.01  | 44.9        | 17.9      | 73.1      | 3.33  |
| Yb                                          | 0.046               | 0.040     | 0.050     | 0.003 | 172       | 132       | 207       | 15.5  | 477         | 209       | 739       | 31.81 |
| Lu                                          | 0.007               | 0.004     | 0.011     | 0.001 | 20.2      | 16.2      | 24.9      | 1.67  | 109         | 51        | 164       | 6.58  |
| Hf                                          | 0.106               | 0.059     | 0.165     | 0.004 | 25.7      | 22.6      | 30.6      | 1.12  | 9652        | 7670      | 11240     | 184   |
| Ta                                          | 0.705               | 0.532     | 0.793     | 0.012 | 189       | 139       | 276       | 21.9  |             |           |           |       |
| Pb                                          | 2.478               | 2.170     | 2.860     | 0.039 | 1.61      | 1.54      | 1.74      | 0.03  |             |           |           |       |
| Th                                          | 0.028               | 0.006     | 0.106     | 0.005 | 269       | 236       | 312       | 11.0  | 884         | 147       | 1950      | 127   |
| U                                           | 0.024               | 0.005     | 0.055     | 0.003 | 67        | 56        | 86        | 4.39  | 1051        | 364       | 1707      | 97    |

**Table 1** Mean trace element composition of the fresh microlite-free interstitial glass in the felsic clot and the main mineral phases. Minimum and maximum values and the calculated SEE (standard error estimate) values are also presented

| Calculated mineral/melt partition coefficients |                  |           |                  |         |                |         |                |         |                |         |  |
|------------------------------------------------|------------------|-----------|------------------|---------|----------------|---------|----------------|---------|----------------|---------|--|
|                                                | amphibole/melt   |           | plagioclase/melt |         | biotite/melt   |         | titanite/melt  |         | zircon/melt    |         |  |
|                                                | K <sub>d</sub> * | uncert.** | K <sub>d</sub>   | uncert. | K <sub>d</sub> | uncert. | K <sub>d</sub> | uncert. | K <sub>d</sub> | uncert. |  |
| Li                                             | 1.089            | 0.079     | 0.301            | 0.010   | 0.057          | 0.007   |                |         |                |         |  |
| Sc                                             | 4.630            | 0.160     | 0.548            | 0.018   | 0.973          | 0.031   | 0.534          | 0.036   |                |         |  |
| V                                              | 84               | 9         | 0.156            | -       | 104            | 10      | 194            | 21      |                |         |  |
| Rb                                             | 0.015            | 0.001     | 0.012            | 0.003   | 2.409          | 0.041   | 0.002          | 0.000   |                |         |  |
| Sr                                             | 0.740            | 0.051     | 17.9             | 1.0     | 0.133          | 0.008   | 0.482          | 0.047   | 0.003          | 0.000   |  |
| Y                                              | 6.224            | 0.298     | 0.025            | 0.011   | 0.025          | 0.003   | 358            | 42      | 210            | 19      |  |
| Zr                                             | 0.559            | 0.026     | 0.006            | 0.001   | 0.046          | 0.001   | 5.891          | 0.411   |                |         |  |
| Nb                                             | 1.361            | 0.038     | 0.019            | 0.009   | 2.668          | 0.048   | 154            | 10      | 0.629          | 0.074   |  |
| Ba                                             | 0.080            | 0.005     | 0.378            | 0.009   | 3.643          | 0.156   | 0.002          | 0.001   | 0.000          | 0.000   |  |
| La                                             | 0.845            | 0.045     | 0.284            | 0.009   | 0.017          | 0.003   | 44.8           | 4.8     | 0.005          | 0.002   |  |
| Ce                                             | 1.821            | 0.084     | 0.184            | 0.006   | 0.023          | 0.005   | 111.3          | 9.8     | 1.651          | 0.230   |  |
| Pr                                             | 3.260            | 0.159     | 0.147            | 0.006   | 0.043          | 0.008   | 205.8          | 18.8    | 0.042          | 0.005   |  |
| Nd                                             | 5.179            | 0.328     | 0.124            | 0.007   | 0.067          | 0.013   | 320.0          | 34.8    | 0.213          | 0.030   |  |
| Sm                                             | 7.976            | 0.588     | 0.295            | 0.192   | 0.258          | 0.045   | 510.3          | 64.8    | 2.987          | 0.401   |  |
| Eu                                             | 7.083            | 0.460     | 2.206            | 0.128   | 0.716          | 0.048   | 415.1          | 34.2    | 5.572          | 0.672   |  |
| Gd                                             | 8.447            | 1.245     | 0.155            | 0.031   | 0.118          | 0.024   | 508.2          | 92.4    | 19.8           | 3.5     |  |
| Tb                                             | 9.163            | 0.827     | 0.360            | 0.228   | 0.114          | 0.027   | 580.2          | 85.0    | 53.7           | 6.9     |  |
| Dy                                             | 7.989            | 0.400     | 0.128            | 0.015   | 0.053          | 0.010   | 498.3          | 66.3    | 106.3          | 10.6    |  |
| Ho                                             | 7.367            | 0.740     | 0.162            | 0.023   | 0.058          | 0.012   | 434.3          | 67.7    | 205.9          | 26.0    |  |
| Er                                             | 6.736            | 0.507     | 0.123            | 0.037   | 0.041          | 0.005   | 385.8          | 52.2    | 368.1          | 38.7    |  |
| Tm                                             | 6.209            | 0.703     |                  |         |                |         | 351.0          | 53.6    | 567.2          | 74.2    |  |
| Yb                                             | 4.796            | 0.491     |                  |         | 0.067          | 0.008   | 253.0          | 33.5    | 703.2          | 82.7    |  |
| Lu                                             | 3.876            | 0.528     |                  |         | 0.053          | 0.009   | 160.3          | 25.1    | 860.3          | 126.0   |  |
| Hf                                             | 0.846            | 0.055     | 0.021            | 0.002   | 0.045          | 0.003   | 10.9           | 0.8     | 4087.4         | 233.5   |  |
| Ta                                             | 0.351            | 0.022     | 0.024            | 0.004   | 0.614          | 0.029   | 165.0          | 20.4    |                |         |  |
| Pb                                             | 0.046            | 0.004     | 0.421            | 0.031   | 0.063          | 0.005   | 0.041          | 0.003   |                |         |  |
| Th                                             | 0.010            | 0.001     | 0.005            | 0.001   | 0.001          | 0.000   | 13.6           | 0.6     | 44.8           | 6.5     |  |
| U                                              | 0.009            | 0.001     | 0.007            | 0.001   | 0.003          | 0.000   | 9.2            | 0.6     | 146.1          | 13.7    |  |

\* For mineral phases, cleared average trace element concentration data was used. Melt phase is represented by the average trace element concentration data of interstitial glass in the felsic clots.

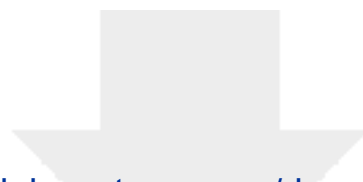
\*\* Uncertainty estimation for the calculated mineral/melt trace element partition coefficients was performed with standard error estimate (SEE = standard deviation / square root of n, where n is the number of measurements) and uncertainty propagation:

$$\text{SQRT}((\text{mineral SEE}/\text{mineral mean})^2 + (\text{glass SEE}/\text{glass mean})^2) * K_d$$

**Table 2** Calculated mineral-melt partition coefficients with the uncertainty values for the Haramul Mic low-T silicic volcanic system

|           | $D_0$    | $D_0 \pm 1\sigma$ | $E$ (GPa) | $E \pm 1\sigma$ | $r_0(\text{\AA})$ | $r_0 \pm 1\sigma$ |
|-----------|----------|-------------------|-----------|-----------------|-------------------|-------------------|
| amphibole | 9.2179   | 0.1796            | 363.1     | 10.1            | 1.0489            | 0.0006            |
| titanite  | 580.9719 | 4.5073            | 410.7     | 5.0             | 1.0537            | 0.0004            |
| zircon    | 994.0153 | 40.7910           | 619.9     | 11.5            | 0.9401            | 0.0019            |

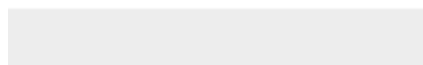
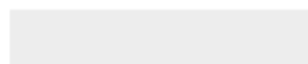
**Table 3** Crystal lattice parameters for the trivalent cations in amphibole, titanite and zircon, determined by the best-fit parabola using the fitting method of Dalou et al. (2018) in the Onuma plots presented on Fig. 8.

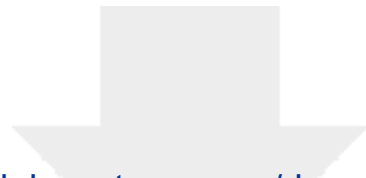


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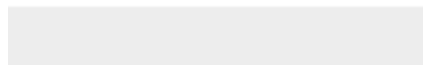
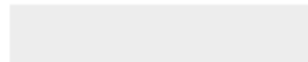


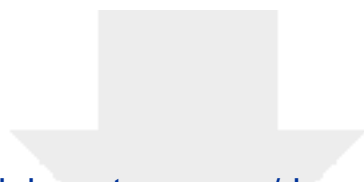


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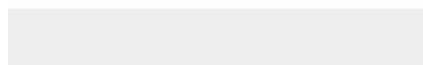
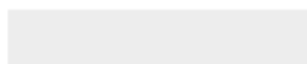


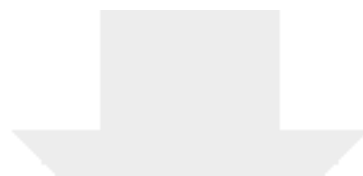


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