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Abstract

This study presents the first cosmogenic 36Cl surface exposure data from a moraine in the Former Yugoslav Republic of Macedonia (FYROM). Five limestone boulders from a terminal moraine in the Galicica Mountains (40.94°N, 20.83°E, 2050 m a.s.l.) were used for cosmogenic 36CI surface exposure dating. The 36CI concentrations from the five boulders are identical within their measurement uncertainties ruling out major effects of inheritance, erosion, or snow cover. The calculated ages are very consistent ranging from 11.3 to 12.8 ka (mean 12 ka) after applying a Caspallation production rate of 56 at g-1 a-1 (LSD scaling) and correction for 5 mm ka-1 carbonate weathering and 2 % snow shielding. The applied corrections for weathering and snow shielding cause a shift to older ages in the order of magnitude of ca. 5 % on average, making the production rate the main impact on exposure ages. The ages point to a moraine formation during the Younger Dryas period, consistent with the timing of the last deglaciation in the Galicica Mountains derived from previous geomorphological studies in the area. The formation of a glacier was likely favoured by several topoclimatic factors, accounting for additional snow input. This interpretation is in line with regional studies on glaciation chronologies from Šara Range (FYROM/Republic of Kosovo), Retezat Mountains (Romania), Mount Orjen (Montenegro) and Durmitor (Montenegro). Lake sediment analyses of lakes Prespa (Republic of Albania/ FYROM/Greece), Malig (Republic of Albania) and Dojran (FYROM/Greece) indicate that cold conditions promoted the formation of a local circue glacier. However, studies of sediment records of the adjacent lakes Ohrid (Republic of Albania/FYROM) and Prespa do not indicate the presence of a proximal glaciation. An explanation might be a combination of the small size of the circue glacier, generating only small amounts of debris, and the karstic bedrock. which hampers fluvial transport and acts by its aquifer system as a natural sediment trap, as the fluvial transport of the sediments to the lakes is absorbed by the karst system.

Keywords	Galicica Mountains; Chlorine-36 (36Cl); Cosmogenic surface exposure dating; Younger Dryas; Balkan Peninsula; Moraine
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To the editor of Quaternary International



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Köln den 21.02.17

To whom it may concern,

We conducted a study in the Galicica Mountains (Former Yugoslav Republic of Macedonia) on a terminal moraine of Younger Dryas age using cosmogenic 36-Chlorine surface exposure dating. The data has been set in context to other Balkan deglaciation chronologies. Also paleoclimate reconstructions derived from nearby lake records were evaluated concerning a possible imprint of a local glaciation on the sedimentation pattern of the lake on the one hand, and if the climate reconstructions reveal favourable climate during the time of a local glaciation on the other hand.

We are confident that Quaternary International would be an excellent journal to publish this data since it would stand in line with similar publications (e.g. Reuther et al., 2007, Makos et al., 2013, Kuhlemann et al., 2013). This study contributes to the reconstruction of glaciations on the Balkan Peninsula.

Thank you in advance for considering to publish our research.

Best regards

Raphael Gromig



Quaternary International

We the authors declare that this manuscript is original, has not been published before and is not currently being considered for publication elsewhere.

We confirm that the manuscript has been read and approved by all named authors and that there are no other persons who satisfied the criteria for authorship but are not listed. We further confirm that the order of authors listed in the manuscript has been approved by all of us.

We understand that the Corresponding Author is the sole contact for the Editorial process. He/She is responsible for communicating with the other authors about progress, submissions of revisions and final approval of proofs.

Sincerely,

On behalf of all authors

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Evidence for a Younger Dryas deglaciation in the Galicica Mountains (FYROM) from
 cosmogenic 36-Chlorine

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13 Abstract

14

15 This study presents the first cosmogenic ³⁶Cl surface exposure data from a moraine in the 16 Former Yugoslav Republic of Macedonia (FYROM). Five limestone boulders from a terminal 17 moraine in the Galicica Mountains (40.94°N, 20.83°E, 2050 m a.s.l.) were used for 18 cosmogenic ³⁶Cl surface exposure dating. The ³⁶Cl concentrations from the five boulders are 19 identical within their measurement uncertainties ruling out major effects of inheritance, 20 erosion, or snow cover. The calculated ages are very consistent ranging from 11.3 to 12.8 ka 21 (mean 12 ka) after applying a Ca-spallation production rate of 56 at g⁻¹ a⁻¹ (LSD scaling) and 22 correction for 5 mm ka⁻¹ carbonate weathering and 2 % snow shielding. The applied 23 corrections for weathering and snow shielding cause a shift to older ages in the order of 24 magnitude of ca. 5 % on average, making the production rate the main impact on exposure 25 ages. The ages point to a moraine formation during the Younger Dryas period, consistent 26 with the timing of the last deglaciation in the Galicica Mountains derived from previous 27 geomorphological studies in the area. The formation of a glacier was likely favoured by 28 several topoclimatic factors, accounting for additional snow input. This interpretation is in line 29 with regional studies on glaciation chronologies from Sara Range (FYROM/Republic of 30 Kosovo), Retezat Mountains (Romania), Mount Orjen (Montenegro) and Durmitor 31 (Montenegro). Lake sediment analyses of lakes Prespa (Republic of 32 Albania/FYROM/Greece), Malig (Republic of Albania) and Dojran (FYROM/Greece) indicate 33 that cold conditions promoted the formation of a local cirgue glacier. However, studies of 34 sediment records of the adjacent lakes Ohrid (Republic of Albania/FYROM) and Prespa do 35 not indicate the presence of a proximal glaciation. An explanation might be a combination of 36 the small size of the circue glacier, generating only small amounts of debris, and the karstic 37 bedrock, which hampers fluvial transport and acts by its aquifer system as a natural sediment 38 trap, as the fluvial transport of the sediments to the lakes is absorbed by the karst system.

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Keywords: Galicica Mountains, Chlorine-36 (³⁶Cl), Cosmogenic surface exposure dating,
 Younger Dryas, Balkan Peninsula, Moraine

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44 **1 Introduction**

45 The glaciation history on the Balkan Peninsula is subject of research since the late 19th 46 century (Hughes et al., 2011 and references therein). It is of particular interest since glacier 47 reconstructions provide a valuable tool to contribute to Quaternary paleoclimate 48 reconstructions. The southerly latitude position of these former glaciers on the Balkan Peninsula made them especially sensitive to climatic changes, i.e. they react faster with 49 50 glacier retreat or advance if there are even minor shifts in the climatic conditions (Hughes 51 and Woodward, 2008; Sarıkaya et al., 2014). Several glaciations occurred and are 52 documented on the Balkan Peninsula since the Mid Pleistocene. To date, numerous glacial 53 studies in the Balkan Mountains have been carried out, for example in the Durmitor Massif, 54 Mount Orjen and Mount Lovćen in Montenegro (Hughes, 2010; Hughes et al., 2011; Žebre 55 and Stepišnik, 2014), Mount Prokletije in the Albanian Alps (e.g. Milivojević et al., 2008), the 56 Rila Mountains in Bulgaria (e.g. Kuhlemann et al., 2013), or Mount Tymphi and Mount 57 Smolikas in northern Greece (Hughes et al., 2003; Hughes et al., 2006a; 2007b, c.f. Fig. 6). 58 In most regions, the glaciation during the Marine Isotope Stage (MIS) 12 (Skamnellian Stage, 59 ca. 478 – 424 ka BP) (Lisiecki and Raymo, 2005) is considered to represent the maximum 60 glacier advances (e.g. Hughes et al., 2010; Hughes et al., 2011; Adamson et al., 2014). 61 However, a precise dating of glacial features, such as of moraines, is lacking in most studies. 62 One reason for the fragmentary age control is that many of the investigated areas are 63 composed of limestone, where age determination is limited to U-series dating of secondary 64 carbonate cements on moraines, which does not necessarily date the actual formation of a 65 moraine but the precipitation of the calcitic cement (Hughes et al., 2013). In some areas, 66 where quartz-bearing rocks are common, cosmogenic ¹⁰Be surface exposure dating was 67 conducted (e.g. Reuther et al., 2007; Kuhlemann et al., 2013).

Studies in the Former Yugoslav Republic of Macedonia (FYROM, in the following referred to as Macedonia) are very rare, which is surprising, as Macedonia is situated in a central position of the Balkan Peninsula. The Šara Mountain Range at the border of Macedonia and the Republic of Kosovo was studied by Kuhlemann et al. (2009). In the Galicica Mountains, which separate Lakes Ohrid and Prespa, a precise mapping of geomorphological features allowed a reconstruction of the expansion of local cirque glaciers (Ribolini et al., 2011).

We now present the first cosmogenic ³⁶Cl exposure ages of a glacial moraine in the Galicica Mountains. The results are set in context with the reconstructed deglaciation history of the Balkan Peninsula in order to improve the understanding of the regional climate history. The results are also compared to other sediment records from nearby lakes in order to test if the results are in line with the paleoclimatic interpretation.

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80

82 **2** Location, Regional Geology and Climate of the Galicica Mountains

83 The Galicica Mountain Range is situated in the southern Balkan region (Fig. 1) and is mostly 84 belonging to the Republic of Macedonia. The highest peak is Magaro with a maximum 85 elevation of 2255 m above sea level (a.s.l.). The mountain range is framed by the Ohrid 86 graben with Lake Ohrid (693 m a.s.l.) to the west and the Prespa graben with Lake Prespa (849 m a.s.l.) to the east. Towards the south, the Galicica Mountain Range extends into the 87 88 Republic of Albania, where it is named Mali i Thatë Mountains, with the highest peak being 89 Kota with an elevation of 2265 m a.s.l. (Watzin et al., 2002; Bordon et al., 2009; Hoffmann et 90 al., 2010).

91

92 The Galicica Mountains are a horst structure, which is intersected by a E-W striking normal 93 fault, creating a wind gap extending from Lake Prespa to Ohrid (Hoffmann et al., 2010). The 94 Galicica Mountains are mainly composed of Triassic and subordinated Early Jurassic 95 (Hoffmann et al., 2010) unfossiliferous limestones and locally dolomites (Wagner et al., 2008; 96 Ribolini et al., 2011). The limestones are strongly karstified, rugged and broken (Wagner et 97 al., 2008). Isolated clastic sedimentary rocks of Triassic age occur occasionally (Vogel et al., 98 2010b). However, the area studied here in more detail, located ca. 2 km south of the wind 99 gap (Fig. 1, further described below) is entirely composed of Triassic limestone. The circue is 100 approx. 400 m wide. The headwall is partly very steep, producing large slope debris 101 surfaces. Rockfalls, generating large boulders debris, are also active today, forming a belt of 102 large blocks at the base of talus fan (Ribolini et al., 2011).

103

104 The large scale climate in Macedonia is controlled by the proximity to the Aegean Sea in the 105 south-east and the Adriatic Sea in the west. Additionally, the surrounding mountains have a 106 crucial influence on the regional climate, which shows characteristics of Mediterranean as 107 well as continental influences (Watzin et al., 2002; Leng et al., 2013). At higher altitudes, 108 montane climate predominates, with higher daily temperature differences compared to the 109 areas in close proximity of lakes Ohrid and Prespa, that buffer daily temperature variations 110 (Watzin et al., 2002). The climate at lakes Ohrid and Prespa for the period between 1961 and 111 1990 is recorded by several weather stations at altitudes below 1020 m a.s.l.. Present day 112 weather stations at higher altitudes unfortunately do not exist. The Ohrid meteorological 113 station (760 m a.s.l., near the city of Ohrid) recorded an annual average temperature of 11.1° 114 C, whereas Resen meteorological station (881 m a.s.l.) at Lake Prespa recorded a mean 115 annual temperature of 9.5°C (Popovska and Bonacci, 2007). Applying a thermal lapse rate of 116 0.6 °C 100 m⁻¹ to the average annual temperature of Ohrid meteorological station leads to an 117 annual average temperature of ca. 3.4 °C at the altitude of the sampled moraine sequence, 118 which is situated at 2010 - 2050 m a.s.l. (ca. 2.5 °C based on the Prespa meteorological

119 station). The climate in the Lake Prespa watershed is considered to exhibit more temperature 120 variations compared to the Lake Ohrid watershed, as Lake Ohrid, which has a water volume 121 15 times larger than Lake Prespa, acts as a strong thermal buffer (Popovska and Bonacci, 122 2007). The precipitation data recorded between 1961 and 1990 reveals an annual average 123 precipitation of 907 mm in Lake Ohrid watershed and 795 mm in Lake Prespa watershed (Popovska and Bonacci, 2007). After 1990 no continuous climate record is published. 124 125 Snowfall in the mountains is assumed to exceed 1 m annually and to last until late spring 126 (Hollis and Stevenson, 1997).

127 In total four moraine sequences were mapped in detail by Ribolini et al. (2011), and range in 128 altitudes from 1780 to 2050 m a.s.l. (Fig. 2). The studied moraine is located in the southern 129 of two cirques. The lower moraines (G1-3 sequences) are less well shaped and lack material 130 suitable for exposure dating. Ice moulted rocks occur and are partly covered by fluvio-glacial 131 deposits. Dating with other methods, i.e. radiocarbon dating, was also not possible, as a 4-m 132 deep borehole in the sediments dammed by the G3 moraine did not deliver datable material 133 and it was not possible to get deep enough, for a rough dating of the G3.

- 134 In contrast, at 2010 – 2060 m a.s.l. a series of well preserved nested moraines forms the G4-135 moraine sequence (Fig. 2), which is considered to have built up in the course of the same 136 glaciation phase (Ribolini et al., 2011). The largest moraine ridge was sampled for 137 cosmogenic ³⁶Cl dating on carbonates (Fig. 3). The distance of the G4 moraine to the 138 headwall of the circue is approx. 400 m and thus most likely not affected by rockfalls from the 139 headwall, also due to the elevated position. The moraine is mainly composed of limestone 140 blocks, pebbles and coarse gravel. Depressions between the ridges originated from glacier 141 retreat stages. Karst dissolution and permafrost creep may have intensified deformation of 142 the original topography of some ridges (Ribolini et al., 2011).
- 143

144 **3 36Cl dating method**

36CI dating has successfully been applied to reconstruct glacial chronologies in several
areas (e.g. Phillips et al., 1997; Makos et al., 2013; Sarıkaya et al., 2014).

With known production rates, the built up of cosmogenic nuclides can be converted into ages, which reflects the time, when the dated material was first exposed to secondary cosmic rays. For this application, the surface exposure ages mark the point of a glacier retreat when fresh rock is deposited in terminal moraines, which then start to accumulate cosmogenic nuclides.

152

153 3.1 Sample collection

Five limestone boulders in or near crest-position of the well-defined terminal moraine G4 were sampled for ³⁶Cl exposure dating (Fig. 3). Today, the moraine exhibits a crest height of

156 approx. 14 m with reference to the base of the uphill side of the moraine and shows 157 implications of only moderate degradation since deposition. Boulders were chosen based on 158 their size, their plausibility to be in stable position and their post-depositional preservation. 159 The samples themself were collected at the upper surfaces of each boulder with a hammer 160 and chisel (Fig. 4). Horizon angles were measured in the field using an inclinometer in 20° 161 intervals in order to correct for topographic shielding.

162

163 3.2. Sample preparation

164 The samples were treated mechanically and chemically at the laboratory facilities of the 165 Institute of Geology and Mineralogy at the University of Cologne, Germany, following mainly 166 the chemistry protocol of (Stone et al., 1996). The weathered rock surface and any 167 contaminations like recrystallized calcite or lichen were mechanically removed using a hand 168 held rotary tool with a diamond bit. This is relevant as they can contain a relevant amount 169 ³⁵Cl, which acts as a target element to produce ³⁶Cl by thermal neutrons. Subsequently, the 170 boulders were crushed and sieved and the $250 - 500 \,\mu m$ grain size fraction was treated with 171 30 % H₂O₂ at 40° C for 96 hours in order to remove any residual organic matter. Each aliguot 172 was rinsed and leached twice in 0.3 M HNO₃ until ~13% of material was removed in order to 173 eliminate any meteoric ³⁶Cl. Afterwards the samples were spiked with a known amount of 174 isotopically enriched stable chloride carrier to allow simultaneous determination of the natural 175 chlorine and ³⁶Cl (Desilets et al., 2006 and references therein). After adding the carrier and some excess AqNO₃ to avoid degassing, ~22 g of each sample were dissolved using MilliQ 176 177 water and subsequent careful addition of 2 M HNO₃. As sulphur displays a major issue for 178 Accelerator Mass Spectrometer (AMS) ³⁶Cl measurement, because ³⁶S it is an isobar of ³⁶Cl, 179 it was removed by extraction of crystallized $BaSO_4$ after addition of $Ba(NO_3)_2$. Finally, the 180 precipitated and dried AgCI powder was pressed into AMS cathodes and measured at the 181 CologneAMS facility (Klein et al., 2011). To track any chlorine contamination in the chemistry 182 process, a chemistry blank is coupled with each series and measured in the same AMS run.

183

184 3.3. Sample Measurements (AMS; Actlabs, ICP-OES)

To ensure the accuracy and precision of the AMS measurement, the AMS was calibrated and normalized to three standards with different concentrations (36 Cl/Cl: 5.000 x 10⁻¹³, 1.600 x 10⁻¹², and 1.000 x 10⁻¹¹) from the NIST SRM 4843 material (Sharma et al., 1990). Additionally, we prepared four pure spike blanks (Oak Ridge National Laboratory batch 150301, NaCl enriched with 35 Cl), and five blanks with the 35 Cl/ 37 Cl natural ratio of 3.127 (four Standard Reference Material 975 and a commercial NaCl purchased from VWR company).

191 Major and trace element contents of bulk non-leached sample material were measured by 192 fusion inductively coupled plasma and fusion mass spectrometry at Actlabs, Canada (Table

193 1), in order to calculate the sample specific production rate of ³⁶Cl. These analyses were only 194 performed for RG14/001 and RG14/005 since the limestone is very homogeneous, the 195 catchment is small and the natural chlorine content of the sample is very low. The 196 measurement of the two samples, therefore, can be considered as representative. In 197 addition, an aliquot of each dissolved sample was analyzed on the ICP-OES at the Institute 198 of Geology and Mineralogy in Cologne to reveal the composition of the target AMS-199 measured fraction (Table 1). The concentration of ³⁶Cl and natural chlorine was measured 200 via AMS at the CologneAMS facility (Table 2).

201

202 **4 Results**

203 *4.1 Sample chemistry*

204 Major as well as minor element concentrations point to a relatively pure limestone (Table 1). 205 For samples RG14/002, RG14/003 and RG14/004, values from the measurement of 206 RG14/001 were taken. Applying the value of RG14/005 instead of RG14/001 was tested and 207 the effect was minor i.e. in the range of ca. 1%. Elements, which can have a crucial influence 208 on the formation of ³⁶Cl via the thermal neutron pathway are consistently very low. Also K, Fe 209 and Ti, on which ³⁶Cl can be produced spallogenically, are low. Radiogenic production of ³⁶Cl 210 can also be excluded, as Uranium and Thorium, which can act as a neutron source once 211 they decay (Bierman et al., 1995), were below detection limit in the sampled rocks.

	Major elem	nents										Trace elements					
Sample ID	SiO2	TiO2	AI2O3	Fe2O3	MnO	MgO (wt-	CaO	Na2O	K2O	P2O5	CO2 (LOI)	CI	В	Sm	Gd	U	Th
	(wt-%)	(wt-%)	(wt-%)	(wt-%)	(wt-%)	%)	(wt-%)	(wt-%)	(wt-%)	(wt-%)	(wt-%)	ррт	ppm	ppm	ррт	ppm	ppm
RG14/001 - 004	0.71	0.003	0.28	0.04	0.003	0.4	54.92	0.09	0.05	0.03	42.56	23.6	< 0.5	<0.1	0.1	0.3	0.2
RG14/005	0.19	0.01	0.5	0.04	0	0.48	53.29	0	0	0.02	40.02	35.2	30	0.1	< 0.1	0.1	< 0.1

Table 1: Chemical composition of samples. Values below detection limit are marked with "<".

215

216 4.2 Cl and ³⁶Cl concentrations

Stable chlorine concentrations of the samples are consistently low ranging from 16 ± 1 to $34 \pm 2 \ \mu g \ g^{-1}$, indicating that the thermal neutron production of 36 Cl has only a minor contribution to the total production. Blank corrections were obtained from subtracting the number of Cl and 36 Cl atoms in the blank from the respective atoms in the samples, which resulted in a decrease of the uncorrected 36 Cl concentration by 1.2 - 1.4%. The measured 36 Cl concentrations are very consistent, ranging from $1.07(10^6)$ at g^{-1} to $1.21(10^6)$ at g^{-1} . This reflects a high precision of the measurement and a low geological variability (Table 2).

Table 2: Sample specific characteristics. A bulk density of 2.6 g cm⁻³ is assumed for all samples.

226

224

						36CI							
Sample ID	Latitude	Longitude	Altitude	thickness of sample	Topographic shielding factor	conc.	35CI/37CI	36CI/37CI	Prod. Rate	calc. Age° with 0 mm ka-1	calc. Age* with 5 mm ka-1	calc. Age° with 10 mm ka-1	calc. Age° with 5 mm ka-1
				•		(atoms			(atoms g-1 a-				
	°N (DD)	°E (DD)	m.a.s.l.	cm		g-1)			1)	erosion	erosion	erosion	erosion
								6.981E-					
RG 14/001	40.937333	20.825389	2051	2	0.964	1.07E+06	9.231	12	86.26	10.75 ± 1.11	11.28 ± 1.28	11.39 ± 1.39	10.78 ± 1.21
								6.015E-				44 22 + 4 26	
RG 14/002	40.937417	20.825472	2050	3	0.964	1.19E+06	7.726	12	87.83	11.77 ± 1.08	12.25 ± 1.17	11.32 ± 1.20	11.79 ± 1.20
RG 14/003	40 937583	20 825500	2048	2	0 964	1 13E+06	8 239	6 4F-12	87 28	11 24 + 1 18	11 74 + 1 23	11.81 ± 1.08	11 26 + 1 09
	40.001000	20.020000	2040	-	0.004	1.102.00	0.200	9.138E-	07.20	11.24 2 1.10	11.7421.20		11.20 2 1.00
RG 14/004	40.937694	20.825361	2040	2	0.967	1.10E+06	10.555	12	84.80	11.26 ±1.13	11.90 ± 1.29	12.09 ± 1.41	11.30 ± 1.29
								6.151E-					
RG 14/005	40.937889	20.825056	2048	6	0.969	1.21E+06	7.623	12	86.29	12.18 ± 1.23	12.79 ± 1.42	13.01 ± 1.31	12.21 ± 1.06
Blank B3	-	-	-	-	-		19.452	2.70E-13		-	-	-	-

227 ^ total sample specific contemporary depth average production rate

228 * corrected for snow shielding (2%)

229 ° no snow correction applied

230 4.3 ³⁶Cl ages

231 Exposure ages of the sampled boulders were computed based on the measured Cl isotope 232 ratios. For the calculations, we used cosmogenic ${}^{36}CI$ production rates of 56 ± 4.1 at ${}^{36}CI(q)$ 233 Ca)⁻¹ a⁻¹ for Ca spallation (Marrero et al., 2016), which is the main production pathway, 234 accounting for ca. 90-94 % of ³⁶Cl production. K spallation production rates of 155 ± 11 at 235 ³⁶Cl (g K)⁻¹ yr⁻¹ (Marrero et al., 2016) were used, but the impact of this production pathway is 236 low due to the low K content in the samples. Used thermal neutron production rate is 759 ± 237 180 neutrons (g air)⁻¹ a⁻¹ (Marrero et al., 2016). Also production via thermal neutron capture 238 is minor as the stable CI content of the samples is low (Table 2), ranging from ca. 5-7.5 %. 239 Production of ³⁶Cl via muons is also subordinated, accounting for ca. 2.5 % of the total 240 production. These production rates are scaled to sea level and high geomagnetic latitude. In 241 order to account for site-specific variations in the atmospheric secondary cosmic-ray flux 242 scaling factors of Lifton et al. (2014) were applied. The surface exposure ages were 243 calculated from the measured ³⁶Cl concentrations using the CRONUScalc (Cosmic-Ray 244 Produced Nuclide Systematics on Earth) (accessed October 2016) calculator with the 245 software Matlab® (R2016a, MathWorks®). The calculation allows for corrections for e.g. 246 shielding, which can be due to topographic shielding and snow cover, and erosion. 247 Topographic shielding correction factors as well as the sample-specific attenuation length 248 were determined using the CRONUS calculator. The exposure ages range between 10.8 ka 249 and 12.2 ka, with a mean age of 11.4 ka and a median of 11.3 ka (Table 2) without 250 corrections for erosion and no snow shielding and using of 56 \pm 4.1 at ³⁶Cl(g Ca)⁻¹ a⁻¹ for 251 spallation. Correction factors are individually discussed in chapter 5.1.

252

253 5 Discussion

254 5.1 36Cl age calculation uncertainties

The results of the timing of the moraine formation depend on different input parameters used for the age calculations, such as erosion, snow shielding, inheritance, and production rates. A proper discussion of these input parameters is needed to evaluate the robustness of the dating results. Major corrections and assumptions were defined by the following aspects:

259 (i) Erosion: Field observations did not indicate significant erosion of the upper boulder 260 surfaces itself (especially chemical weathering due to dissolution of limestone). However, 261 below the G4 moraine series, limestone surfaces exhibit weathering channels carving 262 several centimeters deep into the rock surface, suggesting dissolution of carbonatic rock 263 surfaces due to precipitation. Chemical weathering of limestone can cause erosion rates of 264 up to 10 mm ka⁻¹, according to observations of Ivy-Ochs et al. (2009). A dissolution rate in 265 this magnitude is in accordance with field experiments of (Plan, 2005). Experiments with 266 micro-erosion meters also confirm limestone dissolution rates in the same order of 267 magnitude or even higher (Stephenson and Finlayson, 2009 and references therein). We 268 think that 10 mm ka⁻¹ represents a maximum possible erosion rate. However, we propose a 269 lower erosion rate/limestone dissolution rate of 5 mm ka⁻¹ since field observations do not 270 suggest any higher limestone dissolution rates on the boulders itself.

(ii) Snow Shielding: The combination of Mediterranean as well as continental climate in the
region (Watzin et al., 2002), leads to substantial snow cover in the Galicica Mountains in
winter (Hollis and Stevenson, 1997). Hence, ignoring the effect of snow could lead to an
underestimation of the surface exposure ages. Correction for snow shielding is difficult to
assess for several reasons:

276 277 i) Climate data from locations with similar altitudes as the sampling site are not available

278 ii) Climatic conditions likely changed throughout the Holocene

279 iii) The effect of elevation of boulders on moraine crest is unclear

280 iv) Topography-related effects on snow accumulation unclear

281 v) The effect of snow shielding is not completely understood yet

282 A potential reduction of the production of the target element due to snow cover (Gosse and 283 Phillips, 2001) was partly disproved by Dunai et al. (2014). A hydrogen rich cover, such as 284 from snow, can significantly increase the production rate of certain nuclides (e.g. ³⁶Cl). 285 However, since the concentration of ³⁵Cl is consistently low throughout all five samples, the reinforcing effect of a snow cover is very limited. Hence, we assume that snow cover has a 286 287 net attenuating effect of the production of ³⁶Cl. Based on the available literature (Hollis and 288 Stevenson, 1997), we suggest a snow cover of 0.3 m for an average duration of 4 months 289 per year with a density of 0.3 g (cm³)⁻¹, leading to an age correction of approx. 2%, using the 290 equation of Gosse and Phillips (2001).

291 (iii) Inheritance: The exposure ages are very consistent with only minor differences within 292 the analytical uncertainties of the ³⁶Cl concentrations. The present debris fans indicate a high 293 supply of new rock material, which leads to a continuous renewal of the cliff surface. 294 According to Putkonen and Swanson (2003) the chance of dating boulders with significant 295 prior exposure is extremely low in settings, where erosion at the source rock delivering rock 296 debris is high. Relatively high temperature differences between day and night cycles as well 297 as frost shattering during winter season might be the driving mechanisms for relatively high 298 physical weathering. Inheritance can thus be ruled out.

(iv) **Production rates**: The amount of studies determining ³⁶Cl production rates is still limited and the published rates scatter significantly, particularly for Ca spallation and the muonic contribution. We used production rates recently published by Marrero et al. (2016) for which one out of three calibrations sites are on the same latitude as our study site and the published production rates are scaled to scaling factors of Lifton et al. (2014), to calculate the exposure ages. Since the geographical position of the Galicica Mountains is very close to the
sampling sites for production rate studies of Schimmelpfennig et al. (2011), we also tested
these production rates, which are in the order of ca. 20-25 % lower for Ca spallation and ca.
2-17 % for K spallation (depending on scaling model, respectively) than those of Marrero et
al. (2016), respectively, in order to calculate ages.

309

310 The uncorrected ages (Table 2) can be regarded as minimum ages. Applying an erosion rate 311 of 5 mm ka⁻¹ does not have any significant impact on the ages but should also not be ignored 312 (Fig. 5). An applied erosion of 10 mm ka⁻¹ shows a more significant effect towards older ages 313 in the order of 0.5 - 0.8 ka. We do not suggest that such a relatively high erosion rate applies 314 to the sampled boulders in this particular geological setting, we rather want to illustrate, that 315 erosion as one of the major correction factors has only a minor influence in such young 316 samples. The incorporation of snow shielding (2%) has an impact on the ages to the same 317 degree than the applied maximum erosion rate. In contrast to erosion and snow shielding, 318 the selection of Ca spallation production rates plays a crucial role regarding the impact on 319 the surface exposure ages. Applying the Ca spallation production rate of Schimmelpfennig et 320 al. (2011) yields significantly older ages, ranging from 13.9 to 15.5 ka (uncorrected for erosion). These ages are consistently younger than the time frame assessed for the global 321 322 last glacial maximum (LGM, 26.5 ka to 19 ka, Clark et al., 2009), even when applying an 323 erosion rate of 10 mm ka⁻¹ and consequently, a moraine formation in the course of a 324 deglaciation after the LGM can be excluded. These ages, however, would correspond to the 325 Older Dryas period, but as a consequence, no evidence for a Younger Dryas moraine would 326 then be evident. We favour the recently determined Ca spallation rate of Marrero et al. 327 (2016) and think that the Ca spallation rate of Schimmelpfennig et al. (2011) indicate 328 maximum ages and rather overestimate the surface exposure ages.

329 In this study because the ³⁶Cl concentrations and the calculated ages cluster very closely, we 330 can rule out a significant impact of moraine degradation and assume a permanent exposition 331 of the boulders, which reflect the actual deposition of the moraine. Consequently, only 332 chemical weathering due to precipitation needs to be taken in to account. Hence, the 333 calculated ages range from 11.3 to 12.8 ka (mean 12 ka) after correction for 5 mm ka⁻¹ 334 erosion, snow cover and topographic shielding. In this case the glacier advance forming this 335 moraine was centered in the Younger Dryas cold period. The sampled moraine is part of a 336 series of nested moraines located in the cirgue. Taking the stratigraphical order into account 337 the sampled moraine is the second oldest one of the G4 moraine sequence. Thus, the 338 moraine likely represents a stage of the glacier stagnation after the maximum advance has 339 already occurred.

341 5.2 Galicica moraine ages

Since the dated moraine is only one part of the G4 moraine series, the deglaciation after the
Younger Dryas glacier advance was likely interrupted by stagnations, leading to the
multicrested shape of the G4 moraine series.

Assuming that the G4 moraine sequence was formed in the course of the Younger Dryas period, the G3 moraine, which is situated on an altitude of ca. 1930 m a.s.l. was likely formed at either the Oldest Dryas period (ca. 17.5 – 14.5 ka BP), LGM or MIS 6 (ca. 190 – 130 ka BP). Also a moraine formation at the Older Dryas can be possible. Moreover it can be stated, that no glacier advances during the entire Holocene took place, due to a lack of geomorphological evidence.

351

352 5.3 Comparison to glaciation timing of other Balkan Mountain Ranges

353 Studies dating glacial retreats are very sparse on the Balkan Peninsula. However, findings in 354 other formerly glaciated areas are in good agreement with the results of this study. The most 355 proximal dated site to the Galicica Mountains is the Sara Range, which is situated at the 356 border of the Republic of Macedonia and the Republic of Kosovo. Kuhlemann et al. (2009) 357 sampled a series of moraines at altitudes between 1355 m a.s.l. and 2300 m a.s.l. for ¹⁰Be 358 surface exposure dating. The lowest moraine yielded ages of 19.4 ± 3.2 ka (1402 m a.s.l., N-359 facing cirque), 16.1 ± 2.3 and 12.4 ± 1.7 ka (1398 m a.s.l. and 1355 m a.s.l., respectively, same moraine ridge, NE-facing cirque) and 14.4 ± 2.1 (1776 m a.s.l., S-facing cirque) 360 361 including corrections for erosion (10 mm ka⁻¹), respectively. The inconsistency of the ages is 362 explained by dislocation of some of the sampled boulders. However, the oldest age points to 363 a moraine formation in the course of the LGM, whereas the others could potentially represent 364 recessional moraines (Hughes et al., 2013). At higher altitudes (2020 – 2300 m a.s.l.) 365 moraines were ascribed to the Younger Dryas, with ages from 11.9 ± 1.7 to 14.7 ± 2.1 ka, 366 with the same corrections applied for erosion. The higher altitude of certain Younger Dryas 367 moraines compared to the G4 moraine is not surprising, since there is a precipitation 368 gradient on the Balkan Peninsula, which decreases from west to east (Kuhlemann et al., 369 2009).

In the Retezat Mountains in Romania, Reuther et al. (2007) investigated glacial features and dated N-facing moraines using cosmogenic ¹⁰Be for surface exposure dating. They identified a Late Glacial glaciation phase clustering around 16.1 \pm 0.5 ka, implying that the local LGM was delayed by several thousand years compared to the global LGM, but coinciding with the Oldest Dryas (Heinrich-event H1 ca. 16.8 ca ka BP). However, they also identified a moraine ascribed to the Younger Dryas with ¹⁰Be ages of 11.4 \pm 1.3 ka and 13.6 \pm 1.5 ka (corrected for 5 mm ka⁻¹ erosion, uplift and snow cover). The boulders sampled for dating were not deposited on the moraine ridge, but directly above and below the moraine, representing thetime frame of the moraine built-up, which is in good agreement with our results.

Kuhlemann et al. (2013) dated moraines in the Rila Mountains (Bulgaria) using cosmogenic ¹⁰Be. They identified two glacial stages during which moraines were formed. Similar as in the Šara Mountains, the glacier retreats were asynchroneous with the LGM, with moraines dating at the beginning (24 – 23 ka BP) and the termination of the LGM (18 – 16 ka BP). Moraines documenting a glacier retreat at the Oldest Dryas and the Younger Dryas are not dated but suggested based on stratigraphical relationship of geomorphological findings, respectively.

A number of studies exist from Montenegro. Studies of Hughes et al. (2010) on Mt. Orjen revealed the presence of Younger Dryas moraines based on U-Series dating of secondary carbonates in moraines. These moraines are situated approx. 500 m lower than the G4 moraine in the Galicica Mountains. The differences in altitudes can be due to different local topoclimatic factors and much higher precipitation in Montenegro (today ca. 5000 – 6000 mm a⁻¹), which may have led to a much thicker sow cover and a lower equilibrium line altitude (ELA).

Also for the Durmitor Massif in Montenegro Younger Dryas moraines are reported by Hughes et al. (2011). Hughes et al. (2006a, 2007b) argue that maximum glacier advances in the Pindus mountains in Greece likely occurred between 30 and 25 ka, based on climate modelling of the pollen record of Lake Ioannina, which would be in agreement with the assumption of Kuhlemann et al. (2013) of dryness during the LGM. However, also evidence for a glacier readvance is reported for the Pindus Mountains (Hughes et al., 2006b).

For Mt. Prokletije at the border to Albania, Milivojević et al. (2008) hypothesize, based on detailed geomorphological mapping, calculation of ELAs and comparison of those to other Mediterranean mountains that the last of a total of three glacier advances occurred during the Younger Dryas. At Mount Lovćen moraine sequences are believed to be of older origin than Younger Dryas, most likely of LGM age (Žebre and Stepišnik, 2014). Precise correlation is not possible due to a lack of a dating of the geomorphological evidence.

405 The lack of evidence for Younger Dryas glaciation in certain areas might have different 406 causes. Topoclimatic factors, such as regional temperature and precipitation can play a key 407 role for the formation of a glacier. Additional snow availability can be promoted by 408 avalanching, and wind blown snow into a cirgue. The latter is likely in the Galicica Mountain 409 range from the plateau topping the cirgue. Hughes (2009) proposes that avalanching in front 410 of steep cliffs and snow drift can increase the snow accumulation by a factor of two as shown 411 for present day glaciers at Mount Prokletije (Albania/Montenegro). Moreover, the orientation 412 of the circue can promote glacier activity. A N-E oriented circue enables a better shading, 413 which can lead to less ablation due to direct solar radiation (Evans, 1977; Hughes et al.,

414 2006a). This feature is in particular common for glaciations younger than MIS 12 (Hughes et 415 al., 2007a). Also the geology may play a significant role in glacier formation. In the Galicica 416 Mountains, the karstic bedrock may have led to a formation of initial depressions, which then 417 acted as a snow-trap and promoted the accumulation of snow. Also the relatively light color 418 of limestone and, hence, relatively high albedo may have set favourable conditions for a 419 glacier growth (Hughes et al., 2006a). Taken together, these factors can explain the 420 formation of a glacier due to only a modest increase of the local ELA, which was calculated 421 to 2130 m a.s.l with respect to the ELA for the G2-3 glacial phase, (calculated to ca. 2000 m 422 a.s.l.) (Ribolini et al., 2011). This is significantly lower than the calculated ELA (2300 m a.s.l.) 423 for the Šara Mountain range for the Younger Dryas.(Kuhlemann et al., 2009).

424

5.4 The Younger Dryas in the sedimentary records of lakes Ohrid, Prespa, Maliq, and Dojran
It is likely that the dated moraine was formed during a more windy or wet period of the
Younger Dryas.

428 Nearby paleoclimate records reveal a sharp transition from the relatively warm and humid 429 Bølling/Allerød interstadial to the Younger Dryas period (Bordon et al., 2009; Lézine et al., 430 2010; Panagiotopoulos et al., 2013). Adrop of the mean annual temperature of around 10°C 431 is reconstructed at Lake Malig (Bordon et al., 2009), and lakes Ohrid and Prespa record (Fig. 432 1) the last occurrence of ice rafted debris during the Younger Dryas (Vogel et al., 2010a; 433 Aufgebauer et al., 2012). However, the Younger Dryas does not seem to have had constant 434 cold and dry conditions. Bordon et al. (2009) subdivide the Younger Dryas into three periods 435 according to best modern analogue technique using pollen assemblages: whilst 12.8-12.2 436 and 11.9 – 11.3 cal ka BP are interpreted as cold and dry, the intermediary interval was 437 slightly warmer and more humid. A paleoclimate record from Lake Prespa indicates rather 438 dry conditions between 13.2 and 12.6 cal ka BP, which were followed by wetter conditions 439 between 12.6 and 11.5 cal ka BP during the overall cold Younger Dryas (Aufgebauer et al., 440 2012). This is in line with a shift in pollen assemblages, where a decrease in Artemisia and 441 Chenopodiacaea is correlated with an increase in tree taxa (Panagiotopoulos et al., 2013), 442 and with a shift in the diatom assemblages, which suggest increased moisture availability 443 after 12.3 ka BP (Cvetkoska et al., 2014). These findings are also in agreement with those 444 from Lake Dojran, located ca. 160 km to the east of the Galicica Mountains (Fig. 6). Lake 445 Dojran records an extreme lake level low-stand between 12.5 and 12.1 cal ka BP and a 446 subsequent higher lake level during the second part of the Younger Dryas period until 11.5 447 cal ka BP (Francke et al., 2013). The different climate conditions in the course of the 448 Younger Dryas are not recorded in Lake Ohrid, as its much larger water volume may have 449 buffered some climatic shifts. However, Vogel et al. (2010a) postulate enhanced aeolian 450 supply and a stronger wind activity during the Younger Dryas period in the surroundings of

451 Lake Ohrid, which potentially promote snow accumulation in the glacier cirque of the Galicica452 Mountains.

Taking into account the errors of the respective age information for the lacustrine successions as well as for the cosmogenic nuclides, an unambiguous alignment of the G4 moraine formation and the existence of a cirque glacier to one of the described more humid periods during the Younger Dryas is not possible.

457

458 5.5 Imprints of glacial erosion on lakes Ohrid and Prespa?

459 Glacial activity usually enhances erosional processes that increase the amount of debris, 460 which is eventually transported into a sedimentary basin such as lakes Ohrid and Prespa. 461 Rather high supply of debris to the lakes would therefore be expected at the onset of the 462 deglaciation (Ballantyne, 2002) and would lead to an increase in the sedimentation rate of 463 both lakes during the Younger Dryas. However, the sediment cores from lakes Prespa 464 (Aufgebauer et al., 2012) and Ohrid (Vogel et al., 2010b) indicate relatively constant 465 sedimentation rates prior, during and after the Younger Dryas. Moreover, increased erosion 466 and transport from Galicica Mountain bedrock should increase the amount of detrital calcite 467 in the sediments of lakes Ohrid and Prespa. However, total inorganic carbon (TIC) is low 468 during the Younger Dryas and isotope investigations on the carbonates in the Lake Ohrid 469 sediments have revealed the formation of calcite is predominantly bound to authigenic 470 precipitation and siderite is formed diagenetically (Lacey et al., 2016).

471 The lack of a significant imprint of the reconstructed glaciation in the Galicica Mountains 472 during the Younger Dryas on the sediments of lakes Ohrid and Prespa might have three 473 different reasons. Firtsly, the limited number of age control points constraining the time 474 period of the Younger Dryas in the sediment cores Co1202 (Ohrid) (Vogel et al., 2010a) and 475 Co1215 (Prespa) (Aufgebauer et al., 2012) is probably not sufficient to record variations in 476 the sedimentation rates on centennial time scales. Moreover, the regional geological setting 477 with the strongly karstified Galicica Mountain range may have buffered increased erosion 478 and sediment transport to the lakes. River runoff to lake Ohrid only accounts for ca. 25% of 479 the total inflow today (Wagner et al., 2010). Popovska and Bonacci (2007) report that 40 % of 480 the water discharging into Lake Ohrid at St. Naum at its south eastern shore close to the 481 investigated moraine is fed by karstic water derived from Lake Prespa. The remaining 60 % 482 of the inflow at St. Naum originate from direct precipitation over the Galicica Mountains, 483 which, however, drains directly into the karst system (Popovska and Bonacci, 2007). As a 484 result, only small rivers and creeks feed lakes Ohrid and Prespa today, and the majority of 485 water and sediments formed by the Galicica glacier may not have reached the lakes via 486 surficial pathways.

487 Similar processes were observed at Mount Orjen in Montenegro where glacier-derived
488 headwater became uncoupled from the depositional basin due to the formation of a
489 subterranean karst system (Adamson et al., 2014).

A third explanation for the lack of a significant change in the sedimentation patterns of the lakes during the Younger Dryas is that the glacial erosion was relatively restricted. This seems plausible taking the small distance between the slopes of the cirque and the dated moraine, as well as the high distances to the shoreline of the recovered lacustrine sediment successions in lakes Ohrid and Prespa.

495

496

497 6 Conclusions

498 Five boulders from a terminal moraine in the Galicica Mountains were dated using 499 cosmogenic ³⁶Cl surface exposure dating on limestones in Macedonia. The ages indicate a 500 moraine formation in the course of the Younger Dryas period based on surface exposure 501 ages centering at ca. 12 ka (corrected for 5 mm ka⁻¹ erosion and snow shielding). 502 Corrections for erosion and snow cover have only a minor effect on the age calculation. The 503 calculated exposure ages are in good agreement with several studies on deglaciation 504 chronology from the Balkan Peninsula. The timing of the formation of the glacier is in line 505 with sediment based paleoclimate reconstructions of the adjacent Lakes Prespa and Malig 506 as well as Lake Dojran located ca. 160 km east of the Galicica Mountains, all of which point 507 to increased moisture availability along with cold temperatures and increased wind activity 508 from ca. 12.5 – 12.1 ka BP onwards.

509 On the other hand, the dated glacial event did not leave a significant imprint in the sediments 510 of the adjacent lakes Ohrid and Prespa, probably due to insufficient age control points to 511 constrain centennial variations in the sedimentation rates, the strong karstification of the 512 Galicica Mountains and decoupling of erosional transport from the sedimentary basins and 513 the restricted erosional force of the local glacier.

514 The results of this study display an important contribution to improve the glacial chronology 515 of the Balkan Peninsula and emphasize the necessity of analysing different archives of 516 paleoclimate information.

517

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- Figure 1: A) Overview map with Macedonia highlighted B) Overview map with locations of the Galicica Mountains, Lake Ohrid, Lake Prespa and Lake Maliq, Asterisks mark the City of Ohrid and Resen, respectively, where climate stations are situated C) Digital surface model (DSM) of the cirque with the G4 moraine series. The model was created using structurefrom-motion photogrammetry with the software Agisoft Photoscan. (please print as colored image)
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- Figure 2: Geomorphological overview map of the two cirques beneath Magaro Mountain.
 Sampled moraine is highlighted in detail map (modified after Ribolini et al., 2011). (please
 print as colored image)
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Figure 3: Panorama picture of the sampled moraine. Arrows indicate position of sampled boulders. Picture is taken in downvalley direction, i.e. picture shows inner side of the moraine. (please print as colored image)

- Figure 4: Sampled boulders on the moraine crest. Arrows point to exact sampling position on each boulder. Hammer for scale. (please print as colored image)
- Figure 5: Probability density plots showing the calculated ages. The dashed box reflects the period of the Younger Dryas, blue area the Last Glacial Maximum. Black lines represent normal distribution of the single ages, red line displays cumulative relative probability.
 Corrections for erosion and snow cover are given in the diagrams, respectively. Thick arrows indicate significant impact of the applied correction on the exposure ages, while thin arrow indicates only minor impact. We used a production rate of ³⁶Cl from Ca spallation of 56 at g⁻¹ (Marrero et al., 2016) and LSD scaling. (please print as colored image)
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Figure 6: Overview map with locations of studied glaciers on the Balkan Peninsula: 1)
Durmitor Massif 2) Mt. Orjen 3) Mt. Lovćen 4) Mt. Prokletije 5) Rila Mts. 6) Mt. Tymphi 7) Mt
Smolikas 8) Galicica Mts. 9) Šara Mts. Lake Dojran is marked with the red asterisk. Authors
corresponding to the Study areas are noted in the text. (please print as colored image)







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