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1 **Growth and erosion rates of the East Carpathians volcanoes**
2 **constrained by numerical models: tectonic and**
3 **climatic implications**

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15 **Abstract**

16 The East Carpathian volcanic range experienced an along-arc, Late Miocene to
17 Quaternary migration of eruptive activity during its ~11Ma-long activity. Here, a novel and
18 complex methodology is presented that yields new geochronological and geomorphological
19 constraints on the evolution of the 20 volcanic edifices. New unspiked K-Ar ages either
20 constrain their lifespan (6.79±0.10 - 6.47±0.09 Ma for Seaca-Tătarca; 5.47±0.08 - 4.61±0.07
21 Ma for Vârghiș) or date the youngest volcanic activity (central Călimani). In parallel,

22 numerical reconstructions of volcanic paleo-topographies were performed to quantify their
23 shape at the end of their construction stage. The inferred initial volcano size shows a wide
24 range (3 to 592 km³), making up the four main successive volcanic segments (910, 880, 279
25 and 165 km³ for Călimani, Gurghiu, North Harghita and South Harghita segment,
26 respectively) totalizing 2200 km³ and an average growth rate of 200 km³/Ma at range-scale.
27 At the volcano-scale, with only consideration to their respective time spans (i.e. avoiding
28 repose periods), growth rate is characterized by two major trends: a moderate growth rate
29 (137 km³/Ma) for the older volcanoes (11-3.6 Ma) followed by a lower growth rate (28
30 km³/Ma) obtained for the Plio-Quaternary volcanoes. Comparing reconstructed and current
31 topographies yielded a total eroded volume of 524±125 km³, defining averaged denudation
32 of 22% and a 20 m/Ma erosion rate. Erosion rates for major climatic periods were computed,
33 which highlight the contrasting climatic contexts since 11 Ma. The highest erosion rate (38
34 m/Ma) occurred during transitional moderate subtropical-continental climate period (9.5-8.2
35 Ma). An intermediate erosion rate (14 m/Ma) characterized a moderate continental climate
36 period (8.2-6.8 Ma) when conditions became less humid. The lowest erosion rate (7 m/Ma)
37 reflects the prevailing continental but occasionally semi-arid climate (6.8-5.8 Ma). The
38 highest erosion rate (28 m/Ma) was obtained for Plio-Quaternary times during the
39 interglacial/glacial cycles. Such a quantitative morphometric and geochronological approach
40 demonstrates its efficiency to study volcanic dynamism, including both constructional and
41 erosional processes, through time.

42 **Keywords**

43 Numerical reconstruction; Volcano growth; Erosion rate; Paleo-climatic constraint

44 **1 Introduction**

45 A major geomorphological approach consists in assessing the time-dependent evolution
46 of paleo and present topographies of landforms alongside the geological processes involved
47 in their genesis, and, if possible, in numerically quantifying such processes (Leverington et
48 al., 2002; Persson et al., 2004; Székely and Karátson, 2004; Wright et al., 2006; Grosse et al.,
49 2009; Tucker and Hancock, 2010; Grosse et al., 2012; Karátson et al., 2012; Lahitte et al.,
50 2012; Salvany et al., 2012). In volcanic fields with readily dateable topographic surfaces, one
51 can constrain both dynamisms of volcanic growth and degrees of dismantling. Obviously,
52 short-lived, recent, well datable volcanoes (i.e. of Late Quaternary age) can be studied with a
53 high level of precision, whereas for older systems the precision will decrease. Growth and
54 destruction of volcanoes may or may not be coupled, controlled by either endogenic
55 (tectonic forces, volcanic eruptions) or exogenic processes (erosion controlled by climatic
56 factors) (Thouret, 1999). A number of studies have aimed at constraining the total long-term
57 erosion of volcanic surfaces using two approaches: 1) investigate long-lived but steady
58 sourced volcanic systems such as stratovolcanoes based on edifice changes to track long-
59 term erosional processes (Hildenbrand et al., 2008; Germa et al., 2011; Lahitte et al., 2012;
60 Karátson et al., 2012; Salvany et al., 2012; Ricci et al., 2015a); and 2) study short-lived but
61 dispersed small volcanic systems such as monogenetic volcanic fields using individual point-
62 like data to extrapolate and understand the entire field behaviour (Kereszturi et al., 2013;
63 Blaikie et al., 2015; Grosse et al., 2018). However, in the case of active volcanoes (with
64 current eruption), a mass balance can be carried out either by quantifying the evolution of
65 the topography of the volcano before, after, during and after the eruption or by carrying out
66 a balance of the sediment load which is then reported on the surface of the watershed
67 (Thouret et al., 2014). Digital elevation models (DEM) allow for characterizing the patterns of

68 erosion, and to extract geomorphic and structural information on large-volume edifices
69 which cannot be easily studied in the field (Favalli et al., 1999; Székely and Karátson,
70 2004). DEM-based quantitative geomorphological investigations enable the calculation of
71 constructed and/or eroded volumes and thus estimate magma output as well as erosion
72 rates. These, in turn, can be linked to hydroclimate variability driving the long-term erosion
73 processes: in the Carpathians, a humid continental climate (Karátson and Timár, 2005;
74 Karátson et al., 2019); in other areas with tropical and/or arid climates (Le Friant et al., 2004;
75 Samper et al., 2007; Hildenbrand et al., 2008; Germa et al., 2010, 2015; Karátson et al., 2012;
76 Lahitte et al., 2012; Salvany et al., 2012; Ricci et al., 2015a, 2015b; Bablon et al., 2018).

77 This study combines new geochronological constraints with a quantitative
78 geomorphological overview of the Călimani-Gurghiu-Harghita¹ volcanic range (hereafter
79 CGH, Fig. 1) in order to decipher its long-term topographic evolution. The unspiked
80 Cassinot-Gillot K-Ar technique has been used to obtain precise ages on four of the 17
81 volcanic centres (Central Călimani, Seaca-Tătarca, Vârghiş and Pilişca volcano). Also, we
82 performed DEM analysis to numerically model the paleo-topography of the volcanoes by
83 computing the best fitting surface to the volcanoes, making it possible to obtain constructed
84 and eroded volumes. Combining geomorphological information with available
85 geochronological data allow us to accurately assess the geomorphic evolution by calculating
86 growth and erosion rates, which in turn help constrain the timing and dynamism of
87 magmatic processes and to investigate links between tectonic and magmatic dynamism
88 (Wadge, 1982, 1984; Grosse et al., 2018).

¹ Official Romanian names are completed by locally used Hungarian names in Tables, also helpful for the reader to find names on local maps.

89 From cumulative erosion rates, computed at the scale of each volcano, we investigate their
90 variations and how they correlate to the main climatic fluctuations under variegated but
91 temperate continental climate-dominated conditions (Karátson, 1996), that the CGH area
92 has experienced since the onset of volcanism started at 10.6 Ma (Pécskay et al., 1995).

93 **2 Geological setting**

94 The Carpathians are a major mountain range in Central and Eastern Europe spanning
95 from Eastern Bohemia (Czech Republic), Poland, Slovakia, Hungary, Ukraine, to Romania and
96 Serbia. Linked to the Alpine and Dinaric systems, they encompass the Pannonian and
97 Transylvanian Basins (inset Fig. 1). Volcanic activity related to complex Tertiary to
98 Quaternary geodynamic processes occurred both along the inner part of the Carpathian
99 orogen and in the Carpatho-Pannonian Basin (Rădulescu and Săndulescu, 1973; Csontos et
100 al., 1992; Szabó et al., 1992; Seghedi, 1995; Mason et al., 1996, 1998; Seghedi et al., 1998,
101 2004a, 2004b, 2005a, 2005b; Fielitz and Seghedi, 2005; Seghedi et al., 2019) .

102 Located in the eastern part of Romania's Inner Carpathians, the 160 km long CGH
103 volcanic range constitutes the youngest, longest and southeasternmost segment of the
104 Carpathian volcanic range (Fig. 1). The CGH is a typical post-collisional volcanic range as it
105 developed after the stacking of the collision-related East Carpathian nappe system (Matenco
106 and Bertotti, 2000). After the collision and the step-by-step propagation of the CGH
107 volcanism parallel with the orogen, the plate boundaries more or less sealed. It was assumed
108 to be associated with asthenosphere upwelling, explained by progressive break-off of an
109 assumed Miocene subducted slab (Mason et al., 1998; Seghedi et al., 1998), as suggested by
110 the present high heat flow coinciding exclusively with the volcanic area (Tari et al., 1999;
111 Demetrescu et al., 2001). The CGH consists of a NW–SE-trending row of several adjoining

112 and partially overlapping volcanoes mostly characterized as composite volcanoes in the axial
113 part and resulting from complex eruptive histories, including several cone-building phases
114 interrupted by destruction phases (Szakács and Seghedi, 1995). The subduction-related,
115 post-collisional volcanism (Rădulescu and Săndulescu, 1973; Pécskay et al., 1995; Seghedi et
116 al., 2004a; Lexa et al., 2010) resulted in products which display calc-alkaline to alkaline
117 compositions with andesite-dominated rock types (Mason et al., 1996, 1998; Seghedi et al.,
118 2011). Volcanism of the overall CGH segment was interpreted as coeval with the last peak of
119 crustal deformation in the Carpathian bend zone, characterized by uplift in the orogenic
120 zone and subsidence in the foreland with amplitudes in the order of 2-4 km (Matenco et al.,
121 2010).

122 **3 Volcanism of the CGH range and its timing**

123 The CGH volcanic range divides into four sub-segments corresponding respectively from
124 the north to south to Călimani, Gurghiu, Northern and Southern Harghita (Fig. 1). As a result
125 of the migration of volcanic activity along the range, the overlap between its 17 eruption
126 centres is limited, allowing one to individualize them quite well. The first comprehensive
127 study of CGH (Szakács and Seghedi, 1995, 1996) pointed out the systematic decrease of
128 volume, height and width of the volcanic edifices from north (Călimani) to south (Southern
129 Harghita), most of them surrounded by extended and partly coalesced volcanoclastic aprons.

130 The pre-volcanic basement of the CGH volcanic range belongs to two different major
131 tectonic units, lithologically characterized by Proterozoic to Lower Palaeozoic metamorphic
132 rocks overlain by Mesozoic, mostly carbonatic, sedimentary rocks of the East Carpathian's
133 "Crystalline-Mesozoic Zone" in the eastern part (Săndulescu, 1988). A thick Paleogene-
134 Neogene sedimentary pile including ductile rocks such as clay and salt prone to plastic

135 deformation (Ciupagea et al., 1970; Szakács and Krézsek, 2006) forms the pre-volcanic
136 basement in the western part along the rim of the Transylvanian Basin. A topographic
137 “anomaly” consisting in reverse-dipping (i.e. towards volcano center instead of towards
138 volcano periphery) of the peripheral outer slopes of some volcanoes (e.g. Făncel-Lâpușna
139 and Seaca-Tătarca in Gurghiu Mts, and Vârghiș volcano in North Harghita, Fig. 1) was
140 interpreted by Szakács and Krézsek (2006) as resulting from complex interactions between
141 the pre-volcanic basement and the overlying volcanic edifices. Namely, the Late Miocene
142 volcanoes grew up on a pre-volcanic basement including a thick salt layer which was
143 squeezed-out, displaced and involved in enhanced diapirism governed by gravitational
144 effects of the volcano loading. These processes modified in a certain degree also the original
145 volcanic topography by volcano spreading (Delaney, 1992; Borgia, 1994; Merle and Borgia,
146 1996; Lagabriele et al., 2000; Wooller et al., 2004; Borgia et al., 2005; Grosse et al., 2009,
147 2012), namely increasing edifice diameter and reducing relative height differences between
148 volcano centre and periphery, hence reducing relief energy and erosion rates.

149 Available ages along the CGH range (Peltz et al., 1987; Szakács et al., 1993; Pécskay et
150 al., 1995, 2006; Seghedi et al., 2004b, 2005; Karátson et al., 2019; Lahitte et al., 2019)
151 defined a quasi-continuous eruptive activity from 10.6 to 0.03 Ma. The southwardly shift in
152 eruption centres has been linked to progressive slab detachment following an oblique
153 subduction stage (Mason et al., 1998; Seghedi et al., 1998).

154 For the past 10.6 Ma, the CGH range has been experiencing a temperate subtropical-
155 continental climate with occasionally arid periods in the Late Miocene and a relatively humid
156 climate with occasional periglacial regimes and interglacial/glacial variability during the
157 Pliocene and Quaternary (Karátson, 1996; Fielitz and Seghedi, 2005; Magyari et al., 2014).

158 Details and evidence concerning climate changes in the Carpathian area are given in the
159 discussion.

160 **4 Methods**

161 **4.1 K-Ar ages by unspiked Cassinot-Gillot technique**

162 Seventeen new groundmass K-Ar ages are first presented here (Table 1). Results were
163 obtained on samples collected from four volcanoes representing the three main sectors of
164 the range (Călimani, Gurghiu, North and South Harghita). Measurements followed the
165 protocol of rock sampling, preparation and measurement as described by Germa et al.
166 (2011) and detailed in the supplementary materials (S.M.).

167 **4.2 DEM-based volume calculations**

168 Using a quantitative DEM-based approach of volumetry (Székely and Karátson, 2004;
169 Karátson and Timár, 2005; Favalli et al., 2009; Grosse et al., 2009; Kervyn et al., 2012; Lahitte
170 et al., 2012; Lavigne et al., 2013; Karátson et al., 2013; Germa et al., 2015; Ricci et al.,
171 2015a), we numerically modelled the paleo-topographies of all CGH volcanoes. To constrain
172 the shape that they displayed directly after the end of their respective activity, we follow the
173 method introduced by Lahitte et al. (2012) and improved since by Germa et al. (2015). The
174 resultant modelled volcanic surfaces are used to assess constructed and eroded volumes
175 (Ricci et al., 2015a), and then, using the available chronological data, to constrain the time-
176 space dynamism of volcano surface evolution through both construction and erosion rates.
177 To achieve our objectives, the topography of the CGH volcanic range was analysed from the
178 Shuttle Radar Topography Mission (SRTM) 30m-resolution DEM. Topographic surfaces were
179 modelled at 100m-resolution that compromises sufficient details of reconstruction and time

180 of calculation without affecting their quality (Wright et al., 2006; Grosse et al., 2009;
181 Karátson et al., 2010; Kervyn et al., 2012).

182 4.2.1 Volcanic paleo-surface model

183 The main body of work relates to the topographic reconstruction of a volcano, i.e. the
184 modelling of the surface that it reached after its growth ceased (the so-called pre-erosional
185 volcanic surface, or PEVS). To model such surfaces, we use as input data the sectors of the
186 current topography that still testify to the PEVS or, in other words, that have undergone
187 ideally zero or minimal erosion since the volcano construction ceased. In detail, each grid cell
188 of the present-day DEM was converted in a XYZ point, forming a huge input database (20
189 million points). Next, different morphometric parameters such as slope, topographic
190 position index (TPI, Weiss, 2001) and topographic openness (TO, Yokoyama et al., 2002;
191 Favalli and Fornaciai, 2017) were computed, assigned as attributes for each point and
192 combined to select only points belonging to PEVS. These so-called constraining points (CPs)
193 are commonly located on quasi-planar surfaces found commonly in the mid- to lower flanks
194 of the volcanoes which, during long-term erosion, tend to be preserved relatively to the
195 upper flanks and central parts that encompassed central depressions (Karátson et al., 2016;
196 Favalli and Fornaciai, 2017). More than 200,000 CPs for the entire CGH volcanic range (Fig.
197 2A) were extracted. For practical reasons, they were reduced to one tenth without breaking
198 the homogeneity of their distribution. The Seaca-Tătarca volcano (06 in Fig. 1) example is
199 shown in Figure 3 to illustrate CPs extraction (purple dots in Fig. 2A, Fig. 3A and Fig. 3C) from
200 its whole surface (green dots in Fig. 3A and coloured surface in Fig. 3C) and in consideration
201 to slope, exposition, TO and TPI parameters. The elevation versus distance to centre diagram
202 (Fig. 3A) highlights how CPs, independently of their position along the volcano profile (lower,
203 mid or upper), belong to planezes surfaces, i.e. to the locally (at hundred-meter scale)

204 uppermost surface of the volcano. Note that only the main edifice was selected without
205 considering the volcanoclastic materials of the ring plain immediately surrounding the central
206 edifice (Zernack et al., 2011; Németh and Palmer, 2019). Finally, a careful visual control has
207 been carried out in order to remove outlier points accidentally present in the ultimate CPs
208 database. From their respective CPs, numerical reconstructions of 15 from the 16 selected
209 volcanoes have been modelled using the ShapeVolc (SV) algorithm (Lahitte et al., 2012). Only
210 Pilişca volcano (a small-sized shield volcano topped by a few lava domes in South Harghita;
211 Szakács and Seghedi, 1995) has been reconstructed by the Kriging interpolation (Krige,
212 1951).

213 The ShapeVolc algorithm dedicated to volcano surface modelling was used successfully
214 in various dismantling contexts: large flank collapse (Lahitte et al., 2012; Germa et al., 2015),
215 caldera formation (Lavigne et al., 2013) or regressive erosion (Salvany et al., 2012; Ricci et
216 al., 2015a, 2015b). This algorithm, as well as the one proposed by Favalli et al. (2014), fit the
217 ideal surface by minimizing the mean square error between this surface elevation and those
218 of the CPs.

219 ShapeVolc algorithm, assuming that a volcano displays at first order an axial symmetry
220 (Karátson et al., 2012), finds, using the simplex algorithm, the set of parameters that define
221 the first order geometry of a volume defined by a surface of revolution. Adjusted parameters
222 are: XYZ coordinates of the axis of revolution (the location and elevation of the
223 vent/summit); the elliptical shape of the edifice (i.e. the shape of its contour lines); and the
224 parameters of the generatrix of the surface revolution (i.e. the function, black dots in Fig. 3B,
225 that best fit the non-eroded upper volcanic surface). As the current elevation of each CPs is
226 the lowest one possible for the uppermost surface, parameters are calculated in order to

227 find the lowest possible surface of revolution passing above most of the CPs (see black curve
228 relatively to purple dots in Fig. 3A).

229 A common way to model volcano shape and volume consists in a simple rotation of the
230 best generatrix curve around an axis located at the volcano summit, defining a pure surface
231 of revolution (Karátson et al., 2010, 2012). However, such kind of modelling does not
232 consider the ellipticity of the volcano that is a common characteristic of edifices (Grosse et
233 al., 2012, 2014; Favalli et al. 2014; Grosse and Kervyn, 2018). The observed improvement of
234 results, when ellipticity is considered advocate for closely considering volcano eccentricity in
235 our reconstructions (see details given in S.M.), has been done in this study: eccentricity and
236 azimuth have been systematically computed.

237 Moreover, as taking into account the volcanic complexity of the surface is crucial to
238 optimize precision in volume calculation, a major improvement is here introduced to allow
239 ShapeVolc to take into account the second order of magnitude of details of volcanic
240 surfaces. Indeed, if most volcanoes can be defined at first order by a mathematically
241 defined, elliptical surface of revolution, the complexity of volcanic construction introduces
242 heterogeneities on this mathematic perfection that commands modelling the second order
243 of magnitude of their shape. Concretely, each PEVS model was performed in two steps. 1)
244 The first order of magnitude of the volcanic surface was obtained by defining the unique set
245 of parameters (listed above) that best fit to all CPs of the volcano. 2) The second step locally
246 adjusts the surface to find elevation of each cell of the 100 m-resolution paleo-surface
247 model with only considering the closest CPs of the given cell and selected by classical
248 research criteria (typically the 20 nearest CPs in each of the eight 45°-large angular sectors).
249 This time-consuming step (up to 120,000 elevation points to be modelled for a large
250 volcano) allows for defining the uppermost PEVS and takes into account the irregularities of

251 the surface such as local elevation variations and small details in the volcanic structure
252 (presence of parasitic and/or adventive cones or domes, such as Tarvez in Fig. 3A). This
253 second order of detail becomes essential for young volcanoes where the very small amount
254 of erosion material imposes the most adjusted surface as possible.

255 Uncertainty of our modelled volcano shapes ($\sigma_{Z_{SV}}$, Table 2) is calculated as the standard
256 deviation of the difference (ΔZ_p) between the altitude of the constraining points (Z_{CP}) and
257 the altitude of the modelled ones (Z_{SV}).

258 4.2.2 Volumetric data, growth and erosion rates

259 Estimating the volcano volume requires knowing the surface of its basement prior to its
260 construction, the so-called pre-volcanic basal surface (PVBS). Details of PVBS modelling by
261 kriging interpolation are given in the S.M.

262 Comparisons of pre-erosional volcano surface to PVBS and the present DEM allow
263 calculations of constructed (V_C) or eroded (V_E) volumes, respectively. Next, the growth rates
264 (GR, in km^3/Ma) defined by V_C divided by volcano lifespan (Δt) constrains the eruptive
265 dynamism; the erosion rate (ER, in m/Ma) defined by V_E divided by the area and the erosion
266 duration (i.e.; volcano age) and the degree of denudation (D_{Den} , the percentage of volcano
267 volume removed by erosion), defined by V_E divided by V_C , are computed. Details of the
268 modelling of PVBS and calculation of volumes and uncertainties, degree of denudation,
269 growth and erosion rates are given in the S.M. The inferred erosion is highlighted in Fig. 3C
270 by the black sticks that connect the present DEM as well as the PEVS surfaces.

271 Our reconstructions include both coherent and fragmented volcanic materials making
272 up the edifices, and thus, they refer to bulk volumes instead of DRE (dense rock equivalent).
273 Our constructed volumes and growth rates take into account the volcanoclastic aprons, but
274 they are excluded from the V_E and ER since our calculations do not differentiate the

275 volcaniclastic aprons (their source and to which volcano they belong); so, an accurate
276 proportioning is quasi-impossible. Our volcano GR differs from magma output rates sensu
277 stricto as they only integrate a part of the whole pyroclastic material. However, it allows the
278 obtaining of a general framework of magmatic activity through space and time along the
279 CGH range and can be used as a good proxy for magma output rates (Wadge, 1982, 1984;
280 Grosse et al., 2018).

281 **5 Results**

282 **5.1 K-Ar ages**

283 New K-Ar ages are reported in Table 1, with all uncertainties quoted at 1-sigma level
284 (1σ). All Ar analyses were successfully duplicated at the 1σ level except sample 17EC14 that
285 required a third measurement (see details of each measurement in the S.M.). Radiometric
286 dating results are presented in chronological order in the following sections, compiled in
287 Table 1 and discussed in Fig. 2A-D.

288 **5.1.1 Călimani edifice**

289 The four oldest dated samples of lava flows and intrusions belong to the Călimani
290 caldera edifice. The oldest ages (8.30 ± 0.12 and 8.33 ± 0.12 Ma) were obtained on two distinct
291 outcrops from the same andesite lava flow, illustrating the good reproducibility of our
292 dating method. The monzodioritic intrusion exposed in the central part of the Călimani
293 caldera is slightly younger (8.26 ± 0.12 Ma). In contrast, the dacitic lava dome at Pietricelul,
294 built up on the Călimani caldera rim, constrains the post-caldera activity (6.69 ± 0.10 Ma) and
295 is comparable with the youngest ages measured previously in the Călimani area (Pécskay et
296 al., 1995).

297 **5.1.2 Seaca-Tătarca volcano (Gurghiu Mts.)**

298 Five andesitic lava flows were dated on this large lava dominated volcano.
299 Stratigraphical coherence is observed, as the oldest ages have been obtained on the
300 innermost and lowest samples (6.65 ± 0.09 and 6.79 ± 0.10 Ma). Two intermediates ages were
301 obtained from lava flows on the upper flanks of the volcano on the SSE and western sides
302 (6.61 ± 0.09 and 6.52 ± 0.09 Ma, respectively). The youngest age was obtained on the
303 uppermost rim of the erosional depression (6.47 ± 0.09 Ma) in accordance with the relative
304 stratigraphy. From the dated samples (that likely constrain the whole Seaca-Tătarca volcano
305 stratigraphy), we infer a constructional period of $\sim 330\pm 140$ ka.

306 **5.1.3 Vârghiş volcano (North Harghita Mts.)**

307 From the six andesite lava flows dated on the Vârghiş volcano, the oldest age (5.47 ± 0.08
308 Ma) has been obtained on the only amphibole lava located in the lowest outcrop of the
309 north sector of the central depression (identified as a sector-collapse depression by (Szakács
310 and Seghedi, 1995, 2000). Two lava flows sampled inside the depression have provided
311 intermediate ages (5.06 ± 0.07 and 4.79 ± 0.07 Ma) whereas two other (4.65 ± 0.07 and
312 4.89 ± 0.07 Ma) came from the upper flank of Harghita Mădăraş summit, described as a
313 depression rim dome (Szakács and Seghedi, 1995). Finally, the post sector collapse and
314 youngest activity is constrained at the southern periphery of the edifice (4.61 ± 0.07 Ma).
315 From these ages, we infer a constructional period of $\sim 900\pm 140$ ka.

316 **5.1.4 Pilişca volcano (South Harghita)**

317 Two dacitic lava rocks from the Pilişca volcano (1.98 ± 0.06 and 2.11 ± 0.04 Ma) might
318 represent late-stage dacitic lava domes stratigraphically overlying andesitic lavas of the early
319 volcano stage located along the riverbed of the Olt river in the Tuşnad Gorge (2.11 ± 0.03 and
320 2.13 ± 0.03 Ma, in Karátson et al. 2019).

321 5.2 Volcanic surface models

322 5.2.1 Paleo-volcanic surfaces

323 From their constraining points (Fig. 2A), 16 CGH volcanoes were modelled (Fig. 4). Their
324 morphometric characteristics are summarized in Table 2 (more exhaustively discussed in
325 S.M.). They display two types of radial profile according to their main concave (modelled by
326 an exponential or polynomial curve) or convex (gaussian curve) shape (Grosse et al., 2009).
327 Ellipticity ranges from 0.16 (quasi-circular edifice) to 0.80 (strongly elongated one).
328 Uncertainties of modelled surfaces range from 24 m to 82 m. Modelled volcanoes are
329 characterized by contrasting sizes, from very small parasitic cones (Râchitis edifice, #12, 3 ± 3
330 km^3) to large composite volcanoes (Central Călimani edifice, #01, $592\pm 115 \text{ km}^3$; Fig. 1 and
331 Fig. 4).

332 In the Călimani sector (Fig. 4A), the Central Călimani (#01, constituted by the current S and
333 SE flanks of Rusca-Tihu and the associated cones of Lucaciul and Tămău) and Moldovanul
334 (#02) edifices have been reconstructed. They display contrasting shapes; the quasi-circular
335 Central Călimani is best fitted by a concave shape (steep summit) whereas the more elliptical
336 Moldovanul fits to a convex shape (relatively flat summit).

337 For Gurghiu sector (Fig. 4B), seven volcanoes have been modelled (#03 to #09, Fig. 4B).
338 Whereas all have quite similar concave shape, four volcanoes (#03, #04, #05, #08) exhibit
339 relatively high ellipticity (0.61-0.80) whereas the others are almost circular (0.16-0.37).

340 In North Harghita, all four modelled volcanoes (#10 to #13, Fig. 4C) display a marked
341 ellipticity (0.33-0.80) and concave shape, except the very small Râchitis with a convex
342 profile.

343 In South Harghita (Fig. 4D), the dual Luci-Lazu (#14a and #14b) and Cucu (#15) volcanoes
344 display contrasting ellipticity (0.20 and 0.49, respectively) but both with quasi-linear profiles

345 (almost conical shape). Pilișca (#16), the youngest volcano in our study (only the late
346 Pleistocene Ciomadul is younger in the whole CGH, Szakács and Seghedi, 1995; Karátson et
347 al., 2019; Lahitte et al., 2019), has a such “un-geometric” shape that it requires kriging
348 interpolation.

349 5.2.2 Volumetric results

350 Using the output from the geomorphological models, the main individual data for each
351 volcano are compiled in Table 2 and comprise the area (A), Height, constructed (V_C) and
352 eroded (V_E) volumes, growth (GR) and erosion (ER) rates and the degree of denudation
353 (D_{Den}). To compute GR and ER, we considered all available radiometric ages from previous
354 studies, most remaining conventional K-Ar ages being obtained on whole rock. For each
355 volcano, we calculated an average age using as bracketing temporal constraints the oldest
356 and youngest available ages and assuming that the average age represents the peak of
357 activity.

358 The V_C computed for the 16 volcanoes modelled in this study ranges between 3 ± 3 and
359 592 ± 190 km³. Fitting the volcano volume and area by a power-law function shows a good
360 positive correlation ($R^2=0.93$; Fig. 5A) defined by:

$$361 \quad V_C = 0.015 \times (A)^{1.51} \quad \text{Eq. 1}$$

362 We note that data on Fâncel-Lăpușna (in Gurghiu) seems out of this general trend; however,
363 its inclusion or exclusion from the analysis practically does not alter the correlation (R^2
364 remains 0.93). Dimensional analysis shows that volume and surface are related by a power-
365 law function with a 1.5 exponent. For instance, a regular cone (radius R, height H, slope α) is
366 defined as:

$$367 \quad V = \tan(\alpha) / (3 \times \pi^{1/2}) \times A^{1.5} \quad \text{Eq. 2}$$

368 Using the computed volume and area of the 16 analysed volcanoes (some showing very
369 irregular shapes due to emplacement above or around older ones and having different
370 shapes, all displaying second order of magnitude, i.e. not a simple mathematical shape), we
371 obtained roughly the same 1.5 correlation exponent between the two variables (Fig. 5A). We
372 interpret this quasi-ideal correlation as the expression that, although each volcano
373 experienced its own history of construction, the individual long-term growth was averaged
374 out as a quite similar and common evolution along the range. Such a quantification
375 approach, based on enhanced morphometry, can be considered as numerical validation of
376 the trend predicted for arc volcanoes evolving from single cone to massif (Grosse et al.,
377 2009).

378 Plotting the height versus volume diagram as proposed by Grosse et al. (2009) (Fig. 5A
379 inset), they fall in the field of the massif trend which is coherent with the complexities of
380 their topography and eruptive history. Furthermore, the power-law curve fitting CGH data in
381 the height versus volume diagram (Fig. 5A inset) has a power exponent close to $1/3$ (0.35).
382 Again, this means, inverting the equation, that volume is almost related to height by a factor
383 of three and confirms the more and less homomorphic shapes of all CGH volcanoes.

384 The V_E computed in this study ranges between 0.2 km^3 (Ciomadul) and $158 \pm 85 \text{ km}^3$
385 (Călimani), increasing toward the north of the range with ER range between 7 and 26 m/Ma.
386 The degree of denudation of the studied volcanoes is in the range of 3–34% (the average
387 weighted mean being 22%), which means that within the studied period ($\sim 11 \text{ Ma}$), less than
388 one third of the original volcanic edifice has been removed by erosion.

389 Plotting the V_C against V_E shows a strong linear positive correlation with $R^2=0.95$ (black
390 line, Fig. 5B) whereas D_{Den} values (expressed as percentages in Fig. 5B) do not systematically

391 fit with the V_C , i.e. small edifices (e.g. Bacta, Jirca, Moldovanul), as well as large ones
392 (Călimani, Fâncel-Lăpușna) can display a high degree of denudation.

393 No correlation ($r^2=0.01$) exists between the degree of denudation (D_{Den}) and height (H)
394 (Fig. 6A) and, logically, with volume or area as they are strongly correlated. Additionally, no
395 effect on erosion by any altitudinal contrast appears. On the other hand, D_{Den} fit relatively
396 well with latitude ($R^2=0.62$, Fig. 6B). As morphometry of volcanoes cannot explain this fit,
397 other parameters such as age, spatial climate fluctuation, rock strength and edifice slope can
398 possibly drive this regional variation of the D_{Den} values. These aspects will be further
399 addressed in the discussion.

400 **6 Discussion**

401 **6.1 Volume comparison with previous results**

402 Summarizing individually modelled constructional volumes (i.e. prior to erosion) leads to
403 a total original volcanic volume of 2240 ± 190 km³ in the CGH range during ~11 Ma, higher
404 than the 1400–1606 km³ range previously proposed (Szakács et al., 1997; Karátson and
405 Timár, 2005; Seghedi et al., 2019) obtained with basic approaches. We interpret this higher
406 value compared to previous studies as due to the complete integration of erosionally
407 removed volumes over the whole edifice. Hence, our approach illustrates the importance of
408 reconstructing a full paleo-topography of the volcanic edifice, especially for heavily
409 dismantled volcanoes such as those in the Călimani and Gurghiu segment.

410 A clear anticorrelation exists between relative uncertainties of constructed volume
411 (σ_{RELV_C} , range from 19% to 50%, details in S.M.) and average height (ΔH_{mean} , Fig. 7) and fits
412 particularly well ($R^2=0.87$) to an inverse function. Even if calculation of relative uncertainty is

413 obtained from a complex integration of elementary uncertainties over the whole area of the
414 volcano, it can be approximated as

$$415 \quad \sigma_{\text{RELV}_C} \approx \text{sqrt} (\sigma_{\text{Mean_upper_surface}}^2 + \sigma_{\text{Mean_lower_surface}}^2) / \Delta H_{\text{mean}} \quad \text{Eq. 3}$$

416 where, ΔH_{mean} is the average elevation of the edifice, and both σ_{Mean} terms are the
417 average uncertainties of the basal and upper surfaces. Consequently, for a given volume, the
418 smaller the ΔH_{mean} , the more poorly constrained its average height, and the higher, the
419 relative uncertainty associated to its reconstructed volume (σ_{RELV_C}).

420 **6.2 K-Ar age constraints**

421 **6.2.1 Comparison with previous radiometric age data**

422 All new K-Ar ages (obtained on Central Călimani, Seaca-Tătarca, Vârghiș and Pilișca
423 volcanoes, Table 1, Fig. 2A-D) match within 1σ with the previous, respective conventional K-
424 Ar ages. As such, the main issue is not necessarily the absolute values but, rather, improving
425 their precision, since the uncertainty has been reduced by a factor of 3 to 5 (typically around
426 0.1 Ma vs 0.4 Ma). This improvement is crucial for correctly defining the dynamism of the
427 volcanism, which requires precise constraints on volcano lifespans.

428 The lifespan of Seaca-Tătarca volcano, for example, is shortened from a previous
429 estimate of 1 ± 0.4 Ma (Pécskay et al., 1995; Seghedi et al., 2004b) to 340 ± 140 ka. For Vârghiș
430 volcano, the new results slightly extend its youngest activity (4.61 ± 0.07 Ma, this work, vs 4.9
431 Ma in Pécskay et al., 1995) and refine its relatively long lifespan from 0.7 ± 0.4 to 0.9 ± 0.1 Ma.
432 For Pilișca volcano, newly dated units do not sufficiently encompass its full activity to allow
433 constraining its lifespan based only on the new K-Ar ages.

434 6.2.2 Example of volcano growth through time: The case of Seaca-Tătarca volcano

435 The study of the well-defined, regular-shaped simple edifice of Seaca-Tătarca benefits
436 from regularly distributed, well-dated samples, allowing for precisely correlating its volume
437 growth rate through time (Fig. 8A and Fig. 8B), and proposes a growth model encompassing
438 its whole construction phase. For this purpose, the dated samples are projected in a graph of
439 elevation versus distance to centre, with correction of the ellipticity of the edifice (Fig. 8A).
440 Next, considering the average topographic profile of the volcano at the end of its
441 construction and assuming a homothetic construction (i.e. shape remains self-similar all the
442 time), we computed the proportion of the volcano volume that was already constructed
443 with constant growth rate when subsequent, dated lava flows erupted.

444 Based on this, the location of the oldest sample (17EC03) is calculated as corresponding
445 to 10% of the construction of the edifice (Fig. 8A) which is consistent with its outcrop in the
446 large, central erosive depression that has not yet sufficiently dismantled the innermost part
447 of the volcano to reach its basement. Though coming from a lower elevation than the
448 previous one, sample 17EC04, belongs to an upper section of the volcano (21% of the
449 construction). It outcrops in a river valley whose topographic profile (represented by the
450 lowest green dots in Fig. 8A) is less sloping than the volcanic sequence it crosscuts.
451 Consequently, outcrops along this valley display younger and younger volcanic units toward
452 the periphery of the volcano. Sample 17EC05 corresponds to an intermediate position during
453 the construction of the edifice (65%) despite its location in the upper flanks. Indeed, as the
454 volcano grew, its area increased, too, requiring more and more material to increase its
455 elevation. Consequently, each new % of growth (i.e. elevation) occupied progressively lower
456 thicknesses (in Fig. 8A, blue lines regularly spaced at 10% are closer and closer upward). The

457 uppermost and youngest dated lava flows (17EC02 and 17EC01, Fig. 8A) correspond almost
458 to the last stage of the edifice construction (92% and 95%, respectively).

459 From these growth proportions through time (white dots in Fig. 8B), and assuming
460 magma output rate as constant (straight line in Fig. 8B), we can define the growth pattern of
461 the whole volcano. In particular, the age of the onset and end of its activity can be inferred
462 as the time interval where the growth line intersects 0% and 100% of growth proportions,
463 respectively. With consideration of uncertainties, this interpolation leads to a full
464 construction of Seaca-Tătarca volcano from 6.80 Ma to 6.46 Ma (red dots in Fig. 8B) and,
465 consequently, a lifespan of ~340 ka. Based on such detailed findings of a selected volcanic
466 edifice, we can assume that detailed chronological studies on similar-sized volcanoes along
467 CGH would result in faster growth rates as well.

468 **6.3 Spatio-temporal evolution of the CGH volcanism**

469 Compiling all available K-Ar ages (Fig. 9), i.e. the new unspiked K-Ar ages along with
470 those on Ciomadul and Pilișca and the numerous conventional K-Ar ages, the previously
471 recognized diachronism in volcanic activity along the CGH range can be confirmed, generally
472 characterized by decreasing ages southward from the late Miocene (~ 11 Ma) volcanism in
473 Călimani to the sub-recent (<0.1 Ma) activity at Ciomadul (Rădulescu and Săndulescu, 1973;
474 Peltz et al., 1987; Szakács et al., 1993, 2018; Pécskay et al., 1995, 2006; Seghedi et al., 2004b;
475 Seghedi et al., 2005; Karátson et al., 2019; Lahitte et al., 2019). However, detailed analysis
476 along the range highlights overlaps in volcanic activity between some segments. In
477 particular, the activity of Călimani and Gurghiu was partially simultaneous, roughly between
478 9.8 to 6.3 Ma (Seghedi et al., 2004b, 2005b; Pécskay et al., 2006). A second overlap is
479 observed between the latest activity in Gurghiu and the early activity of North Harghita (6.3 -
480 5.1 Ma).

481 Detailed observation of southward age progression along the CGH range allows for
482 observing various compartments in each segment (Fig. 9). For Călimani, volcanic activity was
483 coeval in both edifices (Central Călimani and Moldovanul), the earliest activities starting in
484 the northern area and then shifting southwards. This mechanism is highlighted by the
485 trapezoid box in Fig. 9 illustrating how early activity started only in the north but
486 subsequently took place simultaneously in the entire segment. In Gurghiu, also, southward
487 age progression does not establish a real migration, but, instead, an extension of the
488 volcanically active area (green trapezoid in Fig. 9). Isolating the oldest Gurghiu ages for each
489 20 km-large windows allows constraining its southward expansion (black line on Fig. 9).
490 Starting from only Jirca's activity at the beginning, the volcanism experienced a widespread
491 expansion of activity at a rate of 12.6 km/Ma ($R^2=0.75$) over the whole segment (all volcanic
492 centres being active quasi contemporaneously). It seems that during this period, the
493 volcanism reached its paroxysm and accounted for the main activity apparent at around 6.6
494 Ma (Fig. 10).

495 On the contrary, volcanism in North and South Harghita shows a well-defined
496 southward migration. A full progression of ages can be observed toward the south, as
497 activity in each subsequent volcanic centre generally begins at a progressively later date with
498 relatively good correlations. Between 6.3 and 4.6 Ma, volcanism migrated at a rate of 13.4
499 km/Ma ($R^2=0.72$) over the whole North Harghita segment. Subsequently, from 4.3 to 0.03
500 Ma, volcanism continued to progress southward along South Harghita. Samples obtained for
501 each volcanic centre show a narrow age range (typically <1 Ma), reflecting the migration of
502 activity but at a slower rate (4.6 km/Ma, $R^2=0.68$; Fig. 9) than along the older parts of the
503 CGH.

504 6.4 Growth rates evolution through time and tectonic implications

505 Combining ages with the new volume constraints allows for characterizing the major
506 phases of volcanic activity, the quiescence periods and the evolution of magma output (Fig.
507 10A). To quantify this, the volume of each volcano is assumed to be erupted between the
508 youngest and the oldest available ages, with a maximum output occurring at half-time of the
509 age range (Grosse et al., 2018) and individual GR through time is defined, at volcano-scale,
510 by a Gaussian-like curve (coloured curves in Fig. 10A). Next, summing all individual outputs
511 through time, we computed cumulative growth rate (dashed grey line in Fig. 10A). Finally,
512 the derivation of this curve gives the GR through time for the whole CGH volcanic activity
513 (bold line in Fig. 10A). Even though the regional GR does not consider the possible
514 subvolcanic bodies (see discussion in Karátson and Timár, (2005), it remains a good proxy for
515 investigating overall magma output rate.

516 Starting around 11 Ma, the CGH range experienced three main periods of volcanic
517 activity isolated by two long periods of relative quiescence (3.6–2.8 Ma and 1.5–0.9 Ma, Fig.
518 10A): first, a long and continuous period of activity between 10.6 to 3.6 Ma with a maximum
519 pulse of activity at 6.6 Ma (contemporaneous to the growth of Seaca-Tătarca volcano); and
520 next, a drastic decrease of volcanic activity with only episodic activities at 2.8–1.5 Ma and 1–
521 0.03 Ma during the growth of Pilișca and Ciomadul. The migration of volcanism can be
522 correlated to changes in the magma storage system (along with magma composition)
523 influenced or triggered by changes in plate tectonic processes (see global context in Crisp,
524 1984; White et al., 2006; East Carpathian features in Mason et al., 1998; Seghedi et al.,
525 2011). Before 3.6 Ma, most volcanoes (Fig. 10A, Fig. 10B and Fig. 10C) erupted relatively high
526 volumes (>50 km³) of basaltic andesite to andesite lavas with a typical calc-alkaline
527 composition (Seghedi et al., 2011). Their homogeneous Sr-Nd-Pb isotopic composition may

528 reflect a common mantle source (Mason et al., 1996, 1998; Seghedi et al., 2011). On the
529 contrary, South Harghita volcanoes of the 3.6–0.03 Ma period (Fig. 10A and Fig. 10D) are
530 smaller (<30 km³) and show different chemical composition with progressively increasing K-
531 content (from calc-alkaline to high-K calc-alkaline and shoshonite) and adakite-like features,
532 reflected by the increased proportions of dacites and shoshonites (Szakács et al., 1993;
533 Seghedi et al., 2011).

534 The two different magma sources should have been responsible for the observed
535 morphometric dichotomy in volcano size between the northern segments (from Călimani up
536 to Luci-Lazu edifice in North Harghita) and the three last edifices (Cucu, Pilișca, Ciomadul in
537 South Harghita). Such conclusions fit with observations of Crisp (1984) and White et al.
538 (2006), who showed that more mafic magmas are more easily mobilized in the lithosphere
539 and tend to erupt higher volumes (northern CGH volcanoes) than the differentiated ones
540 (i.e. most of South Harghita).

541 Subsequent GR values computed at segment-scale (234, 276, 116 and 39 km³/Ma from
542 north to south segments, Table 2) as well as values at volcano-scale (>100 km³/Ma until 3.6
543 Ma and <50 km³/Ma after) clearly support the decrease of volcanic intensity along the range,
544 as previously identified by Szakács et al. (1995, 2018), and the effect of different magma
545 sources (Seghedi et al., 2011). Moreover, Bouguer and magnetic anomalies surveys (Seghedi
546 et al., 2019) revealed positive residual anomaly in the northern and axial part of the range,
547 indicating the presence of high-density subvolcanic bodies which may be correlated to the
548 high eruptive volumes computed in those segments. Contrarily, in the Southern Harghita,
549 residual magnetic anomalies are negative, indicating the presence of low-density structures
550 in coherence with the small volumes of volcanoes.

551 In Gurghiu segment, the main magmatic pulse of activity observed around 6.6 Ma (Fig.
552 10A) can be plausibly related to the migration of the subducted slab through the magma
553 generating zone in the mantle and the initiation of the progressive detachment of the slab
554 following an oblique subduction stage (Mason et al. 1998; Seghedi et al. 1998, 2011). This
555 main pulse temporally coincides with an E–W compressional event that occurred across the
556 entire Carpatho-Pannonian region during the late Miocene, interpreted as reflecting the
557 arrival of thick and buoyant continental crust at the trench and its resistance to subduction
558 (Peresson and Decker, 1997). Moreover, Gurghiu zone is mostly located at the intersection
559 of regional tectonic elements with normal faults associated with paleo-basins (Seghedi et al.,
560 2019). Interaction of tectonic structures would have created an area of low resistance,
561 facilitating the penetration of magma and development of its ascent paths, thereby allowing
562 rapid magma output and construction of large volcanic centres in this segment. Moreover,
563 the largest positive magnetic anomaly identified under Gurghiu segment (Seghedi et al.,
564 2019) was suggested to have been the solidified plumbing system responsible for generating
565 the Seaca-Tătarca and Şumuleu volcanoes. Under a collisional regime, the progressive slab
566 detachment toward the south (Fig. 10D) may have triggered gradual migration of magma
567 production in South Harghita, along with emplacement of adjacent or overlapping volcanic
568 structures (Mason et al., 1998; Seghedi et al., 2005b).

569 A decreasing volcano volume towards southeast (Szakács and Seghedi, 1995) (Fig. 4A;
570 Table 2) can also be explained by the progressive opening of the lithospheric tear during slab
571 steepening (Mason et al., 1998; Seghedi et al., 1998, 2011). Finally, during the final stages of
572 subduction, a decrease in the convergence rate in the extreme south of the CGH (Fig. 10D)
573 was evidenced (Seghedi et al., 2011), possibly also responsible for the smaller dimensions of
574 the last three volcanoes.

575 The two Plio-Quaternary quiescence periods (3.6–2.8 and 1.5–0.9 Ma, Fig. 10A-D) were
576 interpreted as linked to changes in magma sources by Seghedi et al. (2011). Interestingly, the
577 1.5–0.9 Ma time gap is a period when K-alkalic and Na-alkalic volcanism developed in other
578 areas (shoshonites at Bixad/Malnaş and alkali-basalts in Perşani Mountains; Downes et al.,
579 1995; Panaiotu et al., 2004).

580 Growth rate can be used as a good proxy for revealing the dynamism, fluctuation and
581 intensity of volcanism. Globally, long-term eruption rates for silicic volcanism range from
582 $<10^{-5}$ km³/ka to 10¹ km³/ka, the average being 2.3 ± 0.8 km³/ka for andesitic systems (Vidal
583 and Bonneville, 2004; White et al., 2006). Plotting CGH growth rates results (Fig. 11) in the
584 volcano volume versus lifespan graph of White et al. (2006) highlights two distinct magma
585 output trends. The first group (most edifices of the three northernmost segments, plus Luci-
586 Lazu volcano, blue symbols in Fig. 11) shows an average GR of 137 km³/Ma. The second one
587 (the three youngest Pliocene-Quaternary edifices of South Harghita plus a small Late
588 Miocene edifice in Gurghiu, red symbols in Fig. 11) is characterized by five times lower GR
589 (28 km³/Ma).

590 Seaca-Tătarca volcano with the highest GR (825 km³/Ma, yellow symbol in Fig. 11)
591 remains isolated from these two trends. The exceptionally high GR value does not only
592 originate from the high precision on dating results, which allows drastically refining its
593 lifespan (340 ka). Indeed, even considering the typical 1 Ma-long lifespan computed for most
594 northern CGH volcanoes, its GR would remain two times higher than the given general trend.
595 This implies both an exceptionally intense dynamism of Seaca-Tătarca activity and the call
596 for getting more accurate age results for other volcanoes, since the lifespan of most CGH
597 volcanoes probably could be in the same order of magnitude.

598 Over the entire CGH range, the calculated average GR is 200 km³/Ma (ratio between
599 total volume emitted by all volcanoes during the whole range lifespan ~11Ma, without
600 consideration of the repose periods), slightly larger than previously proposed 152-164
601 km³/Ma range (Szakács et al., 1997; Karátson and Timár, 2005). Even if fluctuations in CGH
602 growth rate occurred through time (10-825 km³/Ma), values remain low compared to values
603 for andesitic volcanoes (blue squares in Fig. 11 background) from other subduction-related
604 arcs (White et al., 2006). It testifies to the less intense magma generation processes in the
605 long term here than in areas where subduction is more intense, such as the West Indies,
606 Japan and the Andes (White et al., 2006). Again, higher-precision dating may result in more
607 intense GR at the lifetime scale of individual edifices as is the case with Seaca-Tătarca. Such a
608 finding matches with worldwide (real, short-term) GR values of composite cones such as at
609 Parinacota volcano, Chile (Hora et al., 2007).

610 **6.5 Erosion rate and degree of denudation through time**

611 Summing eroded volumes of all CGH volcanoes gives a total of 524±125 km³ (Table 2)
612 and an average D_{Den} of 22%, close to the 23% proposed by Karátson and Timár (2005). At
613 segment-scale, only the inferred eroded volume for Călimani segment (265±105 km³) does
614 not match at 1 σ with the previous results of Karátson and Timár (2005), the new value being
615 three times larger. The discrepancy, as presented above, rise from the difference in
616 estimation of pre-erosional volumes (V_C) that directly impact on the estimation of eroded
617 volumes.

618 At volcano-scale, the V_E vs V_C plot can be interpreted with respect to D_{Den} values as it is
619 the ratio between these two parameters (Fig. Fig. 5B). Volcanoes with different volumes but
620 experiencing the same D_{Den} rate (i.e. build at the more and less same period) would follow a
621 linear correlation with constant slope (dashed lines in Fig. Fig. 5B). South Harghita volcanoes

622 built during Plio-Quaternary present the lowest D_{Den} values (~17%); average D_{Den} reaches
623 20% for 5.1-6.8 Ma old volcanoes and 27% for the older ones (>7.1 Ma; Călimani and
624 northern volcanoes). On this graph, the vertical black arrows represent the additional of
625 erosion affecting the older volcanoes and relatively to the average behaviour of the younger
626 ones. This additional erosion increases with the volume of the volcano (bigger volcanoes
627 produced more eroded material). However, in CGH range, larger volcanoes are also mostly
628 the older ones, another parameter positively correlated to the V_E values (older volcanoes
629 suffered longer erosion leading to larger eroded volume). Therefore, as volcano size (V_C) and
630 age are globally positively correlated (Fig. 10A), these parameters sum these effects on V_E
631 behaviour ($V_E=F(V_C)+G(Age)$). Nonetheless, even if the amount of erosion (V_E) seems to be
632 nicely correlated to V_C , it is less relevant than the correlation between D_{Den} and the time
633 (dashed line in Fig. 5B). By averaging D_{Den} values (Table 2 and black dots in Fig. 12) on each
634 CGH segment leads to the following D_{Den} values: 30%, 25%, 20% and 15% (bottom of Table
635 2), confirming the progressive degradation of volcanoes toward north, as first quantified by
636 (Karátson, 1996).

637 Correlation of the D_{Den} versus time allows establishing relationships between erosion
638 rate (ER, i.e. aerial denudation or lowering of edifice height) and time. To better constrain
639 the fast evolution of D_{Den} for the early stage of dismantling, we added to the CGH volcanoes
640 dataset (this work, black dots in Fig. 12) the values proposed for the dozen edifices of the
641 Ciomadul volcanic complex dating to between 0.03 and 0.8 Ma (Karátson et al., 2019; Lahitte
642 et al., 2019, deduced D_{Den} summarised in supplementary material, grey dots in Fig. 12).

643 In the case of dismantling evolving through time according to a single behaviour, the
644 trend of D_{Den} would evolve from 0 at $t=0$ (no erosion right after the volcano construction) to
645 100% after an infinite duration (with any volcano completely eroded). In order to verify D_{Den}

646 (0) = 0, the best function to fit such behaviour is a reverse exponential function ($D_{Den}(t)=1 -$
647 $k \cdot e^{-k \cdot t}$), with $k=1$.

648 However, considering the exhaustive data set of denudation rate (0 to 11 Ma of
649 denudation duration) leads to a best fit function with $k \sim 0.9$ that intersects the D_{Den} axis at
650 10% (dash blue line and y axis in Fig. 12). This means that a perfectly newly constructed
651 volcano would have already lost 10% by dismantling right after its construction. This leads to
652 an improvement of the D_{Den} behaviour, with the consideration of a more complex
653 dismantling history, during the first 1 Ma of the erosion, when erosion occurred
654 simultaneously to (and competed with) the growing volcano and just after the end of its
655 construction. It is highlighted by the fast denudation rates affecting the domes of the
656 Ciomadul complex (grey dots in Fig. 12), “instantaneously” erupted along the past 1 Ma. The
657 good correlation between their age (i.e. erosion lifespan) and D_{Den} values supports
658 interpretation of D_{Den} changes through time as better approximated by two distinct trends.
659 First, a very fast evolving trend in the early stage with D_{Den} evolving from 0 to 14% during
660 about the first 1 Ma (grey dots in Fig. 12). This trend also incorporates the proportion of the
661 volcano dismantled during its 0.4-1 Ma-long construction phase (with an average proportion
662 of 1% lost by 0.1 Ma-long lifespan). Then, a long-term (constrained by all other CGH
663 volcanoes) trend fitted by the following equation (inset Fig. 12; see supplementary material
664 for the detail of calculation):

665
$$D_{Den} = 1 - 0.89e^{-0.024t} \quad (R^2 = 0.62) \quad \text{with } t \text{ in Ma} \quad \text{Eq. 4}$$

666 Namely, the first stage of erosion (the first hundred thousand years [0 - <0.4 Ma] and,
667 subsequently, with less intensity up to 0.8 Ma) is much more intense due to the presence of
668 unafforested slopes, unconsolidated material (and possibly the erosion pattern inherited

669 from the activity period of the volcano) which result in high erosion rates. It is followed by a
670 second slowly decelerating long-term trend that can be considered as steady-state erosion in
671 the <20 Ma window.

672 Reciprocally, the averaged through time erosion rate (red and orange dots in Fig. 12) is
673 fitted by a quickly decreasing function in the early stage, followed by a slowly decreasing
674 rate (red curve in Fig. 12) with $R^2=0.72$. Both cumulated ER and D_{Den} parameters support a
675 general behaviour that is time dependent.

676 In conclusion, our results fit with the already observed (Ruxton and McDougall, 1967;
677 Francis, 1983; Karátson, 1996; Karátson et al., 2012) changes in volcano degradation: a
678 short-term post-volcanic erosion (<0.8 Ma) was characterized by a rapid lowering and slope
679 dissection/valley formation of the edifices with relatively high ER (24-39m/Ma, Fig. 12),
680 followed by a drastic decrease until reaching an equilibrium stage, where erosion rate slowly
681 decrease as mainly operating through a well-established drainage network (Ollier and
682 Brown, 1971; Karátson, 1996; Karátson et al., 2012) (shown by the red curve based on
683 orange and red dots in Fig. 12).

684 On the other hand, within the long-term erosion, there are obvious changes in ER values
685 which can be linked to past climate variability. A crucial record for disentangling
686 hydrographic evolution in the Black Sea–Carpathian area from Late Neogene has been
687 provided by the marine core DSDP Site 380A (Leg 42B; Popescu, 2006). This record, along
688 with the long-term trend in the palynological steppe index curve (Traverse, 1978) which is a
689 proxy representative of past climate changes between cooler and warmer phases (Suc,
690 1984), indicates a continuously decreasing trend in warm and wet taxa for the Carpathian
691 area throughout Late Neogene and a strong variability during the Quaternary (Popescu,
692 2002, 2006).

693 However, ER is expected to depend on several factors in addition to climate conditions
694 such as lithology or edifice topography such as local slope (e.g. Cotton, 1952; Ollier and
695 Brown, 1971; Francis, 1983; Karátson et al., 1999; Thouret, 1999; Reiners et al., 2003). Again,
696 Fig. 6A clearly shows that morphometric parameters (e.g. height, area, volume) do not
697 control denudation; therefore, erosion of CGH volcanoes is mostly controlled by
698 hydroclimate variability and/or slope characteristics. For instance, Champagnac et al. (2014)
699 showed, based on mountainous relief with 10-35° slope, that the behaviour followed by
700 erosion over time can be approximated by a quadratic relationship. CGH volcanoes, with
701 typically 10-21° slopes (Karátson, 2007), fit well with this observation. Therefore, since
702 volcanoes in CGH present relatively uniform and moderate slopes and are largely dominated
703 by andesite composite volcano types, we suggest that the contrasts in erosion have been
704 mostly driven by past hydroclimate fluctuations with the relatively small impact of
705 topography and rock strength.

706 From the integrated erosion rate (ER) at segment-scale (17, 11, 9, 28 m/Ma in Călimani,
707 Gurghiu, North Harghita and South Harghita, respectively, Table 2), it is possible to infer the
708 ER that the main specific time intervals the CGH range experienced (Karátson, 1996;
709 Popescu, 2002, 2006; Erdei et al., 2007): 1) a short time period of subtropical-moderate
710 continental transitional climate (Late Miocene, ~9.5-8.2 Ma); 2) a moderate continental
711 climate with intervening semiarid periods (Late Miocene up to Pleistocene, ~8.2-4 Ma); and
712 3) a humid continental climate with periglacial regimes during the Pleistocene glaciations
713 (Late Pliocene-Quaternary, from ~3.0 Ma up to present). Knowing the cumulated ER of the
714 period t_1 and t_1+t_2 , we computed the specific ER driving dismantling during the period t_2 that
715 leads to the cumulative ER of the period t_1+t_2 . For instance, the cumulated ER for North
716 Harghita equals the erosion regime that specifically controlled the dismantling of North

717 Harghita before the construction of South Harghita plus the erosion regime during the
718 dismantling of South Harghita. Obviously, the ER calculated for a certain period allows
719 inferring the behaviour of erosion in response to prevailing climatic conditions.

720 This approach leads us to infer four successive erosion regimes showing a strong
721 fluctuation over time (Table 3 and large diamonds in Fig. 12). 1) The highest ER (38 m/Ma,
722 for the 10.8-6.9 Ma period, red diamond), obtained for the construction period of Călimani
723 (Fig. 12), fits with the influence of the transitional moderate subtropical-continental climate
724 period as defined above (Karátson, 1996; Popescu, 2002, 2006). 2) Decreasing ER for Gurghiu
725 (14 m/Ma, 8.7-5.5 Ma, green diamond) and North Harghita (7 m/Ma, 7.0-4.6 Ma, blue
726 diamond) accords with the subsequent, drastically decreased erosional processes
727 characterizing a moderate continental climate. The extremely low value of 7 m/Ma, pointed
728 out for Late Miocene to Pliocene times, can be explained by occasional semi-arid conditions
729 (Lisiecki and Raymo, 2005). Indeed, Pliocene was already characterized by long-term shifts
730 between the humid forest biotopes and run-off and the dry steppes, with changes linked to
731 large amplitude climatic and moisture variations (Suc, 1984; Popescu, 2002, 2006). 4) Finally,
732 the 28 m/Ma ER obtained for the Plio-Quaternary period (<3 Ma, yellow diamond) can
733 reflect the prevailing more humid conditions (Suc, 1984; Karátson, 1996; Popescu, 2002,
734 2006); Schreiber and Unger, 2006) with a minor impact of glacial erosive activity and
735 periglacial regime.

736 These results of erosion rate specific to the main time intervals analysed are following
737 reconstructions of variability in sea levels and progressively stronger land glaciations from
738 Late Neogene to recent times (Lisiecki and Raymo, 2005). These are correlated to a shift
739 from prevalent chemical weathering during Late Neogene to enhanced surface processes,
740 run-off and physical weathering prominent during the Quaternary (Gibbard and Lewin,

741 2009). The latter reflect rapid and high-amplitude successive hydroclimate variability
742 between humid and glacial periods (colder and generally drier periglacial conditions) and
743 shorter, wetter and warmer interglacials (Lisiecki and Raymo, 2005).

744 For the entire range, average ER is ~ 20 m/Ma, lower than the 31.5 m/Ma proposed by
745 Karátson (1996). The latter value was calculated for the highest crater-topped summit
746 regions only and consequently overestimated the average erosion rate over the whole area
747 as omitting the peripheral aprons affected by lower ER. Our calculated ER for the highest
748 volcanoes (Central Călimani, Fâncel-Lăpușna, Seaca-Tătarca, Vârghiș and Șumuleu) give high
749 values (>14 m/Ma), whereas smallest edifices (Jirca, Bacta, Borzont, Râchitis, Pilișca) show
750 the lowest values (<11 m/Ma, Table 3), in accordance with these ages, the most elevated
751 (and larger in size) volcanoes are also the older ones.

752 Relying on inferences of past climatic variability for the broader Carpathian region and
753 experienced by the CGH range throughout its evolution (Traverse, 1978; Suc, 1984; Karátson,
754 1996; Popescu, 2002, 2006; Erdei et al., 2007), we suggest that the best way to express ER
755 variations is to detail them at the scale of an individual segment. Indeed, work at the scale of
756 the whole CGH would only propose a regional value without consideration of variations
757 through time, whereas work at the scale of a volcanic edifice would introduce local bias due
758 to the own history of construction/dismantling that each volcano experienced (even if the 16
759 ERs define a general trend).

760 Globally, few studies have investigated ER over a long-term period, hence the
761 relationship with the geodynamic context or past climate variability is still poorly understood
762 (Karátson and Timár, 2005; Germa et al., 2010; Karátson et al., 2012a; Smith et al., 2016).
763 Studies in tropical environments pointed out much higher ER (Louvât and Allègre, 1997;
764 Hildenbrand et al., 2008; Germa et al., 2010; Ricci et al., 2015b). The systematically higher ER

765 values support the dominant role of climate as a main factor during dismantling. Moreover,
766 differences in prevailing lithology can also affect the behaviour of ER. For example, basaltic
767 rocks experience a faster weathering compared to andesitic ones and hence show higher ER
768 (Gislason et al., 1996; Louvat and Allègre, 1997).

769 The termination of erosional processes on volcanic landforms is the complete
770 destruction of the volcano with a remnant morphology of necks and dykes (i.e. skeleton
771 stage: Cotton, 1952) and a D_{Den} reaching almost 100% (inset of Fig. 12). It defines a very
772 long-term stage (tens to hundreds of Ma) when the volcano is very slowly dismantled as ER
773 tend simultaneously to zero. The equation fitting D_{Den} long-term evolution (Eq. 4) leads to a
774 duration required to destroy 95% of the entire range of 120 -20/+50 Ma. Even if such
775 duration would be significantly modified by changes in the tectonic (e.g. uplift) and/or
776 climatic contexts (Peulvast and Claudino Sales, 2005), its order of magnitude is reliable given
777 some known relict morphologies that testify to Cretaceous or even older Mesozoic
778 volcanism (Klein, 1975; Sturt et al., 1979; Simon-Coinçon, 1989; Slingerland and Furlong,
779 1989).

780 **7 Conclusions**

781 From the modelled volcanic surfaces performed in this study, the constrained
782 constructed volumes allow classifying the CGH volcanoes in the field of volcanic massif,
783 which is in accordance with their topographic complexities and eruptive lifespan, as pointed
784 out by new ages of the Seaca-Tătarca (~340 ka) and Vârghiș (~900 ka) edifices. The 1/3-
785 exponent in the power-law function relying original heights and volume advocates for CGH
786 volcanoes as being self-similar, i.e. exhibiting homomorphic shapes irrespective of the
787 peculiar complexities of their constructional histories. The overall volume computed for the

788 entire CGH is $\sim 2200 \text{ km}^3$, characterized by decreasing individual volumes from north to
789 south, matches the evolution in terms of influence from different magma sources and plate
790 tectonics. Individual growth rates per volcano distinguish a first moderate regime (137
791 km^3/Ma) for the majority of volcanoes (11-3.6 Ma), followed by a drastically decreased
792 output rate ($28 \text{ km}^3/\text{Ma}$) for the mostly Plio-Quaternary volcanoes. Apart from these two
793 trends (representing the average of individual growth rate per volcano), the much higher GR
794 obtained for Seaca-Tătarca ($825 \text{ km}^3/\text{Ma}$) is the consequence of probably both its shorter
795 activity (inferred from new K-Ar datings) and a stronger magmatic regime. However, this
796 finding also points out the poorly constrained duration of volcanic activity (and related
797 output rates) if computed from poorly constrained ages in the case of CGH volcanoes for
798 which modern standard dating is not yet available. Fluctuations of GR (relatively to the
799 moderate overall averaged value of $200 \text{ km}^3/\text{Ma}$) is interpreted as the manifestation of
800 strong fluctuations of volcanic activity driven by drastic changes in subduction mechanisms
801 with decrease of convergence rate during the Plio-Quaternary causing breakoff beneath
802 South Harghita and subsequent upwelling of asthenospheric mantle.

803 Beyond the average values of dismantling parameters (total eroded volume of 524 ± 125
804 km^3 , averaged denudation and erosion rate of 22% and $20 \text{ m}/\text{Ma}$, respectively), the
805 correlation established between denudation and time defines two contrasting intensities of
806 dismantling processes given their position along the erosional timeline. The first denudation
807 trend shows an intense and rapid denudation ($< 0.8 \text{ Ma}$) after the cessation of volcanic
808 activity while the second long-term trend slows down over the remaining erosional lifespan.
809 Whereas this behaviour implies an almost constant erosion rate during the lifespan of the
810 CGH erosion, detailed erosion rate values computed for shorter time intervals evidence the
811 variety of intensity of erosion having occurred through time. Since volcanic edifices present

812 relatively uniform slopes and composition, these second order variations suggest that
813 contrasts of ER along CGH range were mostly driven by climate fluctuations through time:
814 highest ER (38 m/Ma) during the transitional moderate subtropical-continental climate
815 period; intermediate ER (14 m/Ma) characterizing a moderate continental climate period
816 when conditions became less humid; extremely low ER (7 m/Ma) coeval with continental,
817 but occasionally semi-arid, climate; and, finally, higher ER (28 m/Ma) when Plio-Quaternary
818 experienced generally wetter conditions with alternating glacial and interglacial stages.

819 The presented quantitative morphometric approach of paleo-volcanic landforms,
820 performed at the detailed scale of individual volcanoes, demonstrates the possibility of a
821 wide range of applications in various geological domains. They allow constraining
822 constructive processes of volcano-tectonic dynamism at the scales of both individual
823 volcanoes and global geodynamic context. Furthermore, by correlating eroded volumes and
824 climatic variations, the intensity of the processes driving the dismantling of the volcanic
825 range can also be well assessed.

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1103 **Figure Captions**

1104 **Fig. 1:** Geological setting of the studied area (East Carpathian Volcanic Range). Each volcano
1105 is labelled as following (Romanian names, Hungarian ones can be found in Table 1): Călimani
1106 sector (surrounded by red outline): 01 – Central Călimani including Rusca-Tihu cones
1107 Lucaciul, Tămăul and Călimani caldera; 02 – Moldovanul. Gurghiu sector (green outline): 03 -
1108 Jirca; 04 – Fâncel-Lăpușna, including cones Fâncel (04a) and Obarsia (04b); 05 –Bacta; 06 –
1109 Seaca-Tătarca; 07 – Ciumani; 08 – Borzont; 09 – Șumuleu including cone Șumuleu (09a) and
1110 Vf. Ascuțit (09b). North Harghita sector (blue outline): 10 – Ostoros; 11 – Ivo-Cocoizaș; 12 –
1111 Râchitis; 13a – Vârghiș and 13b – Szeles Vésze cone. South Harghita sector (yellow outline):
1112 14 – Luci-Lazu (14a and 14b); 15 – Cucu; 16 – Pilișca; 17 –Ciomadul lavas domes. Inset:
1113 Regional topographic sketch map of Carpathian orogen.

1114 **Fig. 2 :** A) Constraining points (CPs) of each single volcanic edifice. New unspiked K/Ar ages
1115 (in Ma): A) Călimani. B) Seaca-Tătarca volcano. C) Vârghiș volcano. D) Pilișca volcano.

1116 **Fig. 3 :** Modelling of the best fitting surface with all relevant parameters, example of Seaca-
1117 Tătarca volcano: A) Elevation versus distance to centre diagram, green dots project the
1118 whole volcano surface, purple dots are CPs and black dots are the best model profile. B)
1119 Projection of mean (orange), max (yellow) and min (grey) elevation of the whole volcano
1120 surface, after a summarization by a 100m-long window. Note how the best model profile
1121 (black dot) fit well with the upper surface (yellow dots). Average altitude loss (blue dots)
1122 computed as the difference between the best model profile and the mean elevation profile.
1123 C) Perspective view of the best modelled surface: purple dots are the CPs extracted from the

1124 DEM (coloured surface). Coloured dots show the best modelled surface. Black sticks
1125 highlight the loss of elevation (i.e. the intensity of erosion).

1126 **Fig. 4** : Modelled paleo-volcanic surfaces (as they would be directly after the cessation of
1127 volcanism, i.e. before erosion effect): A) North (red colours) - South (yellow colours) cross-
1128 section and perspective views of the whole reconstructed CGH range. B), C), D) zoomed
1129 representation of Gurghiu, North Harghita and South Harghita sectors, respectively.
1130 Numbers refer as the same labels as in Fig. 1 and Table 2. Cream coloured surface is the pre-
1131 volcanic basement surface (PVBS).

1132 **Fig. 5** : **A)** Correlation between total constructed volume with volcano area (uncertainties
1133 displayed at 1σ -level): Green line and box parameters: power-law fitting function obtained
1134 with consideration of all volcanoes. Black line and box parameters: fitting without
1135 consideration of Făncel-Lâpuşna volcano (green dot). Inset: height and volume of CGH
1136 volcanoes plotted on the height versus volume diagram of Grosse et al. (2009) with fitting
1137 power-law function. **B)** Correlation of eroded volume with constructed volume of each CGH
1138 volcano: circle diameters with respect to degree of denudation values (expressed in %).
1139 Colour of symbols, line and parameters box refer to volcano ages: older (red), intermediate
1140 (green) and youngest (yellow) volcanoes. Black line and parameters box represent the V_E - V_C
1141 correlations for all volcanoes whereas colour lines show fitting for the corresponding
1142 periods. Black arrow explained in text.

1143 **Fig. 6** : Correlation between degree of denudation of each volcano of the range with **A)** the
1144 height. **B)** the latitude. Colours symbols with respect to belonging segment (yellow: South
1145 Harghita, blue: North Harghita, green: Gurghiu, red; Călimani).

1146 **Fig. 7** : Relative uncertainty of constructed volume versus average height diagram with best
1147 fitting function. Circle diameter proportional to volcano volume.

1148 **Fig. 8** : Model of growth of Seaca-Tătarca volcano through time: **A)** Projection in elevation
1149 versus distance to centre of the whole Seaca-Tătarca volcano surface (green dots) with
1150 projection of the dated samples (white dots). Blue line displays the chronological model of
1151 growth based on the modelled uppermost volcanic surface. Note how the blue lines
1152 regularly spaced every each 10% of volume become closer upward. **B)** Proportions of
1153 constructed of Seaca-Tătarca volcano versus time diagram. White dots: growth proportion
1154 associated to each dated sample deduced from Fig. 8A, red dots represent interpolation of 0
1155 and 100% of construction (i.e. begin and end of the activity).

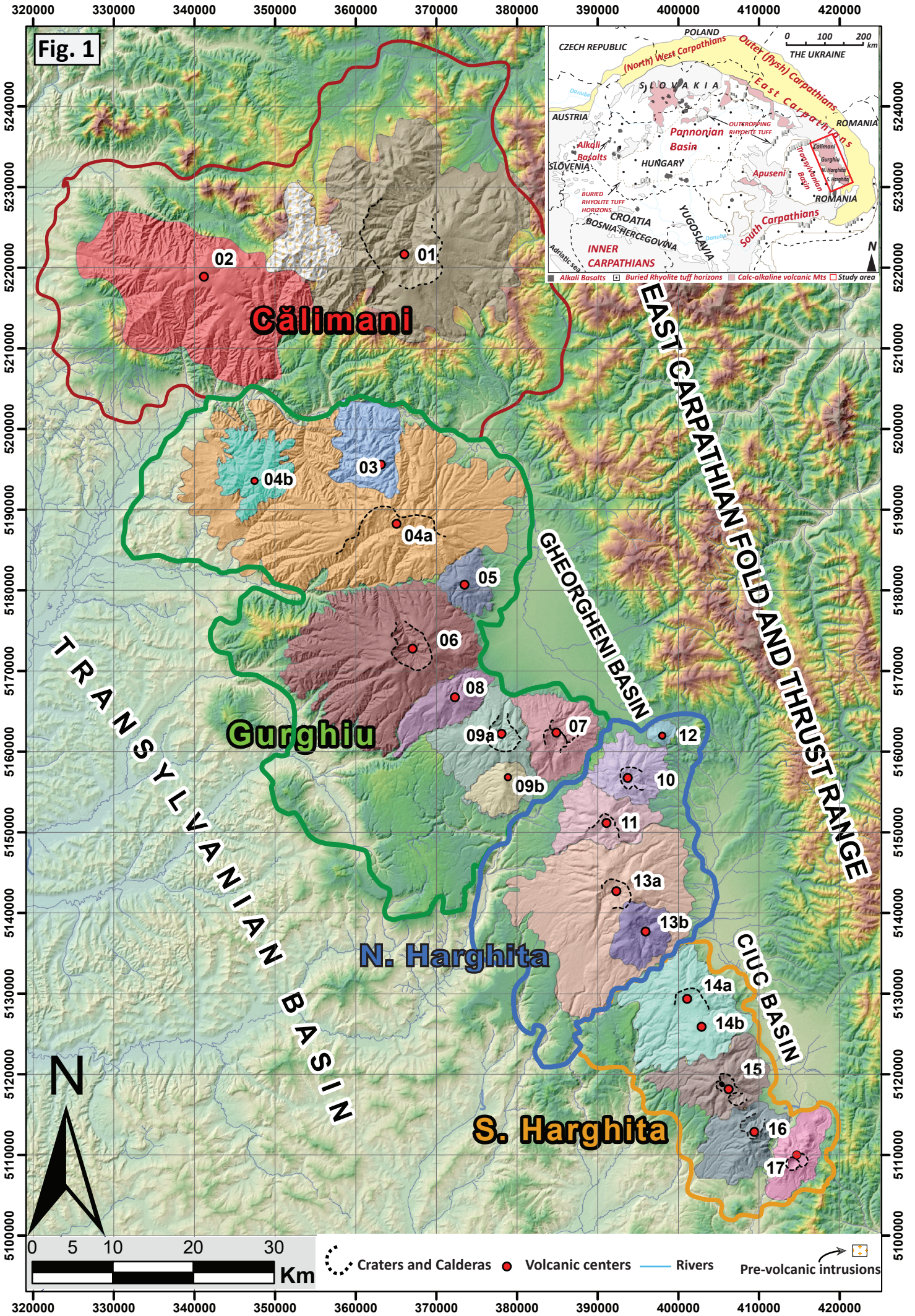
1156 **Fig. 9** : All available ages of CGH volcanic range projected along a N-S profile (Y-axis is the
1157 UTM coordinates of samples). Colours symbols as in Fig. 5 except grey symbols plotting
1158 newly dated samples. Coloured outlines display the spatio-temporal range of volcanic
1159 activity. Heavy lines show either the expansion (Gurghiu case, using only heavily outlined
1160 green symbols) or migration (N. and S. Harghita cases, using all their respective ages)
1161 through time (details in the text), with quantification in the coloured rectangles. Black
1162 double arrows highlight the synchronism in sectors.

1163 **Fig. 10** : Cumulative volume and magma output rates through time (A), correlated to the
1164 main Tertiary tectonic events affecting the East Carpathian range (displayed as cartoons B, C
1165 and D). **A)** Individual variation of volcanic output through time (coloured lines). Cumulative
1166 erupted volume (dashed line and left axis) and global growth rate (plain line and right axis)
1167 variations through time over the whole CGH range. Inset displays Fig. 10 to highlight volume
1168 – space correlations. **B)** Oceanic crust subduction occurring during the Early–Mid-Miocene

1169 convergence episode. **C)** Thinned continental crust subduction and collisional features in the
1170 overriding plate. **D)** Slab detachment of the dense oceanic lithosphere from buoyant
1171 continental lithosphere and upwelling of the hot asthenospheric mantle into the gap to be
1172 juxtaposed against the base of the mechanical lithosphere.

1173 **Fig. 11** : Volumes versus lifespan in a log-log diagram. Trends of both group (red and blue
1174 diagonal lines) as well as grey dashed lines represent constant rates of magma output.
1175 Background of the figure displaying various volcanic contexts around the world from White
1176 et al. (2006).

1177 **Fig. 12** : Graph of dismantling parameters versus time. Black and grey circles (left axis) are
1178 the computed degree of denudation values of each volcano (i.e. averaged along its whole
1179 erosion history), fitted by the reverse exponential curve (with parameters in the blue box).
1180 Grey circles are values from the dozen domes of the Ciomadul volcanic complex (Karátson et
1181 al., 2019; Lahitte et al., 2019; details in S.M.), black dots are D_{Den} values from this work (all
1182 other CGH volcanoes). Orange and red circles are the cumulative averaged erosion rate of
1183 each volcano (right axis) and fitted by the red quadratic curve. Contrarily to red/orange
1184 circles that show average-integrated erosion rates, large diamonds (right axis) represent the
1185 erosion rates computed for specific periods when specific climatic context (summarized on
1186 top of the figure) occurred: red diamond for erosion regime occurring during 10.8 - 6.9 Ma;
1187 green diamond: 8.7 - 5.5 Ma; blue diamond: 7.0-4.6 Ma; yellow diamond: Quaternary.



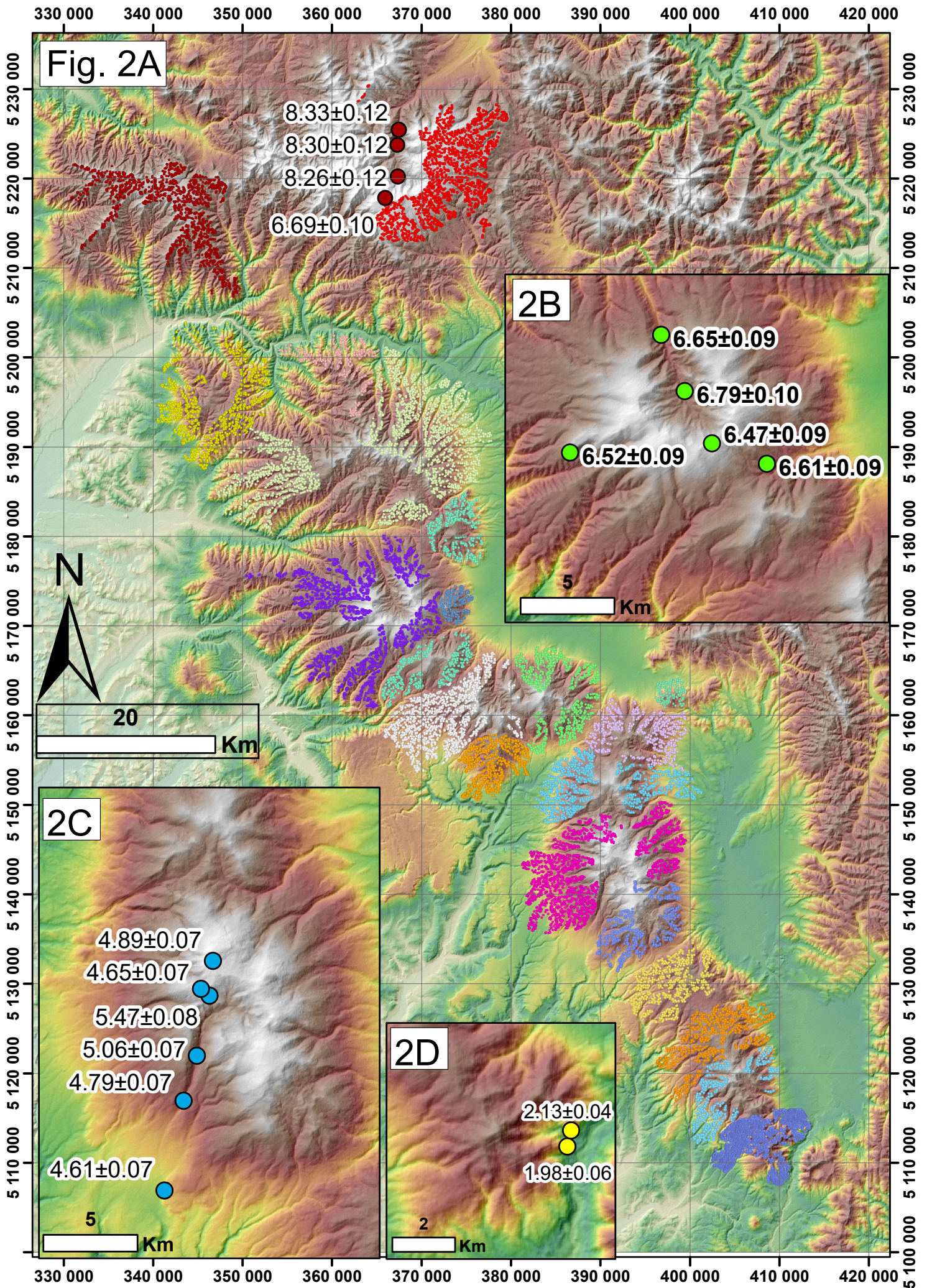


Fig. 3

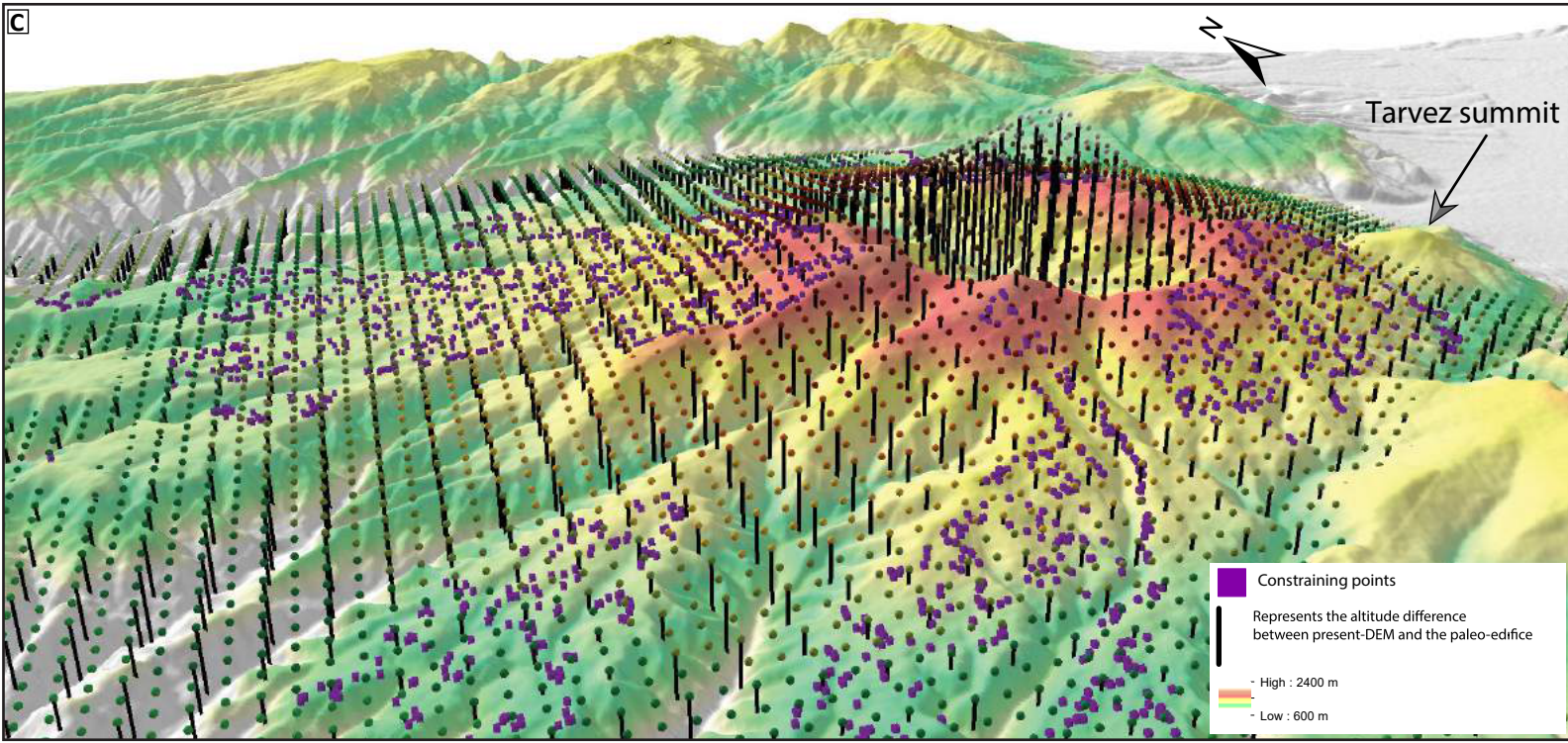
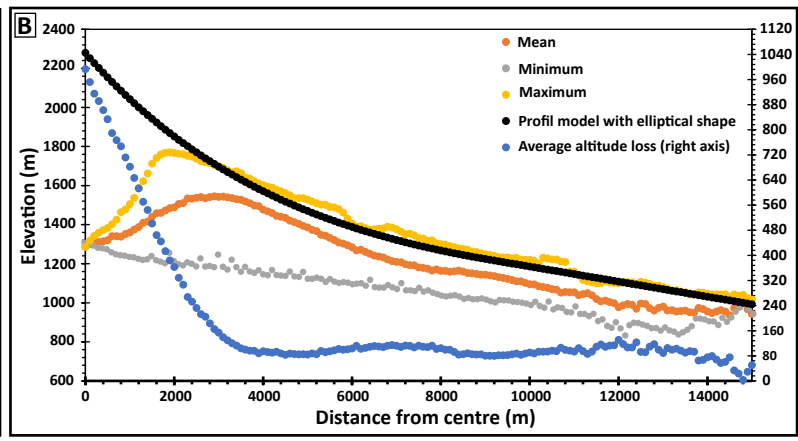
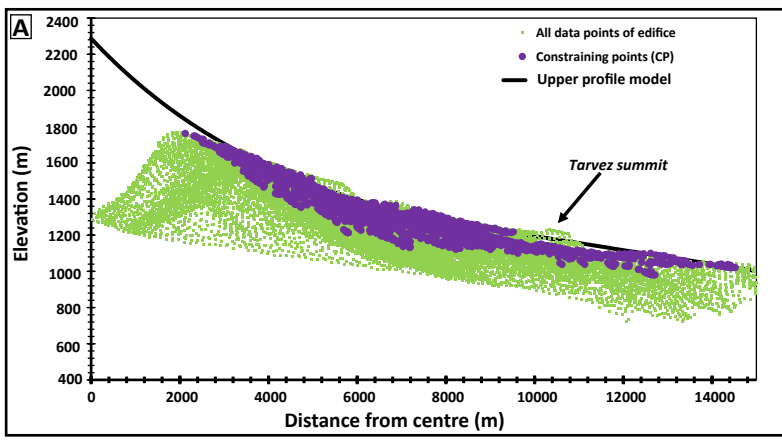


Fig. 4

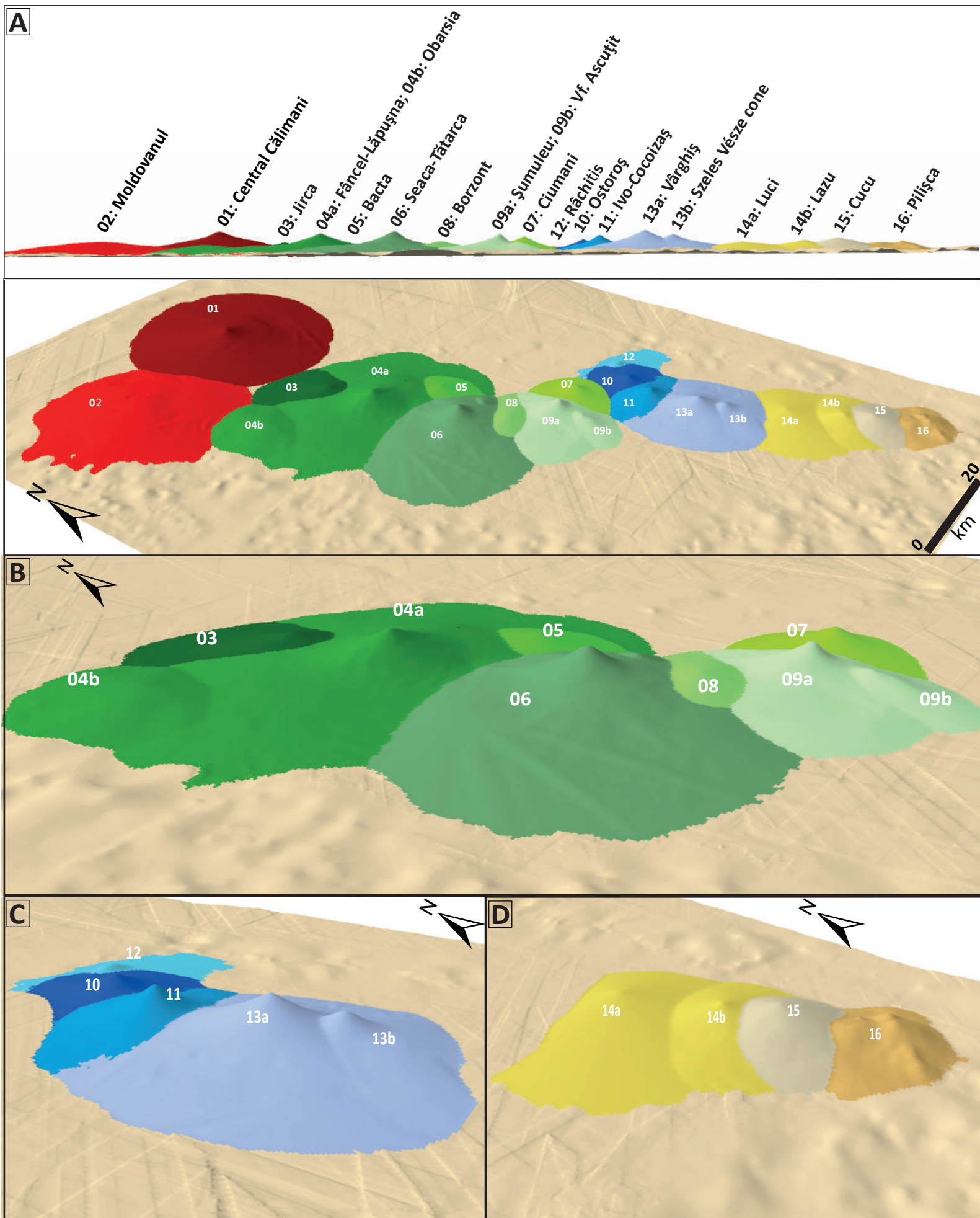


Fig. 5

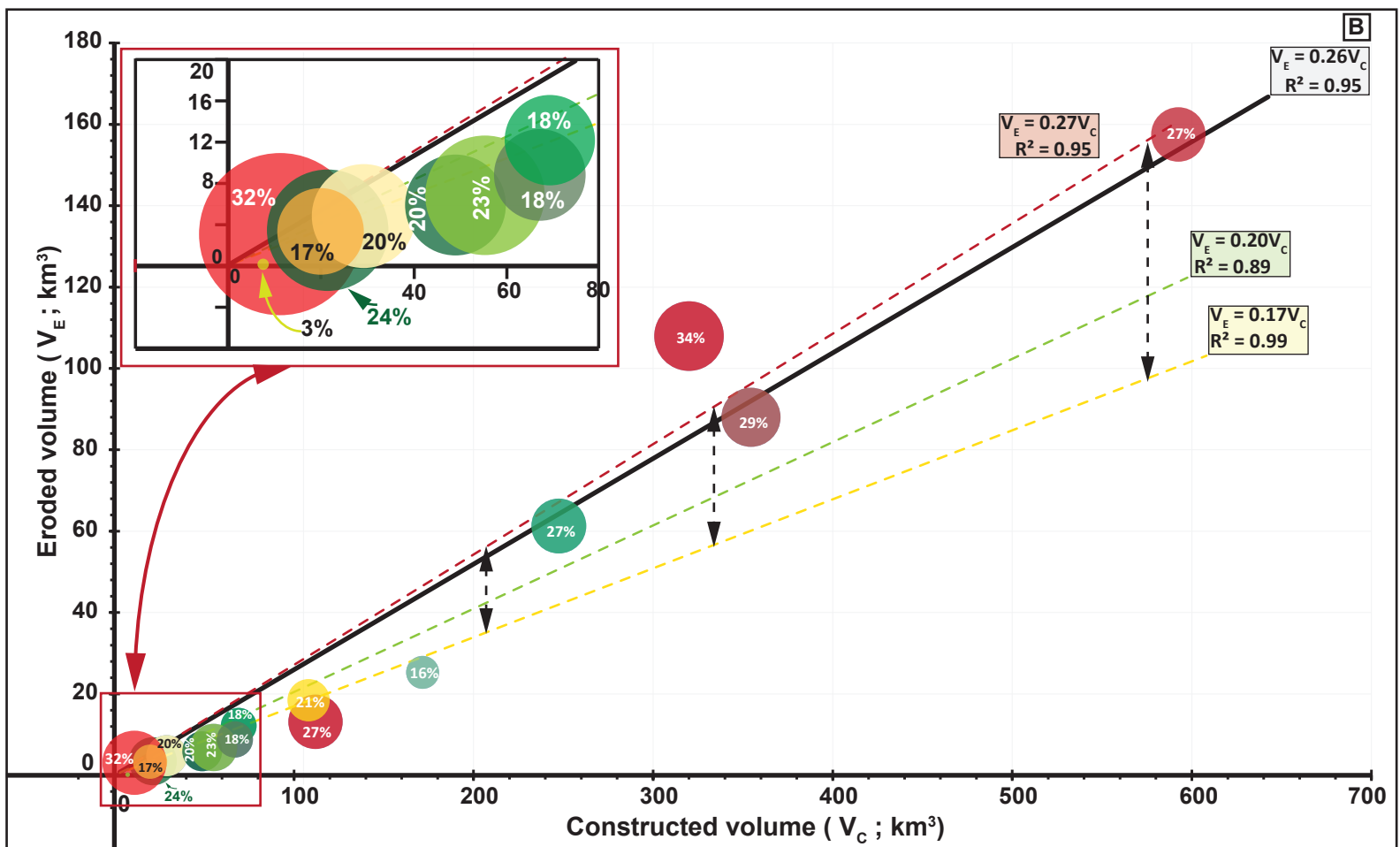
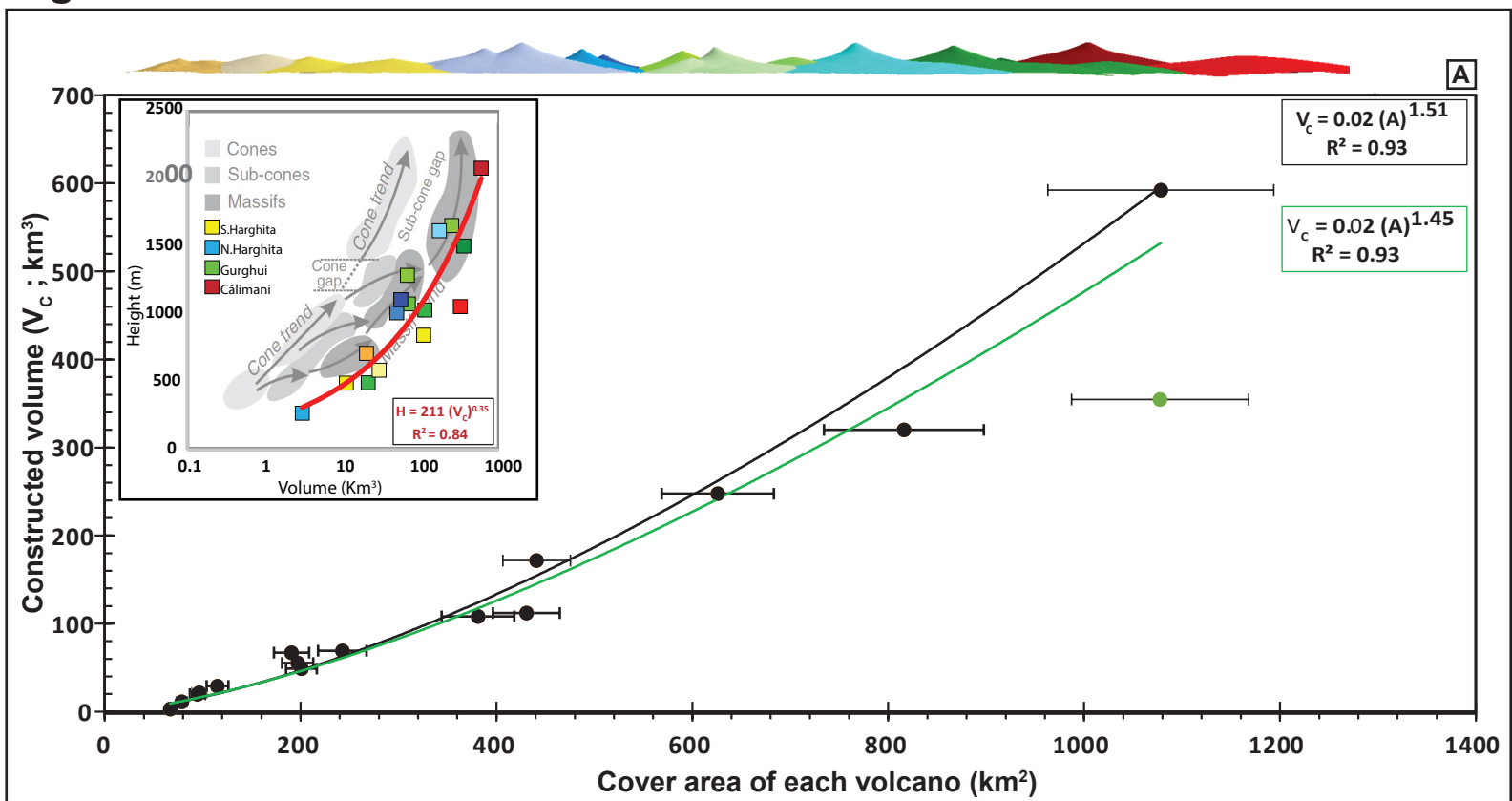


Fig. 6

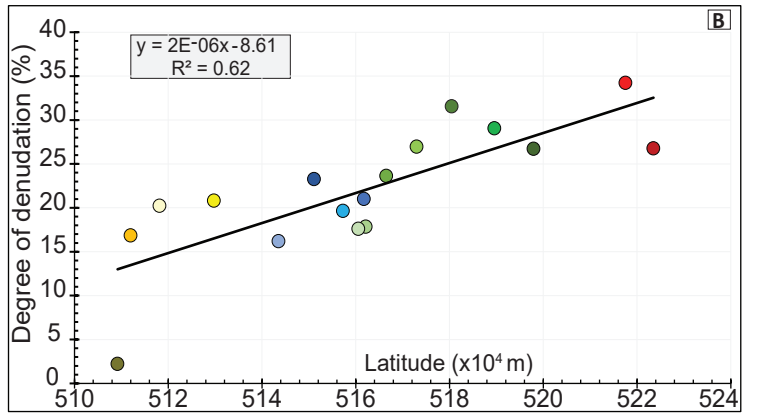
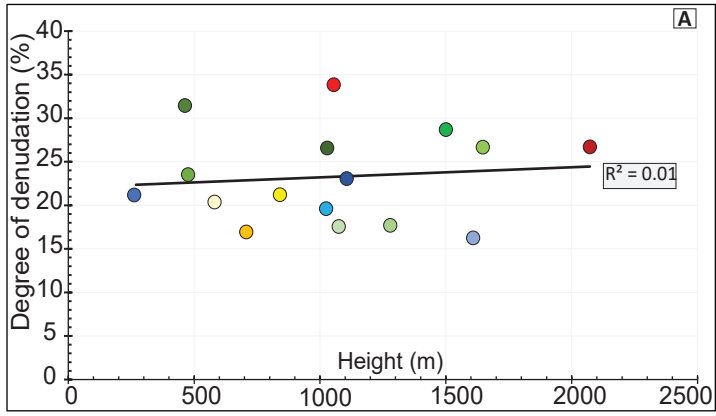


Fig. 7

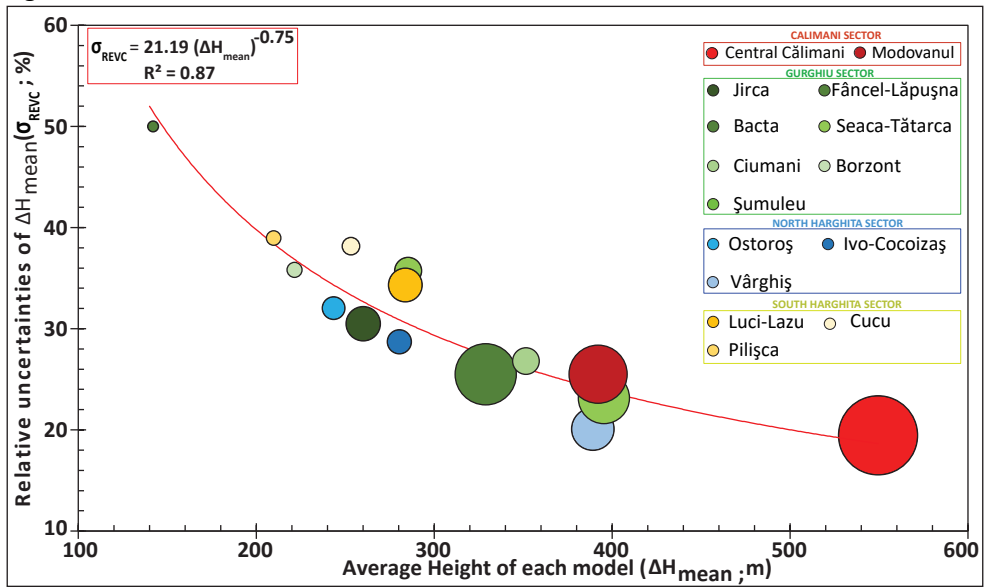


Fig. 8

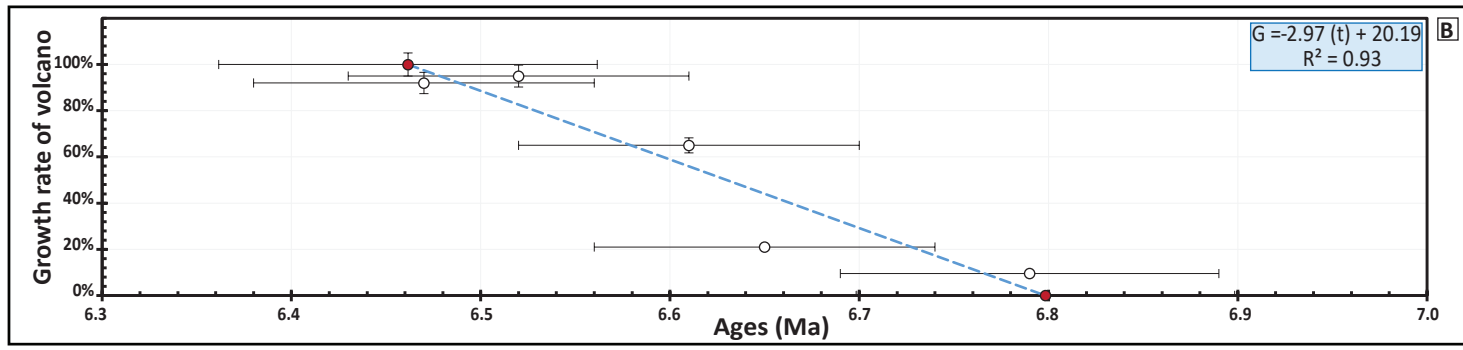
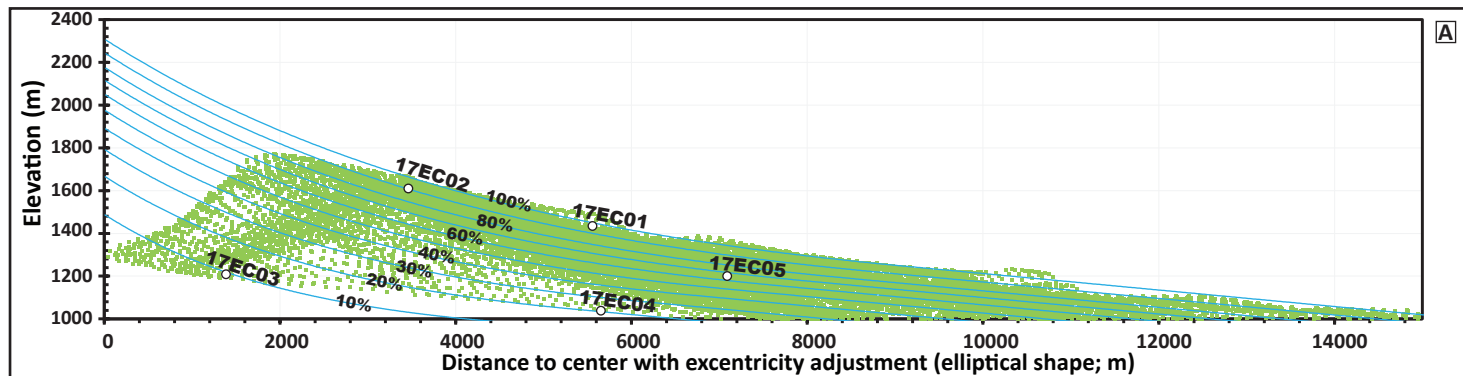


Fig. 9

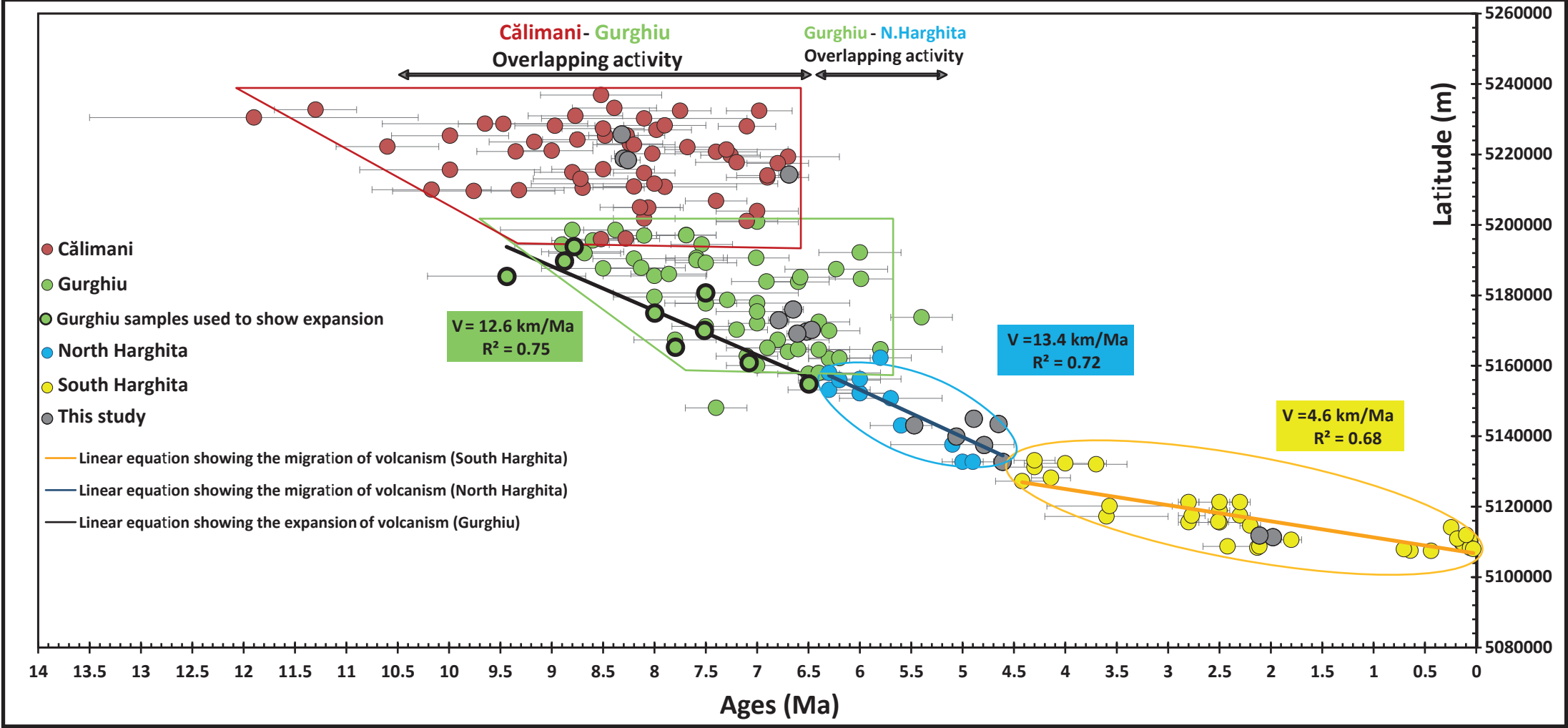


Fig.10

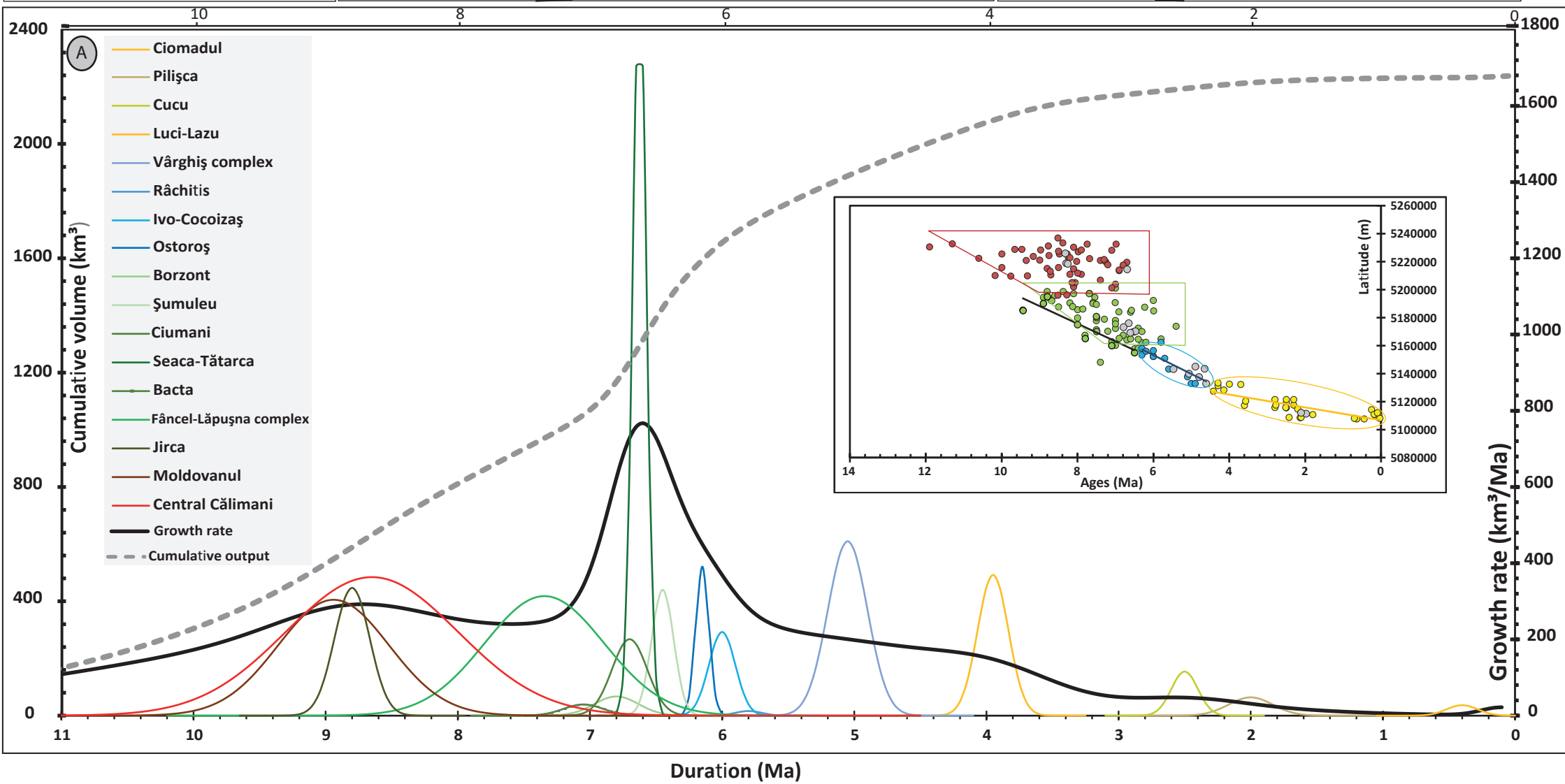
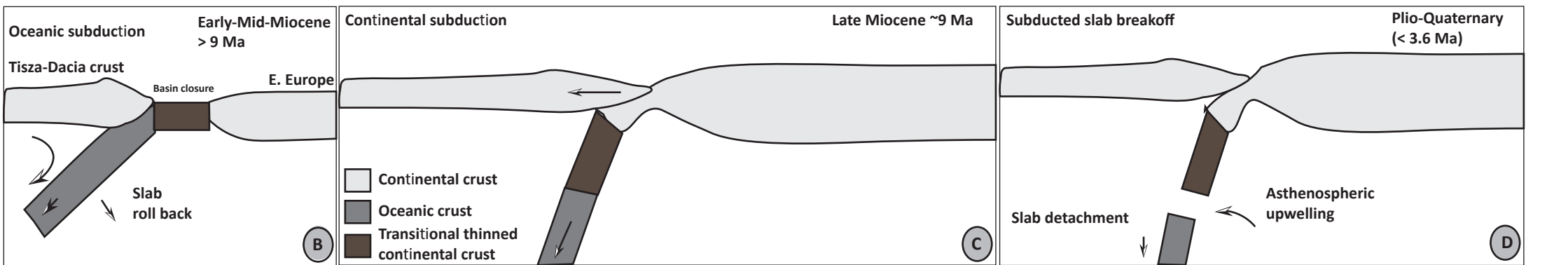


Fig. 11

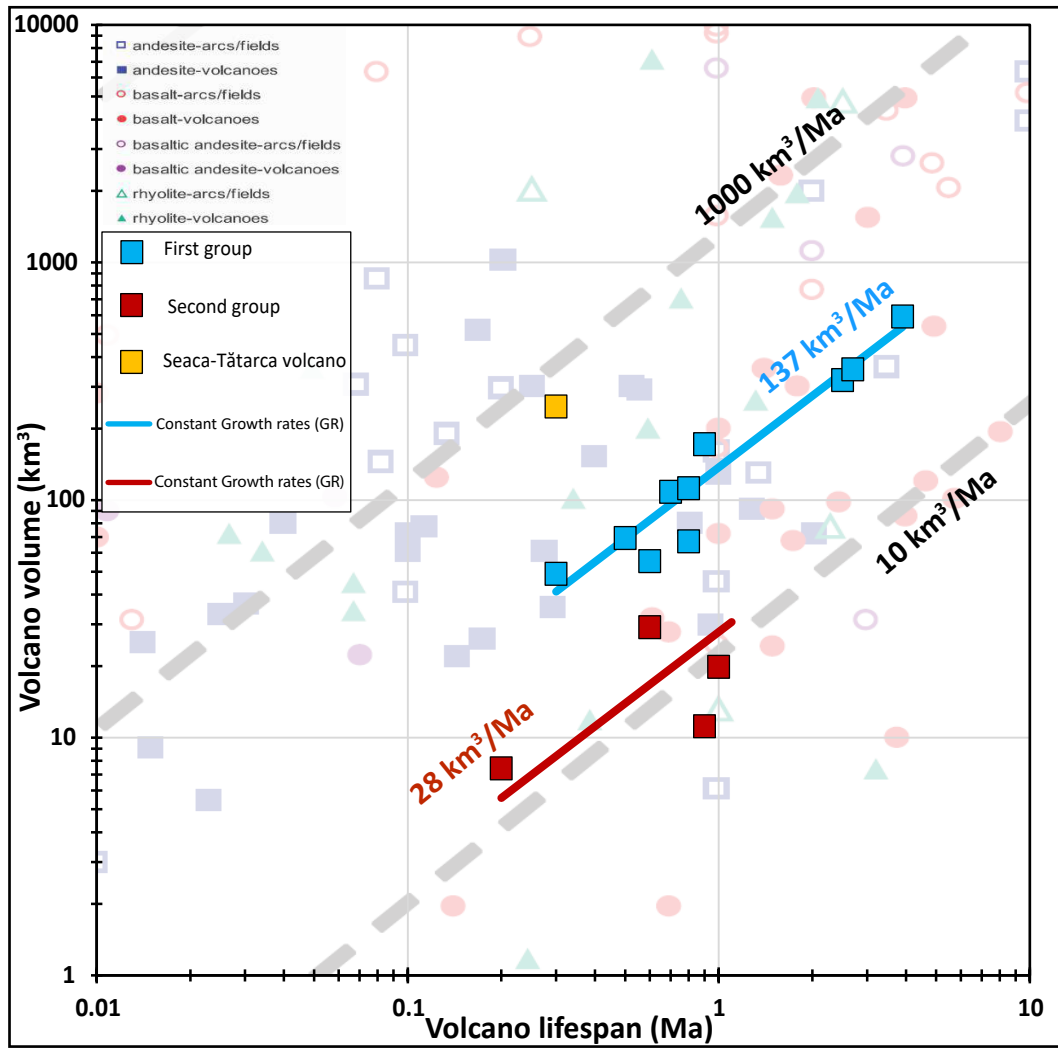
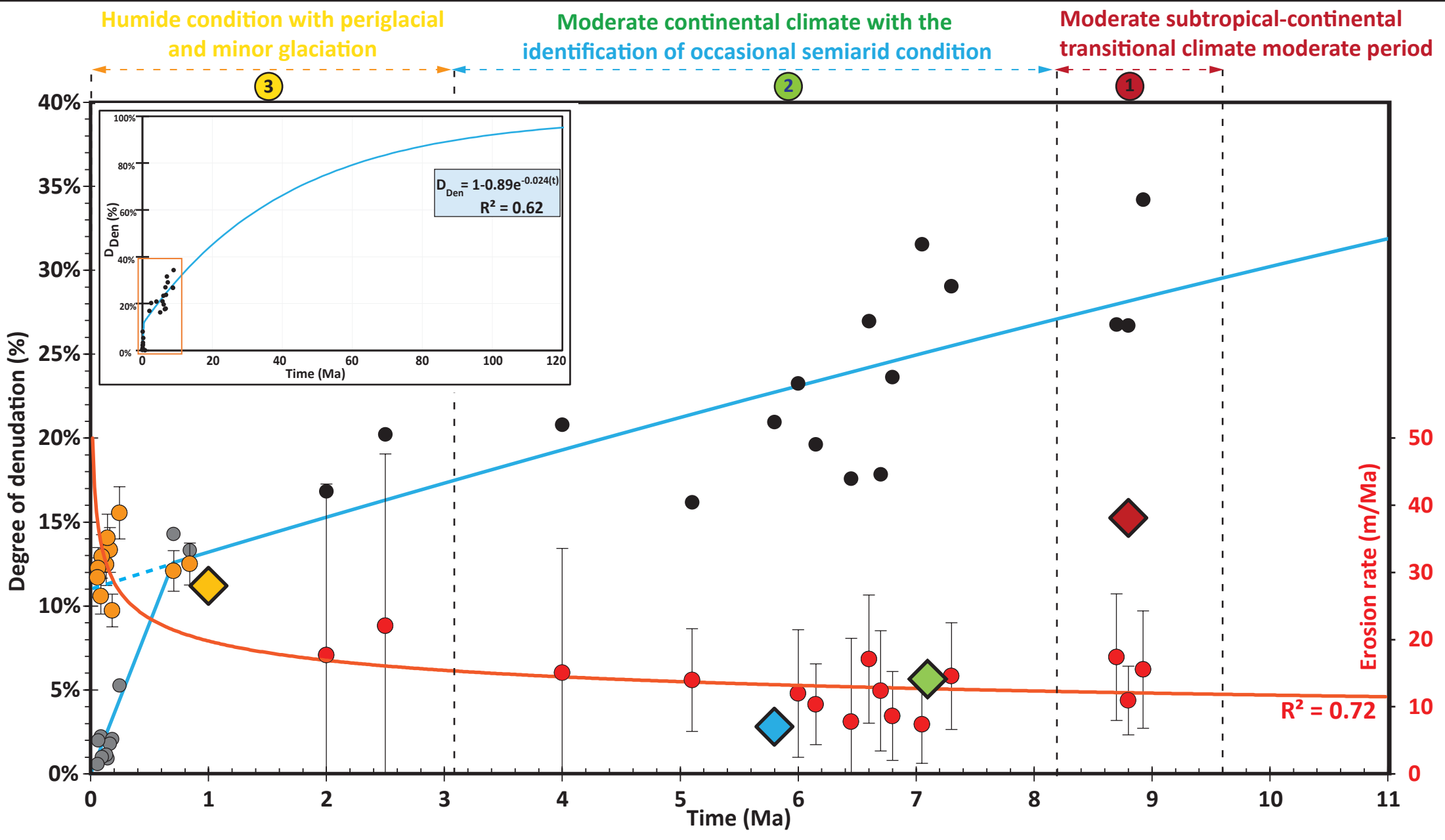


Fig. 12



Tables

Table 1. New K-Ar dating results (details of measurement in S.M).

Sample code	Rock type	K%	Mean age \pm 1 σ (Ma)	
17EC14	Dacite	2.730	1.98 \pm 0.06	Pilișca
17EC15	Dacite	2.841	2.11 \pm 0.04	
17EC11	Pyr. andesite	1.503	4.61 \pm 0.07	Vârghis
17EC09	Pyr. andesite	1.551	4.65 \pm 0.07	
17EC06	Pyr. andesite	1.932	4.79 \pm 0.07	
17EC10	Pyr. andesite	1.995	4.89 \pm 0.07	
17EC07	Pyr. andesite	1.326	5.06 \pm 0.07	
17EC08	Am. andesite	2.373	5.47 \pm 0.08	
17EC02	Pyr. andesite	1.219	6.47 \pm 0.09	Seaca-Tătarca
17EC01	Pyr. andesite	1.328	6.52 \pm 0.09	
17EC05	Pyr. andesite	1.517	6.61 \pm 0.09	
17EC04	Pyr. andesite	1.461	6.65 \pm 0.09	
17EC03	Pyr. andesite	1.193	6.79 \pm 0.10	
17KEL6	Px-Amph And.	1.680	8.33 \pm 0.12	Călmăni
17P-KEL6	Px-Amph And.	1.648	8.30 \pm 0.12	
17KEL5	Monzodiorite	2.755	8.26 \pm 0.12	
17KEL1	Dacite	2.291	6.69 \pm 0.10	

Table 2. Volumetric data (computed from modelled volcanic uppermost surface) and growth rate for each of CGH volcano (labelled as in Fig. 1) and each CGH sectors (at bottom of the table). Values are quoted with uncertainties at 1σ -level. Constructed volume (V_C , in km^3), degree of denudation (D_{Den}): proportion of volcano lost by dismantling processes (in %). All morphometric values from this study except for Ciomadul volcano (Karátson et al., 2019)

Sector	Label and Volcanoes	Radiometric data			Volumetric data						
		Age _{Beginning} (Ma)	Age _{Ending} (Ma)	Lifespan (ΔT) (Ma)	Visible Area (km^2)	Height (m) a.base.l \pm uncertainty (σ_{ZSV}) (m)	Constructed volume (V_C in km^3)	Growth rate (GR) (km^3/Ma)	Eroded Volume (V_E in km^3)	Erosion rate (ER) (m /Ma)	Degree of Denudation (D_{Den})
Călimani	01 : Central Călimani (Kelemen)	10.6	6.7	3.9	1041	2072 \pm 82	592 \pm 115	152	158 \pm 85	17 \pm 9	27%
	02 : Moldoveanul (Moldovánka)	10.2	7.7	2.5	779	1053 \pm 78	320 \pm 82	128	108 \pm 61	16 \pm 9	34%
Gurghiu	03 : Jirca (Nagy-Erdős)	9.2	8.4	0.8	137	1028 \pm 45	112 \pm 34	140	13 \pm 6	11 \pm 5	27%
	04 : Fâncel-Lăpușna (Fancsal)	8.7	6.0	2.7	828	1499 \pm 58	355 \pm 90	131	88 \pm 48	15 \pm 8	29%
	05 : Bacta (Bakta)	7.5	6.6	0.9	59	490 \pm 41	11 \pm 6	12	3 \pm 2	7 \pm 6	32%
	06 : Seaca-Tătarca (Mezőhavas)	6.8	6.5	0.3	543	1650 \pm 63	248 \pm 57	825	61 \pm 34	17 \pm 10	27%
	07 : Ciumani (Csomafalvi Délhegy)	7.1	6.3	0.8	106	1284 \pm 60	67 \pm 18	84	9 \pm 6	12 \pm 9	18%
	08 : Borzont	-	-	-	59	493 \pm 45	21 \pm 8	-	3 \pm 3	-	24%
	09 : Șumuleu (Somlyó)	6.7	6.2	0.5	242	1073 \pm 80	69 \pm 25	138	12 \pm 19	8 \pm 12	18%
North Harghita	10 : Ostoros (Osztoróc)	6.3	6	0.3	93	1008 \pm 37	49 \pm 16	163	6 \pm 3	10 \pm 6	20%
	11 : Ivo-Cocoizaș (Fertő-tető)	6.3	5.7	0.6	95	1104 \pm 57	55 \pm 16	92	7 \pm 5	12 \pm 10	23%
	12 : Râchitis (Csíkmagosa)	-	-	-	67	269 \pm 24	3 \pm 3	-	1 \pm 2	-	21%
	13 : Vârghiș (Madarasi-Hargita)	5.5	4.6	0.9	355	1611 \pm 39	172 \pm 34	191	25 \pm 14	14 \pm 8	16%
South Harghita	14 : Luci-Lazu (Nagykőbük- Tetőfenyő)	4.3	3.6	0.7	306	843 \pm 74	108 \pm 37	155	18 \pm 23	15 \pm 19	21%
	15 : Cucu (Kakukkhegy)	2.8	2.2	0.6	88	586 \pm 64	29 \pm 11	49	5 \pm 6	22 \pm 26	20%
	16 : Pilișca (Piliske)	2.5	1.5	1.0	95	709 \pm 51	20 \pm 8	20	3 \pm 5	18 \pm 26	17%
	17 : Central Ciomadul (Csomád)	0.2	0.03	0.2	72	-	7 \pm 1	37	0.2 \pm 0.1	26 \pm 4	3%
CGH sector		Activity duration (Ma)		Total volume constructed per area (km^3)	Growth rates per area (km^3/Ma)		Degree of Denudation per sector (D_{Den} %)		Eroded volume (km^3)	Erosion rates (m/Ma)	
Călimani		3.9		910 \pm 140	235		30		265 \pm 105	17	
Gurghiu		3.4		883 \pm 117	270		25		190 \pm 63	11	
North Harghita		2.4		279 \pm 41	116		20		39 \pm 15	9	
South Harghita		4.3		165 \pm 40	39		15		27 \pm 24	28	
CGH total		~11 Ma		2239\pm192	~200		~ 22		524\pm125	~20	

Table 3. Specific erosion rates along CGH range history for each contemporaneous climate period (details in Fig. 12 legend).

Sector	Period of activity (Ma)	Average age (Ma)	Erosion rate (m/Ma)	Climate fluctuation
Călimani	10.8 - 6.9	8.8	38	Short-time moderate period subtropical-continental transitional climate (More Humid; 3)
Gurghiu	8.7 - 5.5	7.1	14	Moderate continental climate (Less humid ; 2)
North Harghita and Luci-Lazu edifice	7- 4.6	5.8	7	Moderate continental climate with the identification of occasional semiarid condition (2)
South Harghita (Cucu, Pilișca, Ciomadul)	<3 - present	1.0	28	Moderate continental Humid condition with periglacial and minor glaciation (1)