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Sedimentary response to a collision orogeny recorded in detrital zircon provenance of Greater Caucasus foreland basin sediments

Alexander R. Tye^{a,*}, Nathan A. Niemi^a, Rafiq T. Safarov^b, Fakhraddin A. Kadirov^b, Gulam R. Babayev^b

^aUniversity of Michigan, Ann Arbor, Department of Earth and Environmental Sciences, MI 48109

^bInstitute of Geology and Geophysics, Azerbaijan National Academy of Sciences, 119, H. Javid Ave., Baku, AZ1143

Abstract

The Greater Caucasus orogen on the southern margin of Eurasia is hypothesized to be a young collisional system and may present an opportunity to probe the structural, sedimentary, and geodynamic effects of continental collision. We present detrital zircon U-Pb age data from the Caucasus region that constrain changes in sediment routing and source exposure during the late Cenozoic convergence and collision between the Greater Caucasus orogen and the Lesser Caucasus, an arc terrane on the lower plate of the system. During Oligocene to Middle Miocene time, following the initiation of deformation within the Greater Caucasus, deep marine strata were deposited between the Greater and Lesser Caucasus, and detrital zircon age data suggest no mixing of Greater Caucasus and Lesser Caucasus detritus. During Middle to Late Miocene time, Greater Caucasus detritus was deposited onto the Lesser Caucasus basin margin, and terrestrial, largely conglomeratic, sedimentation began between the Greater and Lesser Caucasus. Around 5.3 Ma, upper plate exhumation rates increased and shortening migrated to pro- and retro-wedge fold-thrust belts, coinciding with the initiation of foreland basin erosion. Sediment composition, provenance, and structural data from the orogen together suggest the existence of a wide (230 - 280 km) marine basin that was progressively closed during Oligocene to Late Miocene time, probably by subduction/lithospheric underthrusting beneath the Greater Caucasus, followed by initiation

*Corresponding author: alextye@umich.edu

of collision between the Lesser Caucasus arc terrane and the Greater Caucasus in Late Miocene to Pliocene time.

The pace of the transition from hypothesized subduction to collision in the Caucasus is consistent with predictions from numerical modeling for a system with moderate convergence rates (<13 mm/yr) and hot lower plate continental lithosphere. Basement crystallization histories implied by our detrital zircon age data suggest the presence of two pre-Jurassic sutures between stable Eurasia and the Lesser Caucasus, which likely guided later deformation.

Keywords: detrital zircon, provenance, collision, Caucasus, Tethys

1. Introduction

The collision of two continents following the closure of an intervening ocean basin is a key element in the plate tectonic cycle (e.g., Nance et al., 2014). The transition from subduction to collision, where lower plate buoyancy or other factors inhibit the downward motion of subducting lithosphere into the mantle, constitutes a major change in the balance of forces acting on an orogen (Beaumont et al., 1996; Regard et al., 2003; Duret et al., 2011, 2012). The initiation of collision has been hypothesized to affect topography (e.g., England and Houseman, 1986), plate kinematics (Patriat and Achache, 1984; Dewey et al., 1989), and climate (e.g., Edmond, 1992; Molnar et al., 2010; Jagoutz et al., 2016). Observations from numerous orogens and modeling studies show that the transition from subduction to collision is a complex and diachronous process, beginning with the entrance of continental or transitional lithosphere into a subduction zone (Klootwijk et al., 1985; Lee and Lawver, 1995; Regard et al., 2003; Chung et al., 2005; Madanipour et al., 2017), and subsequently involving diverse effects such as accretion of large parts of the lower plate, locking of the trench and development of fold and thrust belts, slowing of convergence, and/or initiation of far-field deformation (Lee and Lawver, 1995; Regard et al., 2003; Toussaint et al., 2004a; van Hinsbergen et al., 2012; Cowgill et al., 2016). In order to understand orogenic mass balance and the effects of collision on topography, climate, and plate kinematics, we need well-preserved records of the transition from subduction to collision (e.g.,

21 DeCelles et al., 2014; Zhuang et al., 2015, and references therein).

22 Foreland basin stratigraphic records of collisional orogens are commonly used to
23 constrain the timing of collision (Dewey and Mange, 1999; Ding et al., 2005; Weis-
24 logel et al., 2006; Zagorevski and van Staal, 2011) via dating of events such as initial
25 arrival of upper plate detritus on a lower plate continental margin (Garzanti et al., 1987;
26 Najman et al., 2010; Hu et al., 2015; Koshnaw et al., 2019), cessation of marine sedi-
27 mentation (Garzanti et al., 1987; Najman et al., 2010), and initiation of foreland basin
28 subsidence (Ershov et al., 2003; Fakhari et al., 2008). However, interpretation of fore-
29 land basins in collisional tectonic systems is complicated by multiple factors includ-
30 ing evolving source areas (Axen et al., 2001), changing topography (Pusok and Kaus,
31 2015), and varying base levels (Krijgsman et al., 1999). Preservation of stratigraphic
32 and other (e.g., thermochronometric, structural, kinematic) records is also an issue in
33 mature collisional orogens (e.g., Hu et al., 2015). In the case of one mature collision
34 zone, the India-Asia collision, diachronous transitions in foreland basin sedimentation
35 in several studied stratigraphic sections have historically led to interpretations of colli-
36 sional ages that differed from one another by up to 10 Myr (e.g., DeCelles et al., 2004;
37 Najman et al., 2010; DeCelles et al., 2014; Hu et al., 2012, 2015; Zhuang et al., 2015;
38 Wu et al., 2014). Thus, there is an ongoing need to better understand the stratigraphic
39 record of initial collision and its spatial and temporal variation within a foreland basin
40 system.

41 The optimal setting for investigating the sedimentary response to the initiation of
42 collision is an orogen where collision began recently, so that independent constraints
43 on the structural and kinematic evolution of the orogen are available. There are several
44 examples of orogens thought to be undergoing the initial stages of collision where
45 the sedimentary response to collision could be probed, including Taiwan (e.g., Teng,
46 1990), Timor (Carter et al., 1976; Duffy et al., 2013; Tate et al., 2015), and the Caucasus
47 (Philip et al., 1989; Mumladze et al., 2015). Of these, the Caucasus is unique in that
48 the basin in between the two colliding continents is currently non-marine, permitting
49 ease of access to the foreland basin strata of interest. In addition, published marine
50 magnetic anomaly, geodetic, structural, and thermochronometric analyses constrain
51 the kinematics of the Caucasus and the surrounding region during the transition from

52 subduction to collision (Reilinger et al., 2006; Avdeev and Niemi, 2011; Kadirov et al.,
53 2012; Austermann and Iaffaldano, 2013; Kadirov et al., 2015; Cowgill et al., 2016;
54 van der Boon et al., 2018; van Hinsbergen et al., 2019; Vincent et al., 2019). The
55 goal of this study is to derive from the stratigraphic records available in the Caucasus
56 a preliminary, coupled sedimentary and kinematic framework of collision for further
57 development and comparison with other orogens.

58 In this paper, we first develop a hypothesis of the sedimentary response to the early
59 stages of collision. We then present a new detrital zircon U-Pb age dataset from the
60 Caucasus to probe erosion, sediment routing, and deposition in a natural example of
61 this phase of the plate tectonic cycle. We characterize zircon U-Pb age signatures of
62 potential sources of Cenozoic sediment by using targeted modern river samples. By
63 comparing source age signatures to detrital zircon ages in samples from three fore-
64 land basin sections distributed along strike, we investigate the dispersal of sediment
65 from upland sources into the basin between the Greater and Lesser Caucasus from
66 the Oligocene to Quaternary. We combine this zircon U-Pb age dataset with published
67 stratigraphy for the three sampled sections and published thermochronometric (Avdeev
68 and Niemi, 2011; Vincent et al., 2019), geodetic (Reilinger et al., 2006; Kadirov et al.,
69 2012, 2015; Sokhadze et al., 2018), and structural (Sobornov, 1994; Banks et al., 1997;
70 Forte et al., 2013; Cowgill et al., 2016) records to correlate sedimentary changes with
71 the structural evolution of the orogen and explore implications for collision. We also
72 discuss zircon age distributions of regional basement domains and implications for the
73 distribution of sutures along the southern margin of Eurasia, which may have guided
74 later localization of deformation.

75 **2. Hypothesized response of foreland basin sedimentation to early collision**

76 Modeling and field observations provide perspectives on possible effects of the ini-
77 tiation of collision on an orogen (Tricart, 1984; Garzanti et al., 1987; Beaumont et al.,
78 1996; Lallemand et al., 1992; Regard et al., 2003; Toussaint et al., 2004a; Gürer and
79 van Hinsbergen, 2019), from which we hypothesize the effects on sedimentation be-
80 tween the colliding continental blocks (Fig. 1). During pre-collisional subduction, an

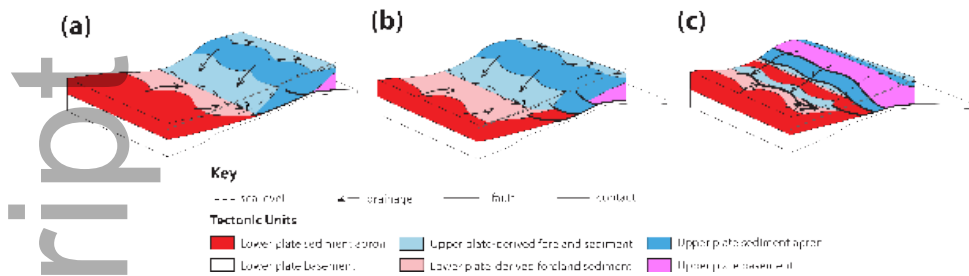


Figure 1: Effects of the transition from subduction to collision on an orogen and its foreland basin (see Section 2 for complete discussion). (a) Narrowing of an ocean basin is accommodated by subduction, resulting in the formation of an accretionary prism above the subduction zone. (b) Lower plate continental slope enters the subduction zone, resulting in accretion of lower plate stratigraphy along a new frontal thrust. (c) Further convergence drives locking of the subduction zone, increased slip on the frontal thrust, and foreland basin deformation and uplift. Foreland basin uplift causes erosion and sediment transport via a longitudinal drainage network. (c) is the current state of the western Caucasus, whereas the eastern Caucasus is in an intermediate state between (b) and (c).

81 accretionary prism may grow on the basin margin above a subduction zone, marine
 82 sedimentation occurs in the basin, and upper plate sediment may be deposited onto the
 83 lower plate as it enters the subduction zone (Fig. 1a; Karig and Sharman III, 1975).
 84 If convergence continues, the lower plate continental margin will eventually enter the
 85 subduction zone and fragments of the lower plate are likely to be accreted to the upper
 86 plate (Fig. 1b; Tricart, 1984; DeCelles et al., 2014). Further continental subduction
 87 increases lower plate thickness and buoyancy, potentially driving further accretion and
 88 accelerating upper plate rock uplift (Lallemand et al., 1992; Beaumont et al., 1996; Toussaint et al., 2004a) and narrowing and uplifting the basin between the two continents
 89 (Fig. 1c). The increasing buoyancy of the incoming lower plate may drive locking of
 90 the subduction zone megathrust and migration of shortening to pro- and retrowedge
 91 fold and thrust belts (Beaumont et al., 1996; Toussaint et al., 2004a). Increasing lower
 92 plate thickness and forward propagation of thrust belts will decrease accommodation
 93 between the two continental blocks and ultimately lead to erosive conditions in the
 94 basin along the plate boundary (e.g., DeCelles and Giles, 1996; Soria et al., 1999).

95 Along-strike variations in buoyancy, structural style, and topography of an incipient
 96 collision zone are greatly influenced by the geometry of the lower plate continent
 97

98 (e.g., Gürer and van Hinsbergen, 2019). At the initial point of contact between the
99 two colliding continents, the foreland basin is expected to undergo uplift and deforma-
100 tion. However, along-strike plate geometries may temporarily preserve lower elevation
101 marine or non-marine basins where the converging continents are not yet in contact
102 (e.g., Şengör, 1976). Tectonic uplift and closure of the foreland basin at the locus of
103 collision is likely to increase the sediment supply of longitudinal drainages that con-
104 vey sediment from the locus of collision to lower elevation sections of the basin along
105 strike (Fig. 1c; Malkowski et al., 2017). As collision continues, further shortening will
106 result in the exposure of the lower portion of the prism, and accelerated upper plate
107 rock uplift rates will lead to the exposure of deeper crustal levels (Fig. 1c; Beaumont
108 et al., 1996; Toussaint et al., 2004a).

109 The predicted responses of the foreland basin to early collision include shallowing
110 and a transition from marine to terrestrial to erosive conditions (Fig. 1b-c); erosion
111 and deposition of material from deeper crustal levels of the orogen (Fig. 1c); and
112 longitudinal drainage away from the locus of initial collision (Fig. 1c). The Caucasus
113 provides a natural setting in which to test whether these expected effects are observed
114 and to constrain the relationships between these effects and the structural and kinematic
115 changes that accompany collision.

116 **3. Geological background**

117 The Caucasus region is located on the southern margin of Eurasia, within the
118 Arabia-Eurasia collision zone (Fig. 2a). To the immediate north of the Caucasus lies
119 the Scythian platform (Natal'in and Şengör, 2005; Saintot et al., 2006b), which is bor-
120 dered to its north by stable Eurasia (Fig. 2a; Allen et al., 2006; Bogdanova et al., 2008).
121 To the south of the Caucasus lies the Turkish-Iranian plateau, which is demarcated from
122 stable Arabia to its south by the Bitlis-Zagros suture, the Arabia-Eurasia plate bound-
123 ary (Fig. 2a; Şengör and Kidd, 1979; Şengör and Yilmaz, 1981; Copley and Jackson,
124 2006).

125 The Caucasus region consists of two parallel, WNW-striking mountain ranges, the
126 Greater Caucasus (~1200 km long) and the Lesser Caucasus (~500 km long; Fig. 2b),

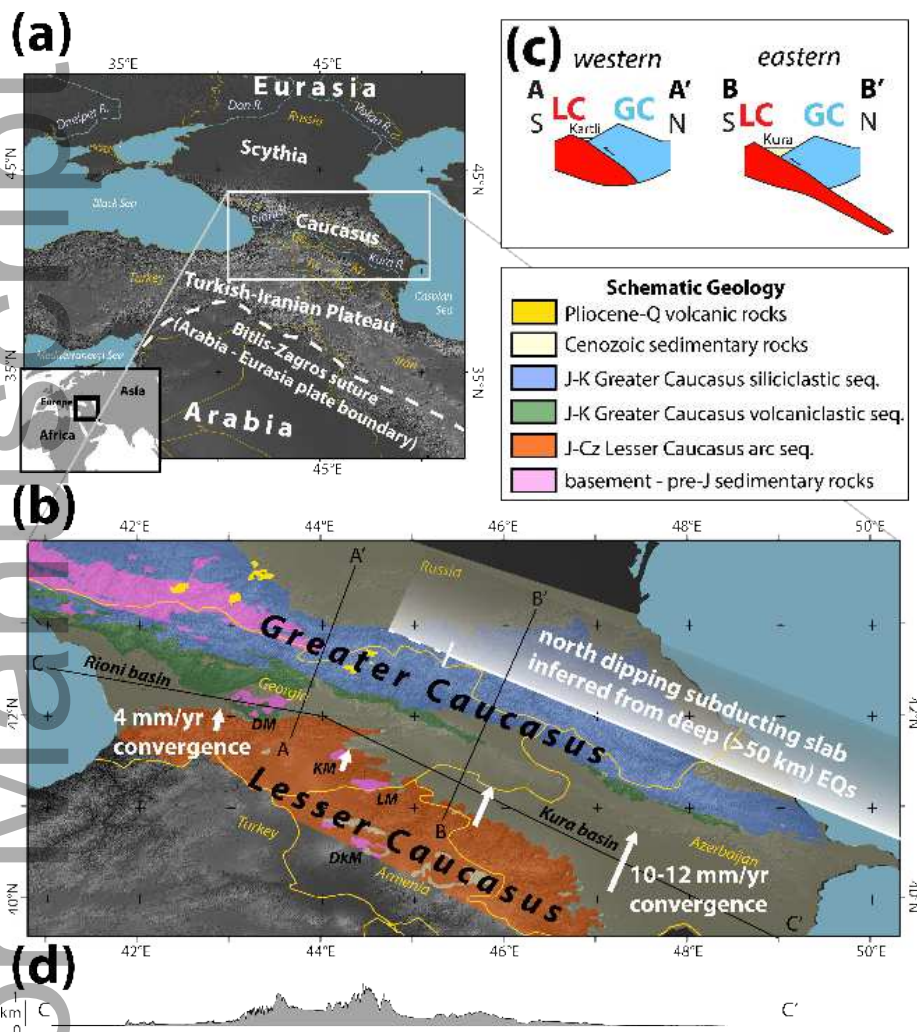


Figure 2: Location and tectonic setting of the Caucasus. (a) The Caucasus region is located on the southern margin of Eurasia in the Arabia-Eurasia collision zone. (b) The Caucasus region consists of the WNW-striking Greater Caucasus and Lesser Caucasus, which are converging toward one another. Schematic GPS convergence rates (Reilinger et al., 2006; Kadirov et al., 2015) between the Greater and Lesser Caucasus and extent of a north-dipping subducting slab inferred from deep earthquakes (Mellors et al., 2012; Mumladze et al., 2015) are shown in white. Key tectonic units are shown in color with key to the upper right (see further discussion in Section 3.2). Abbreviated names of geologic features: DM—Dzirula Massif, KM—Khrami Massif, LM—Loki Massif, DKM—Dzarkuniatz Massif. Black lines show locations of cross sections (A-A', B-B') in (c) and topographic profile (C-C') in (d). (c) Schematic cross sections across the western (A-A') and eastern (B-B') Greater Caucasus. (d) Foreland basin topographic profile along strike of the Greater Caucasus (C-C').

127 separated by a longitudinal drainage network. West of 43° E, the Greater Caucasus
128 is separated from the Lesser Caucasus by the Rioni basin, in which the Rioni River
129 flows west to the Black Sea (Fig. 2a). Between 43° E and 45° E, a contiguous band of
130 elevated topography runs between the Greater and Lesser Caucasus (Fig. 2d). East of
131 45° E, the Greater Caucasus is separated from the Lesser Caucasus by the Kura basin,
132 in which the Kura River flows east to the Caspian Sea (Fig. 2a). The current drainage
133 network of the Greater Caucasus is consistent with the final step of our conceptual
134 model of collision (Fig. 1c).

135 3.1. Tectonic setting and history

136 The present tectonic setting of the Caucasus is constrained by seismic and geodetic
137 data (Fig. 2b, c). Deep earthquakes >50 km beneath the eastern Greater Caucasus sug-
138 gest the presence of a north-dipping subducting slab beneath the range (Mellors et al.,
139 2012; Mumladze et al., 2015), and GPS convergence rates of 10-12 mm/yr accommo-
140 dated between a rigid upper and lower plate are consistent with inferences that sub-
141 duction is currently active (Reilinger et al., 2006; Kadirov et al., 2012, 2015). Seismic
142 tomography indicates the presence of a high-velocity body in the upper mantle beneath
143 the eastern Greater Caucasus interpreted as subducted or underthrust lithosphere (Sko-
144 beltsyn et al., 2014). The Lesser Caucasus mountains are on the lower plate of this
145 subduction system, and the Kura basin separates the eastern Greater Caucasus from
146 the Lesser Caucasus and its eastern extension, the Talysh (Fig. 2b, c). In the west-
147 ern Greater Caucasus, range-normal GPS convergence rates of 3-4 mm/yr (Reilinger
148 et al., 2006; Kadirov et al., 2015; Sokhadze et al., 2018), rapid exhumation (Avdeev
149 and Niemi, 2011; Vincent et al., 2019), and contiguous elevated topography between
150 the Greater and Lesser Caucasus (Fig. 2d) suggest that this part of the range is cur-
151 rently undergoing collision with the Lesser Caucasus (Fig. 2b, c). The combination of
152 ongoing collision inferred in the western Caucasus and active subduction in the eastern
153 Caucasus suggests that the orogen is transitioning diachronously from subduction to
154 collision, with the western part of the range at a more advanced stage of this transition
155 than the eastern part of the range (Fig. 2b; Mumladze et al., 2015). Active fold and
156 thrust belts are located on both the pro- (Banks et al., 1997; Forte et al., 2010, 2013)

157 and retro-wedge (Sobornov, 1994, 1996) sides of the orogen.

158 The Caucasus region has a complex deformation history. The southern Eurasian
159 margin was affected by successive episodes of subduction, terrane accretion, and rift-
160 ing throughout the Phanerozoic that are thought to have generated significant litho-
161 spheric heterogeneity in the region (e.g., Şengör, 1984; Stampfli, 2013). The regional
162 pre-Jurassic tectonic history remains uncertain, in part due to the lack of exposure of
163 rocks old enough to record this history (e.g., Natal'in and Şengör, 2005; Saintot et al.,
164 2006b). Most of the exposed bedrock in the Greater and Lesser Caucasus was deposited
165 in an intra-arc or backarc basin environment during Jurassic to Eocene time (Fig. 2b;
166 Nalivkin, 1976; Alizadeh et al., 2016). During this period, the Lesser Caucasus consti-
167 tuted an active volcanic arc that extended west into the Pontides and east into Iran above
168 the north-dipping subducting slab of Neotethys (e.g., Sosson et al., 2010; Rolland et al.,
169 2011; Adamia et al., 2011b). Concomitant with subduction and arc volcanism, a sys-
170 tem of backarc and forearc basins opened parallel to the arc, including the Black Sea
171 basins, the South Caspian basin, and the Greater Caucasus basin, which opened to the
172 north of the Lesser Caucasus and is where most of the sedimentary bedrock presently
173 exposed in the Greater Caucasus was originally deposited (e.g., Zonenshain and Le Pi-
174 chon, 1986; Adamia et al., 2011b; Vincent et al., 2016; van Hinsbergen et al., 2019).

175 Extant basins that opened during this period are inferred to be floored by oceanic crust
176 (Knapp et al., 2004; Nikishin et al., 2015) or transitional crust with a composition simi-
177 lar to mafic lower continental crust (Mangino and Priestley, 1998). The composition of
178 the basement of the Greater Caucasus basin is poorly constrained and is the subject of
179 controversy, with both an oceanic composition and a thinned, mafic continental com-
180 position having been hypothesized (Cowgill et al., 2016; Vincent et al., 2016, 2018;
181 Cowgill et al., 2018). Structural shortening estimates (Trexler, 2018) and lower plate
182 oroclinal bending estimates (van der Boon et al., 2018) indicating 230-280 km of short-
183 ening accommodated within the Greater Caucasus suggest that the Greater Caucasus
184 basin was originally of comparable width (at minimum) to the extant Black Sea and
185 Caspian Sea basins. Thus, an analogous basement, of thickness 8 - 20 km and com-
186 position similar to oceanic crust or mafic lower crust, is likely (Mangino and Priestley,
187 1998; Knapp et al., 2004; Nikishin et al., 2015).

188 The late Eocene to present history of the region reflects convergence of the Greater
189 and Lesser Caucasus toward one another and closure of the intervening basin. Be-
190 ginning in latest Eocene to earliest Oligocene time, the Greater Caucasus basin be-
191 gan to close by northward subduction/underthrusting, leading to the formation of the
192 Greater Caucasus as a compressive orogen/accretionary prism (e.g., Dotdjev, 1986;
193 Philip et al., 1989; Khain et al., 2007; Vincent et al., 2007; Adamia et al., 2011b; Forte
194 et al., 2014; Alizadeh et al., 2016; Cowgill et al., 2016; Kangarli et al., 2018). The com-
195 plete closure of the backarc basin(s) that separated the Lesser Caucasus from Eurasia
196 was marked by the collision of the Lesser Caucasus arc terrane with the Greater Cauca-
197 sus, the age of which is controversial (Vincent et al., 2016; Cowgill et al., 2016; Vincent
198 et al., 2018; Cowgill et al., 2018). Burial histories suggest that flexural subsidence to
199 the north of the Greater Caucasus was active during Late Miocene to Quaternary times,
200 suggesting significant orogenic growth during that period (Ershov et al., 2003). Pro-
201 and retro-wedge fold and thrust belts began to deform during Late Miocene time, with
202 major deformation occurring in the Pliocene to Quaternary (Sobornov, 1994; Banks
203 et al., 1997; Forte et al., 2013, 2014). Exhumation rates in the western Greater Cau-
204 casus increased by a factor of ten around 7-5 Ma (Avdeev and Niemi, 2011; Vincent
205 et al., 2019), coincident with slowing of Arabia-Eurasia convergence (Austermann and
206 Iaffaldano, 2013) and kinematic reorganization of the Arabia-Eurasia plate boundary
207 (Allen et al., 2004). These coinciding structural and kinematic changes have led to
208 the hypothesis that collision began at ~5 Ma in the western Greater Caucasus and may
209 have affected strain accommodation within the broader Arabia-Eurasia collision zone
210 (Cowgill et al., 2016). An alternative hypothesis for the Eocene to present evolution
211 of the region is that collision between the Greater and Lesser Caucasus was largely
212 complete by 34 Ma (Vincent et al., 2016). The provenance data presented here have
213 implications for the timing of collision.

214 3.2. *Potential sources of Cenozoic foreland basin sediment*

215 The Caucasus and surrounding regions contain three distinct domains of igneous
216 and metamorphic basement and four distinct tectonostratigraphic sedimentary sequences
217 that may have contributed sediment to the basin between the Greater and Lesser Cauca-

218 sus during convergence and collision. Here, we outline these sources and their potential
219 contribution to Caucasus Cenozoic foreland basin sediment.

220 Three distinct basement domains are potential sedimentary sources for Cenozoic
221 Caucasus foreland basins: the Eurasian interior (consisting of the East European Cra-
222 ton and Urals), the Greater Caucasus basement, and the Transcaucasus basement. The
223 Archean to Neoproterozoic crust of the East European Craton (Bogdanova et al., 2008)
224 forms the core of the Eurasian interior at the longitude of the Caucasus and contributes
225 sediment to rivers that drain into the Black and Caspian seas (Fig. 2a; Allen et al.,
226 2006; Wang et al., 2011). The East European Craton may also have contributed sedi-
227 ment to the Cenozoic foreland basin of the Caucasus (Allen et al., 2006). Some rivers
228 that drain the East European Craton also include the Urals in their watershed, so sedi-
229 ment sourced from the Eurasian interior may also include detritus from the Paleozoic
230 Ural orogen (Allen et al., 2006). The second potential basement source is a predomi-
231 nantly late Paleozoic (Hercynian) arc assemblage that constitutes the basement of the
232 Greater Caucasus (Adamia et al., 2011b; Somin, 2011). This arc assemblage is exposed
233 in the core of the western portion of the Greater Caucasus (Fig. 2b; Nalivkin, 1976).
234 The third potential suite of basement sources is the isolated Precambrian to Paleozoic
235 massifs of the Transcaucasus and South Armenian Block, which together lie both be-
236 tween the Greater and Lesser Caucasus and within the Lesser Caucasus (the Dzirula,
237 Khrami, Loki, and Dzarkuniatz massifs of the Transcaucasus are shown in Fig. 2b;
238 Nalivkin, 1976; Knipper and Khain, 1980; Aghamalyan, 1998; Zakariadze et al., 2007;
239 Gamkrelidze and Shengelia, 2007; Mayringer et al., 2011; Rolland et al., 2016).

240 Four tectonostratigraphic sequences in the Caucasus may have contributed sedi-
241 ment to the Cenozoic foreland basin. The oldest sequence is Paleozoic to Triassic in
242 age and does not overlap the ages of the other, younger sequences. The Paleozoic
243 to Triassic sequence is marine and consists of shales, sandstones, and carbonates that
244 are locally found in depositional or structural contact with the Transcaucasus basement
245 and the southern margin of the Greater Caucasus basement (see reviews in Khain, 1975;
246 Adamia et al., 1981; Şengör et al., 1984). Exposures of this sequence immediately to
247 the south of the Greater Caucasus basement are called the Dizi Series (Adamia et al.,
248 2011b; Somin, 2011; Vasey et al., 2020). Paleozoic to Triassic sedimentary rocks are

249 exposed over only a minor area within the Caucasus.

250 The three tectonostratigraphic sequences that constitute the vast majority of ex-
251 posed bedrock in the Caucasus are contemporaneous sequences of predominantly Juras-
252 sic to Cretaceous strata that are markedly different in composition and sedimentology.

253 These three sequences, which we describe here in order of exposure from south to
254 north, are thought to have been deposited on the flanks of the Lesser Caucasus arc and
255 in the Greater Caucasus basin (e.g., Nalivkin, 1976; Zonenshain and Le Pichon, 1986;
256 Saintot et al., 2006a; Sosson et al., 2010; Rolland et al., 2011; Vincent et al., 2016).

257 The southernmost of the three sequences, the Jurassic to Eocene Lesser Caucasus arc
258 sequence is exposed in the Lesser Caucasus and includes calc-alkaline volcanic, vol-
259 caniclastic, and carbonate strata intruded by Jurassic to Eocene plutons that reflect vol-
260 canic arc activity in the Lesser Caucasus (Fig. 2b; Nalivkin, 1976; Kopp and Shcherba,
261 1985; Sosson et al., 2010; Rolland et al., 2011; Sahakyan et al., 2017). Exposed on the
262 southern slope of the Greater Caucasus is the Jurassic to Cretaceous Greater Cauca-
263 sus volcanoclastic sequence, which includes a thick sequence of mafic to intermediate
264 volcanic and volcanoclastic strata and carbonates with local Jurassic intrusions (Fig.
265 2b; Nalivkin, 1976; Mengel et al., 1987; Kopp, 1985). The Greater Caucasus volcani-
266 clastic sequence is thought to have been deposited in the Greater Caucasus basin (e.g.,
267 Vincent et al., 2016). Within the Greater Caucasus, to the north of the volcanoclastic
268 sequence, is a Jurassic to Cretaceous sequence dominated by marine sandstones and
269 shales (Fig. 2b; e.g., Saintot et al., 2006a; Bochud, 2011; Vincent et al., 2013). We
270 term this sequence the Greater Caucasus siliciclastic sequence in order to differentiate
271 it from the Greater Caucasus volcanoclastic sequence, although some carbonates are
272 present. The sedimentary architecture of the Greater Caucasus siliciclastic sequence,
273 inferred from seismic data, suggests the sequence is derived from north of the Greater
274 Caucasus (Sholpo, 1978). Because the Lesser Caucasus arc sequence, Greater Cauca-
275 sus volcanoclastic sequence, and Greater Caucasus siliciclastic sequence together ac-
276 count for the majority of exposed bedrock in the Caucasus today (Fig. 2b), they are
277 anticipated to have been significant sources for Oligocene to Quaternary foreland basin
278 sedimentation.

279 **4. Methods**

280 We report 29 new detrital zircon U-Pb age samples (Table S1) from Cenozoic sand-
281 stones and modern river sands comprising 7,090 total ages (Table S2). Mineral sepa-
282 ration was conducted at the University of Michigan. Heavy mineral fractions were
283 mounted in epoxy and polished to expose crystal interiors. Mounts were made of en-
284 tire heavy mineral fractions, rather than hand selected individual zircon grains, in order
285 to ensure that representative random samples of zircon were analyzed. Mount imaging
286 was conducted at the University of Michigan and the University of Arizona Laserchron
287 Center. U-Pb analyses were conducted at the University of Arizona Laserchron Center
288 using a laser ablation system attached to a Thermo Element 2 single collector ICP-MS
289 (Gehrels et al., 2008; Pullen et al., 2014). Analyses > 20% discordant are excluded
290 from further interpretation. Where practical, we analyzed at least 300 zircon grains
291 per sample, which provides more robust characterization of zircon age signatures than
292 analyses with typical ($n \sim 100$) sample sizes (Pullen et al., 2014).

293 *4.1. Sampling*

294 Understanding provenance changes during the evolution of an orogen (Fig. 1) re-
295 quires characterizing the zircon age signature of potential source areas and quantifying
296 the contribution of those sources to foreland basin deposits. We use 16 new samples
297 of modern river sands from targeted catchments that contain specific bedrock ages
298 and lithologic types, along with published modern and bedrock detrital zircon sam-
299 ples (Allen et al., 2006; Wang et al., 2011; Cowgill et al., 2016; Vasey et al., 2020),
300 to characterize the zircon age signatures of the potential source areas (Figs. 3, 4, 5),
301 as described in the previous section. Using modern river sand samples to characterize
302 potential sources is an efficient way to capture well-mixed, representative zircon age
303 signals associated with erosion from the source area (Fig. 3). This method assumes
304 that present exposures are representative of those that contributed sediment earlier in
305 the Cenozoic (e.g., the Jurassic sandstones presently exposed in the range yield the
306 same detrital zircon age distribution as Jurassic sandstones exposed in the Cenozoic),
307 which we view as realistic given the age ranges of exposed bedrock and the structural
308 style of the Caucasus.

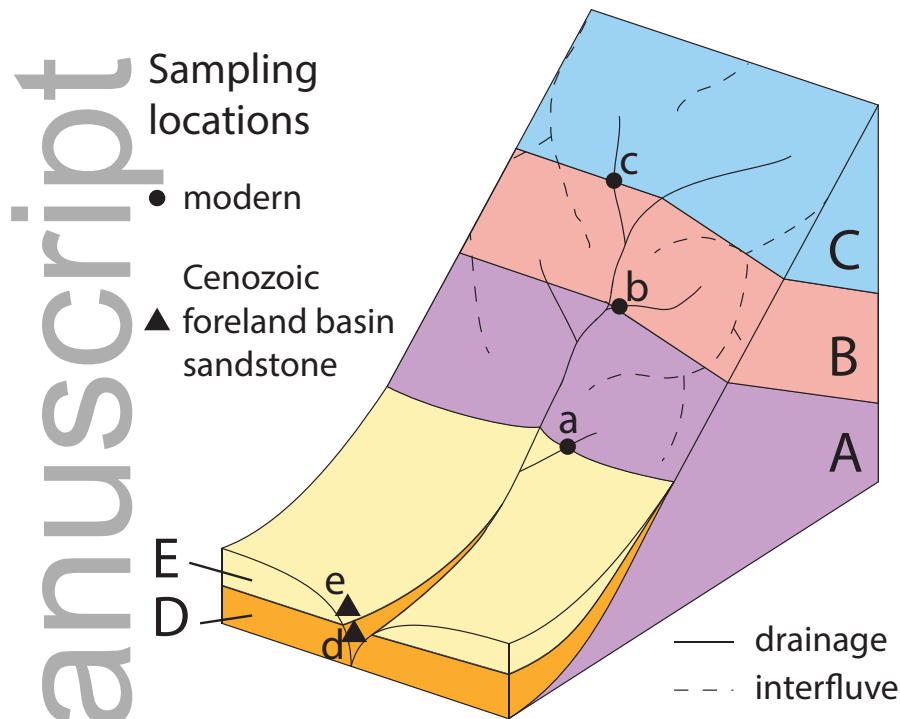


Figure 3: We use a two part detrital zircon sampling strategy to understand the evolution of foreland basin sediment provenance. We collect samples of foreland basin strata (rock samples d, e, of foreland basin units D, E), and we use modern samples of targeted catchments to characterize potential sources contributing to the sampled foreland strata (river sands at locations a, b, and c provide detrital zircon age signatures of units A, B, C, respectively).

309 In order to understand the changing sources of foreland basin sediment over time,
 310 we compare the zircon age signatures of potential sources to the zircon age distributions
 311 of Cenozoic foreland basin rock samples (Fig. 3). We report 13 new samples taken
 312 from different stratal levels of three Cenozoic foreland basin sections (western, central,
 313 and eastern sections) located on the southern margin of the Greater Caucasus (Figs. 4,
 314 5, 6). Five new samples were analyzed from the western foreland basin section from
 315 rocks of Oligocene to Quaternary age (Fig. 5b). Two samples were analyzed from
 316 the central foreland basin section of Middle Miocene and Late Miocene age (Fig. 5a).
 317 Six samples were analyzed from the eastern foreland basin section, including rocks
 318 of Cretaceous-Paleocene to Pliocene age (Fig. 5c). In addition to these new samples,

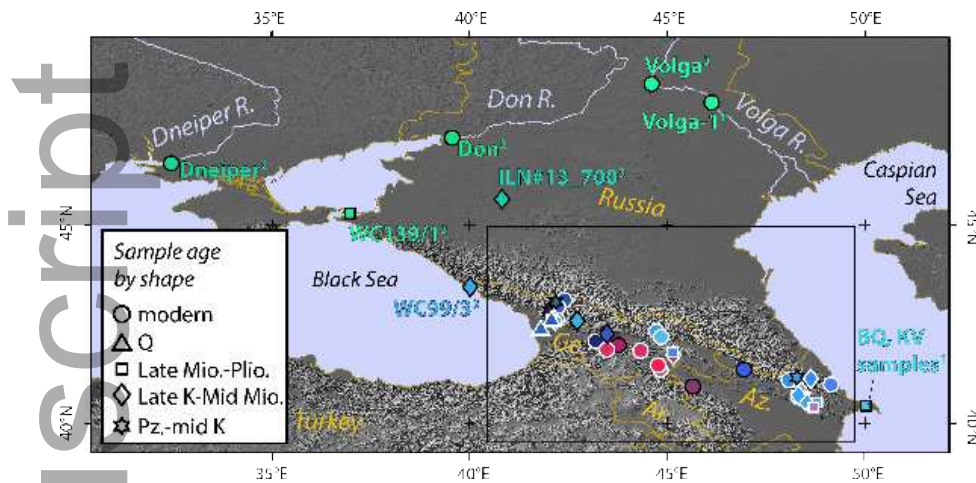


Figure 4: New and published sampling covers the Eurasian interior, the Greater Caucasus, and the Lesser Caucasus. Sample colors indicate affinity to the Eurasian interior (green), the Greater Caucasus (blue), or the Lesser Caucasus (red); see Figure 7 and Section 4.2 for details. Symbol outlines are white for new samples and black for previously published samples. Black rectangle shows the extent of Figure 5a. Samples outside this rectangle have their names displayed and are superscripted according to source publication: 1–Allen et al. (2006); 2–Wang et al. (2011); 3–Vincent et al. (2013). Samples inside the rectangle have their names displayed in Figure 5. Abbreviations for geologic time are as follows: Pz–Paleozoic, K–Cretaceous, Pg–Paleogene, Mio–Miocene, Plio–Pliocene, Q–Quaternary

319 our analyses are integrated with five foreland basin samples from the western Greater
 320 Caucasus (Vincent et al., 2013) and four samples from a Pliocene section at the far
 321 eastern extent of the Greater Caucasus (Allen et al., 2006).

322 4.2. Data visualization

323 Throughout the paper, samples are colored by comparison to three endmember
 324 samples using the Bayesian Population Correlation (BPC) metric (Tye et al., 2019).
 325 BPC values range from 0 to 1 based on the likelihood that two sampled populations are
 326 the same versus different, with values closer to 1 indicating greater population corre-
 327 spondence (Tye et al., 2019). The three endmember samples were chosen because they
 328 highlight first order age distinctions among the potential sources: the Eurasian interior
 329 (represented by sample Volga; Wang et al., 2011) is dominated by Proterozoic zircon
 330 ages, the Greater Caucasus (represented by sample EGC-4) contains predominantly
 331 Paleozoic zircon ages (Adamia et al., 2011b; Somin, 2011), and the Lesser Caucasus

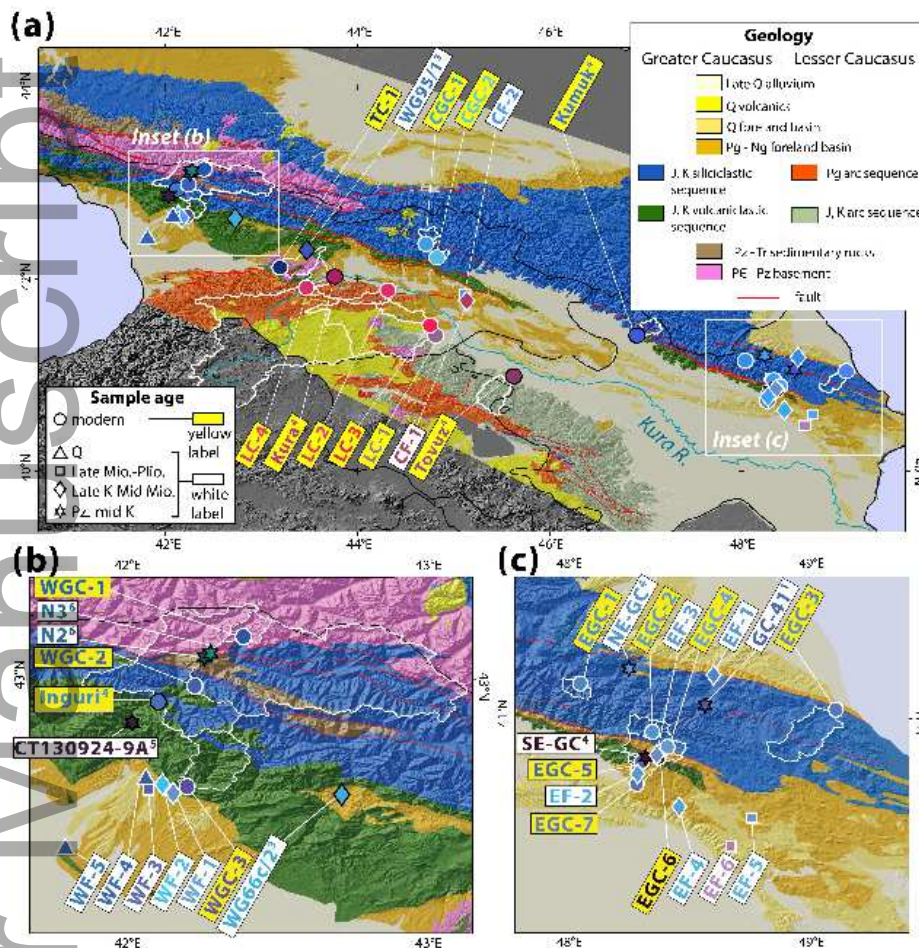


Figure 5: Simplified geology and detrital zircon sample locations in the Greater and Lesser Caucasus. Sample names are shown in rectangles and sample colors are as in Figure 4. Political boundaries are shown in black and catchment boundaries of modern samples are shown in white. The Kura River is shown in blue in (a). Abbreviations for geologic time are as follows: PC–Precambrian, Pz–Paleozoic, Tr–Triassic, J–Jurassic, K–Cretaceous, Pg–Paleogene, Ng–Neogene, Mio–Miocene, Plio–Pliocene, Q–Quaternary. For samples not from this study, sample name superscripts reflect source publication as in Figure 4 with three additions: 4–Cowgill et al. (2016), 5–Trexler (2018), 6–Vasey et al. (2020).

332 (represented by LC-3) is characterized by Jurassic to Eocene zircon ages (e.g., Sosson
 333 et al., 2010). These three endmembers were chosen because they are broadly represen-
 334 tative of samples from their respective source areas and because they have large sample
 335 sizes ($n \sim 300$), where available. The coloring scheme works as follows: each sample

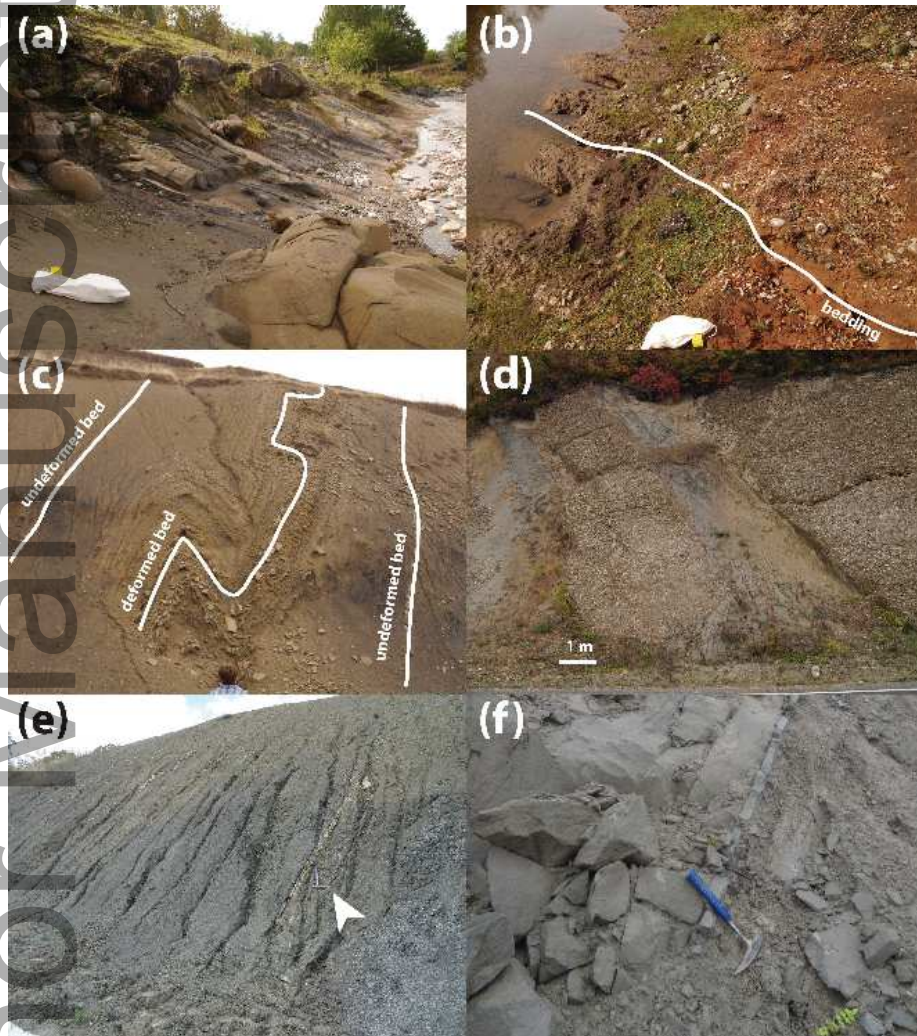


Figure 6: Photographs of sampled Cenozoic foreland basin strata. (a) Oligocene, Early Miocene, or Middle Miocene sandstones and organic-rich shales of the western foreland basin section (sample WF-2). (b) Latest Pliocene conglomerate of western section, bedding marked in white (sample WF-3). (c) Middle Miocene organic-rich sandstone-shale sequence of the central section with bedding of undeformed and deformed horizons marked in white (sample CF-1). (d) Late Miocene conglomerate of the central section (sample CF-2). (e) Oligocene or Early Miocene sandstone and shale of the eastern section with arrow indicating rock hammer for scale (sample EF-4). (f) Pliocene sandstone of the eastern section (sample EF-6).

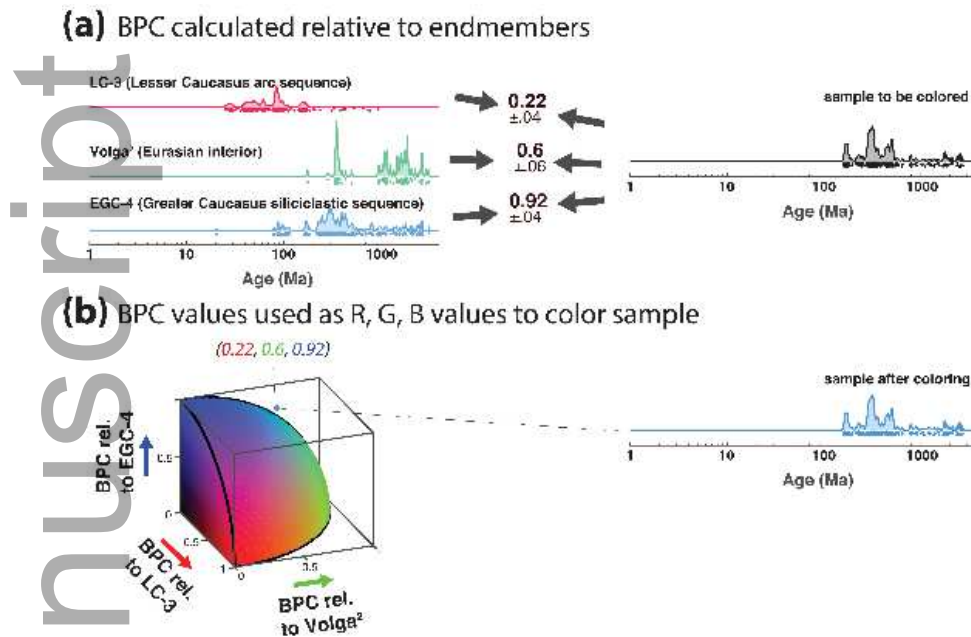


Figure 7: Throughout this paper, samples are colored according to their BPC value (Tye et al., 2019) relative to three representative endmembers of the Eurasian interior, Greater Caucasus, and Lesser Caucasus. (a) BPC values are calculated between each sample and the three endmember samples. (b) Calculated BPC values are used as R, G, B values for coloring each sample. The colored surface shown is a visual aid; samples do not need to fall on this surface.

336 is assigned an RGB triplet where the red value is equal to the BPC value of the sample
 337 compared to the Lesser Caucasus endmember, the green value comes from comparison
 338 to the Eurasian interior endmember, and the blue value comes from comparison to the
 339 Greater Caucasus endmember (Fig. 7).

340 5. Source area detrital zircon signatures

341 5.1. Detrital zircon age signatures of potential sources for Caucasus Cenozoic sedi- 342 ment

343 In order to use detrital zircon data from foreland basin deposits to understand the
 344 Cenozoic tectonic history of the Caucasus, we must first characterize the zircon age
 345 signatures of potential sediment sources for the foreland basin deposits. In this section,

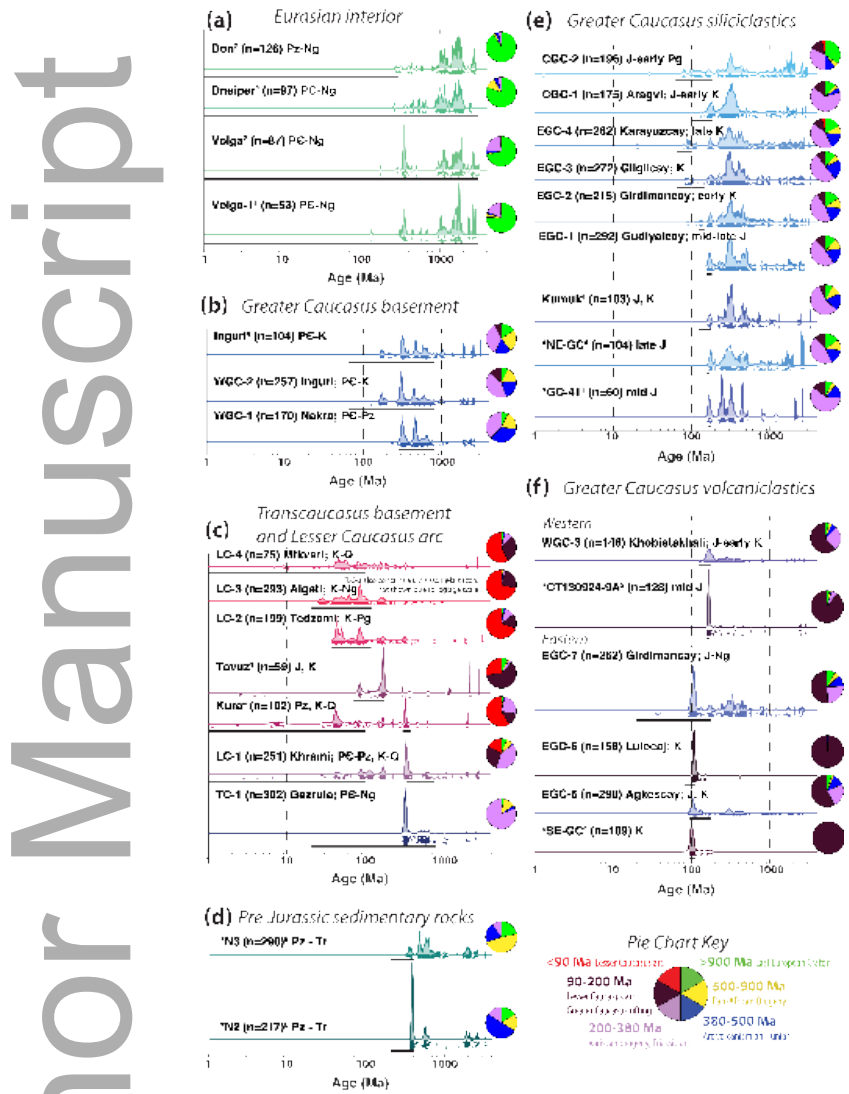


Figure 8: Detrital zircon age signatures from targeted modern river samples from (a) the Eurasian interior, (b) the Greater Caucasus basement, (c) the Transcaucasus basement and Lesser Caucasus arc sequence, (d) pre-Jurassic sedimentary rocks, (e) the Greater Caucasus siliciclastic sequence, and (f) the Greater Caucasus volcaniclastic sequence. Published modern river and bedrock samples from these sources are also shown (bedrock samples are marked with an asterisk *). Each sample is labeled with the sample name, sample size, name of modern river sampled (if applicable/available) and its age or the ages of strata within the sampled catchment (Nalivkin, 1976; Asch et al., 2005). The plot of each sample shows a probability density plot (Hurford et al., 1984) as a solid line, a kernel density estimate (Silverman, 1986; Shimazaki and Shinomoto, 2010; Vermeesch, 2012) as a shaded area, age observations ignoring analytical uncertainty as a band of dots beneath the curves (vertical scatter for visual clarity), and a black bar that shows the age of the sample (for bedrock samples) or the ages of bedrock strata within the sampled catchment (for modern samples). A pie chart of ages is shown to the right of each sample, as outlined in the key (see Section 7 for interpretation of ages). Previously published samples are marked with a superscript, corresponding to references as in Figures 4, 5. Age abbreviations are as Figure 5, plus: Ng–Neogene. The Greater Caucasus volcaniclastic sequence

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19

The eastern portion consisting of Jurassic strata and the eastern portion consisting largely of Cretaceous strata. Samples are colored as shown in Figure 7, and are arranged by region. Empirical cumulative distribution functions of these samples are shown in Figure S1

we discuss the zircon age distributions that distinguish seven potential sources (Figs. 8, S1) that outcrop within the Caucasus and surrounding region (Figs. 4, 5). Three of these sources are regional basement domains (the Eurasian interior, the Greater Caucasus basement, and the Transcaucasus basement). One potential source suite is the pre-Jurassic sedimentary sequences that crop out over small areas adjacent to Greater Caucasus and Transcaucasus basement outcrops. Three sources are Jurassic to Eocene tectonostratigraphic sequences (the Lesser Caucasus arc, Greater Caucasus siliciclastic, and Greater Caucasus volcanoclastic sequences). We first discuss the basement domain sources.

Three distinct basement domain sources can be distinguished by their detrital zircon age signatures: the Eurasian interior (includes the East European Craton and Urals; Figs. 8a, S1a), the crystalline basement exposed in the Greater Caucasus (Figs. 8b, S1b), and the basement massifs of the Transcaucasus (Figs. 8c, S1c). Modern samples from rivers that drain the Eurasian interior contain at least 70% zircon ages >900 Ma, which are associated with the East European craton (Fig. 8a; Allen et al., 2006; Bogdanova et al., 2008). Some samples representing the Eurasian interior also contain a subordinate peak at ~360 Ma derived from the Urals (Allen et al., 2006). Rivers that drain the Eurasian interior contain very few Mesozoic zircon grains and no Cenozoic zircon grains. Detritus of the Greater Caucasus basement (Fig. 8b) is primarily identifiable by concentrated age peaks centered on 300 Ma and 450 Ma. Scattered Neoproterozoic to Middle Paleozoic ages are also present in the Greater Caucasus basement rocks, defining a broad age peak centered on 600 Ma (Fig. 8b). Transcaucasus basement massifs are targeted by sample TC-1 and are also included in the catchments of samples LC-1 and Kura (Figs. 5a; 8c). Pre-Mesozoic ages in these samples are dominated by a single peak at ~300 Ma (Fig. 8c). Samples derived from Transcaucasus basement massifs also contain scattered Neoproterozoic to Paleozoic ages that define a broad peak near 600 Ma (Fig. 8c). The distinguishing detrital zircon age characteristics of the three basement domain sources are that the Eurasian interior is the only source of abundant zircon ages >900 Ma, the Greater Caucasus basement contains large, subequal zircon age peaks at ~300 Ma and ~450 Ma, and the Transcaucasus basement massifs contain only one major age peak, at ~300 Ma (Table 1).

377 One potential sediment source in the Caucasus is a set of Late Paleozoic to Trias-
378 sic, fine-grained clastic to carbonate sedimentary successions exposed over small areas
379 adjacent to the Greater Caucasus basement and Transcaucasus basement massifs (Figs.
380 5b, 8d, S1d; Khain, 1975; Şengör et al., 1984; Adamia et al., 2011b; Vasey et al., 2020).
381 One of these successions, the Dizi Series, is located directly to the south of the Greater
382 Caucasus basement (Fig. 8d; Khain, 1975; Şengör et al., 1984; Adamia et al., 2011b).
383 Detrital zircon age spectra from the Dizi Series are characterized by an age peak be-
384 tween ~500 and 800 Ma, scattered Archaean to Paleoproterozoic ages, and in one case,
385 a 380 Ma age peak that accounts for >60% of measured ages (Fig. 8d; samples N2 and
386 N3; Vasey et al., 2020). The detrital zircon U-Pb age signatures of samples N2 and N3,
387 two bedrock samples from the Dizi Series, differ markedly from modern samples that
388 include the Dizi Series and other Paleozoic to Triassic successions within their source
389 catchments (see samples Inguri, WGC-2, and LC-1; Figs. 5, 8), suggesting the sig-
390 natures of samples N2 and N3 are not effectively propagated through the sedimentary
391 system. In addition, the signatures of N2 and N3 are different from all foreland basin
392 samples, as we later show. The lack of propagation of the Dizi Series age signatures
393 is likely due to the fine-grained clastic and carbonate strata that dominate Paleozoic
394 to Triassic sedimentary sequences on the southern slope of the Greater Caucasus and
395 within the Transcaucasus/Lesser Caucasus (Khain, 1975; Adamia et al., 2011b), and
396 may also be due to the small exposure area of these successions compared to other
397 potential sedimentary sources in the Caucasus (Fig. 5). Because the detrital zircon age
398 signatures of samples N2 and N3 appear not to be effectively propagated through the
399 sedimentary system, it is not possible to use detrital zircon ages to determine whether
400 the pre-Jurassic sequences they represent contributed sediment to the Cenozoic fore-
401 land basin.

402 The final three sources we characterize are three Jurassic to Eocene tectonostrati-
403 graphic packages that outcrop over large areas in the Caucasus region (Fig. 5): the
404 Lesser Caucasus arc sequence (Figs. 8c, S1c), Greater Caucasus siliciclastic sequence
405 (Figs. 8e, S1e), and Greater Caucasus volcanoclastic sequence (Figs. 8f, S1f). Samples
406 derived from the Lesser Caucasus arc sequence can be recognized by the ubiquity of
407 zircon ages 90 Ma and younger (Fig. 8c), which are virtually absent in other potential

408 sources. Lesser Caucasus arc sequence samples also contain an age peak centered on
409 170 Ma. Samples of the Greater Caucasus siliciclastic sequence (Fig. 8e) share two
410 major zircon age peaks with the Greater Caucasus basement (~300 Ma and ~450 Ma),
411 though in the Greater Caucasus siliciclastic sequence these age peaks are wider than
412 in the Greater Caucasus basement. Discordance does not appear to be systematically
413 greater in Greater Caucasus siliciclastic sequence samples than in Greater Caucasus
414 basement samples (Fig. S3), so the increased scatter in the ~300 Ma and ~450 Ma age
415 peaks in the Greater Caucasus siliciclastic samples is likely to truly reflect age scatter
416 in the source area for the Greater Caucasus siliciclastic sequence. Additional age pop-
417 ulations present in some or all Greater Caucasus siliciclastic sequence samples include
418 Permian to Triassic ages, either on the margin of a ~300 Ma peak or as a separate peak;
419 a ~170 Ma zircon age peak; scattered Precambrian to Paleozoic ages ranging from 3 Ga
420 to 500 Ma; and small quantities of ~100 Ma zircon ages (Fig. 8e). The Greater Cauca-
421 sus volcanoclastic sequence yields largely unimodal detrital zircon age samples, which
422 are centered on 170 Ma in the western Greater Caucasus and 105 Ma in the eastern
423 Greater Caucasus (Fig. 8f). Samples that represent the Greater Caucasus volcanoclas-
424 tic sequence and that also contain appreciable quantities of other age peaks (samples
425 WGC-3, EGC-5, EGC-7) come from catchments that include both Greater Caucasus
426 volcanoclastic strata and Greater Caucasus siliciclastic strata. Detrital zircon age sig-
427 natures of the three Jurassic to Eocene tectonostratigraphic sequences in the Caucasus
428 can be distinguished by the fact that the Lesser Caucasus arc sequence contains plenti-
429 ful zircon ages <90 Ma, the Greater Caucasus siliciclastic sequence contains wide age
430 peaks centered on 300 and 450 Ma, and the Greater Caucasus volcanoclastic sequence
431 yields unimodal zircon U-Pb age peaks at 170 and 105 Ma (Table 1).

432 The zircon age signature characteristics described above permit the discrimina-
433 tion of six different potential sources for Caucasus foreland basin sediment (Table 1).
434 These sources include the Eurasian interior, Greater Caucasus basement, Transcau-
435 casus basement, the Lesser Caucasus arc sequence, the Greater Caucasus siliciclastic
436 sequence, and the Greater Caucasus volcanoclastic sequences. Because these sources
437 have distinct detrital zircon U-Pb age spectra, their unique provenance signatures can
438 be distinguished in foreland basin stratigraphic sequences.

Potential source	Map Unit(s)	Notable age peaks	Notes
Eurasian interior	None (Eurasian interior is north of map area)	>900 Ma	Likely ultimate source for most ages >900 Ma in study area
Greater Caucasus basement	PC - Pz basement (Greater Caucasus)	300 Ma, 450 Ma	300 Ma, 450 Ma age peaks subequal, narrow
Transcaucasus basement	PC - Pz basement (south of Greater Caucasus)	300 Ma	no peak at 450 Ma
Paleozoic to Triassic sedimentary sequences	Pz - Tr sedimentary rocks	500-800 Ma, 380 Ma	Not a significant contributor to foreland samples
Lesser Caucasus arc sequence	J, K arc sequence; Pg arc sequence	<90 Ma, 170 Ma	Likely ultimate source for most ages <90 Ma in study area
Greater Caucasus siliciclastic sequence	J, K siliciclastic sequence	300 Ma, 450 Ma	Age peaks wide with scattered ages throughout Paleozoic
Greater Caucasus volcanoclastic sequences	J, K volcanoclastic sequence	170 Ma or 105 Ma	Unimodal

Table 1: Diagnostic detrital zircon age signatures of potential sources of Cenozoic foreland basin strata in the Caucasus. Map Unit(s) column shows corresponding units on Figure 5.

439 **6. Foreland basin zircon U-Pb characteristics and provenance interpretation**

440 We use detrital zircon U-Pb age distributions from foreland basin sedimentary sec-
441 tions, in combination with the source signatures outlined above, to infer which sources
442 contributed sediment to the foreland basin and changes in provenance over time. Here,
443 we describe the zircon age distributions of new and previously published samples from
444 foreland basin sedimentary strata deposited during Cenozoic time in the basin between
445 the Greater and Lesser Caucasus (Figs. 9, S2). In describing these age distributions,
446 we discuss sample composition and sedimentology (Fig. 6 shows photos of selected
447 sampled lithologies) and the stratigraphic context of the samples (Fig. 10). We also
448 compare foreland basin zircon age signatures with both the sources discussed above
449 and published datasets and discuss the implications for source exposure and sediment
450 routing systems. New foreland samples were collected from three sedimentary sec-
451 tions (western, central, and eastern) that were deposited in Paleogene to Quaternary
452 time (Figs. 5, 9). We also discuss published samples from a Pliocene section at the
453 far eastern extent of the range (Allen et al., 2006), as well as a set of previously pub-
454 lished samples that are distributed over a wide area of the western Greater Caucasus
455 (Vincent et al., 2013). Age constraints for our samples are based on published geo-
456 logic mapping (Edilashvili, 1957; Dzhanlidze and Kandelaki, 1957; Gamkrelidze and
457 Kakhadze, 1959; Voronin et al., 1959; Khain and Shardanov, 1960; Mekhtiev et al.,
458 1962; Nalivkin, 1976) unless otherwise noted, and published age constraints are used
459 for previously published samples. The zircon age distributions are discussed roughly
460 in order from west to east, beginning with the previously published distributed samples
461 in the western portion of the range (Vincent et al., 2013) and proceeding with our new
462 western, central, and eastern sampled sections, followed by the published far eastern
463 section (Allen et al., 2006, Fig. 9).

464 *6.1. Distributed foreland basin samples of the western Greater Caucasus*

465 Previously published Cenozoic samples from the western Greater Caucasus include
466 five samples from early Oligocene to latest Miocene/earliest Pliocene time (Figs. 9a,
467 S2a; Vincent et al., 2013). This group of five samples includes two samples that lie

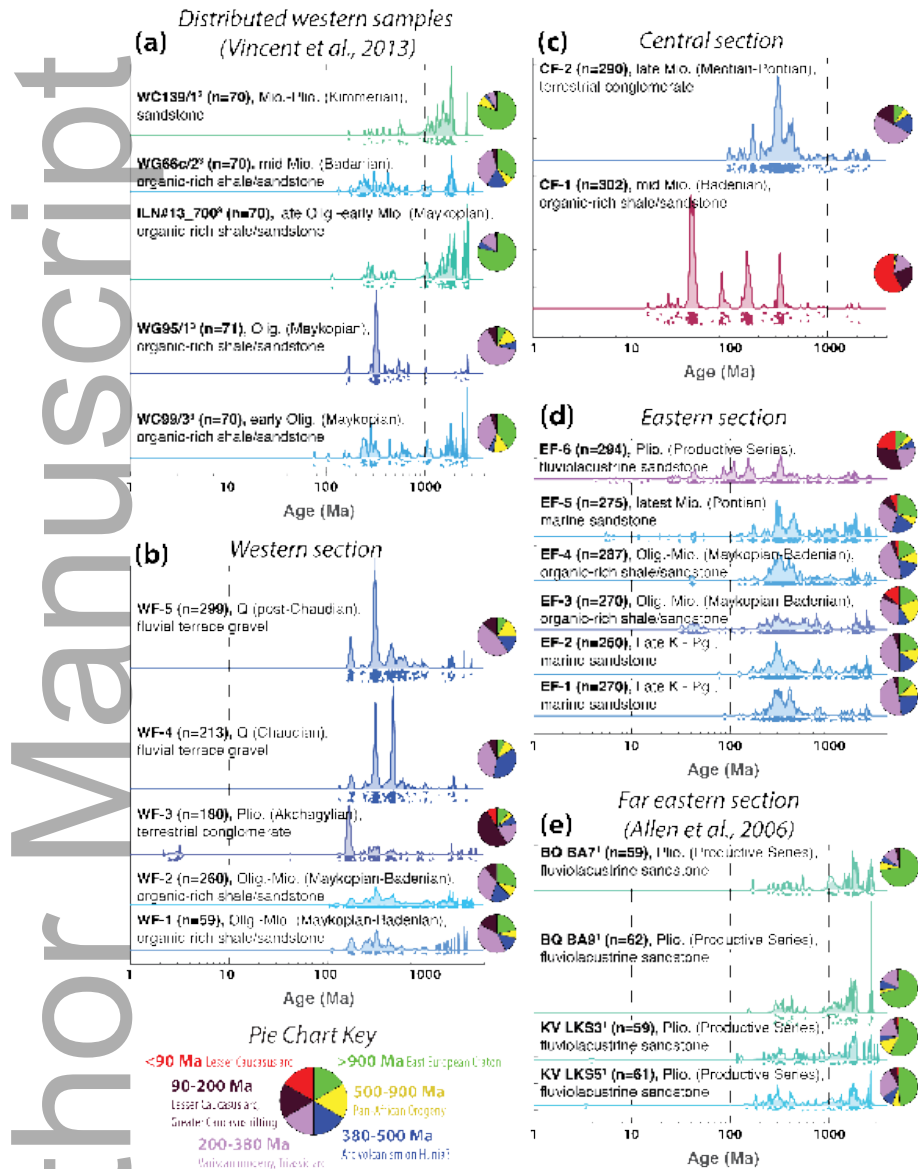


Figure 9: Detrital zircon age spectra of foreland basin sedimentary rocks reflect Cenozoic provenance variation over space and time. Previously published samples from distributed locations in the western Greater Caucasus are shown (a; Vincent et al., 2013). New samples were collected from western (b), central (c), and eastern (d) foreland basin sections. Previously published samples from a Pliocene section at the far eastern extent of the range are also reported (e; Allen et al., 2006). Spectra are shown in reverse stratigraphic order in each panel. Symbolology is the same as Figure 8. Sample ages, with regional stage in parentheses, and rock types, are listed. Abbreviations are as in previous figures, plus: Olig.–Oligocene, Mio.–Miocene, Plio.–Pliocene, Pleis.–Pleistocene. Empirical cumulative distribution functions of these samples are shown in Figure S2.

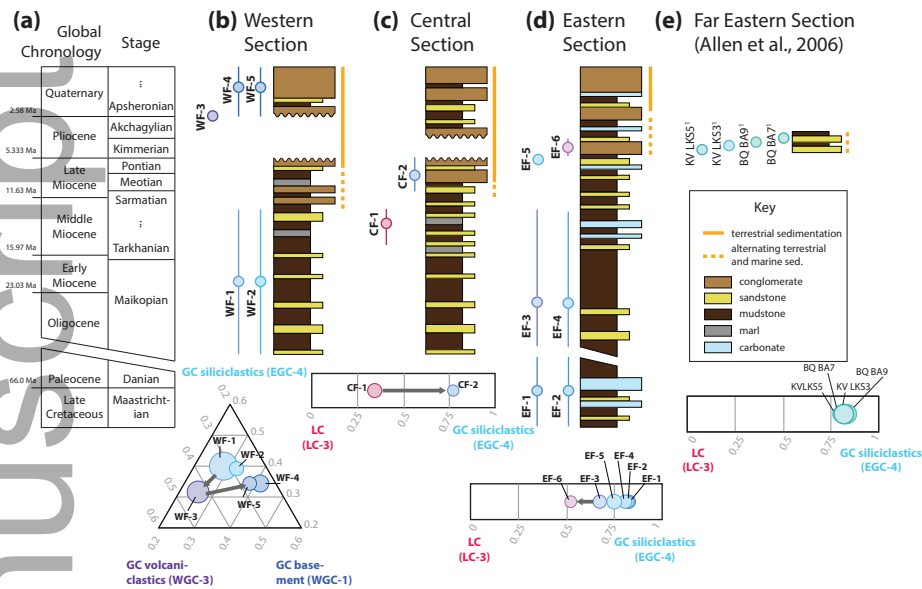


Figure 10: Foreland basin samples are shown in stratigraphic context. (a) Global chronology with stratigraphic stage names (Paratethyan stage names are used for the Neogene; Jones and Simmons, 1998). (b-e) western, central, eastern, and far eastern sampled foreland basin sections. Sample ages are depicted with symbols next to each stratigraphic column, with error bars representing the range of possible ages. Symbols are colored using the BPC coloring scheme used throughout this paper (Fig. 7). Beneath each section is a plot of normalized BPC of the samples relative to potential sources for the section (source sample numbers shown in parentheses), with arrows indicating trends over time (see Tables S3, S4 for BPC results). The endmembers for each plot are chosen based on which endmember sources (Fig. 8) are inferred to have contributed detrital zircon grains to each section (see text for further discussion). Plot symbol size is mean BPC uncertainty (1σ) with respect to the endmember samples, and symbols for the central and eastern sections have been doubled in size for visual clarity. Sources from the Greater and Lesser Caucasus are abbreviated GC and LC, respectively. Stratigraphy is schematic, based on Edilashvili (1957); Dzhanelidze and Kandelaki (1957); Gamkrelidze and Kakhazdze (1959); Voronin et al. (1959); Khain and Shardanov (1960); Mekhtiev et al. (1962); Hinds et al. (2004); Vincent et al. (2014) and field observations. Blank space in sections marks missing time due to unconformities. Unconformities without significant missing time are not shown. The distributed western samples of Vincent et al. (2013) are not depicted stratigraphically because they are from a variety of locations with variable stratigraphy.

468 to the northwest of the Greater Caucasus (samples ILN#13_700 and WC139/1; Fig.
 469 4), and three samples located on the southern margin of the range (samples WC99/3,
 470 WG66c/2, and WG95/1; Figs. 4, 5). The two samples northwest of the Greater Cauca-
 471 sus, late Oligocene to early Miocene sample ILN#13_700 and Miocene to Pliocene

472 sample WC139/1, are composed mostly of zircon grains >900 Ma (Fig. 9a) with
473 some scattered Neoproterozoic and Paleozoic ages. Overall these two samples show a
474 clear affinity to the Eurasian interior (Fig. 8a). Early Oligocene sample WC99/3, an
475 Oligocene marine sandstone sample from the south side of the westernmost Greater
476 Caucasus, contains ~40% zircon ages >900 Ma, as well as a 230-360 Ma age peak
477 (Fig. 9a). This sample shows a partial affinity to the Eurasian interior, with the 230-
478 360 Ma age peak suggesting a partial affinity to the Greater Caucasus siliciclastic se-
479 quence. Middle Miocene marine sandstone sample WG66c/2 was collected from near
480 our western section (Fig. 5b), and contains scattered Paleozoic to Triassic ages that
481 coalesce around two broad age peaks at 450 and 300 Ma, as well as ~35% ages >900
482 Ma (Fig. 8a). These ages indicate that WG66c/2 was likely derived predominantly
483 from the Greater Caucasus siliciclastic sequence. Sample WG95/1 was collected from
484 an Oligocene sandstone in close proximity to the Dzirula Massif (Fig. 5a), a Tran-
485 scaucasus basement massif, and has a detrital zircon age distribution dominated by a
486 narrow ~300 Ma age peak (Fig. 9a) that closely matches that of modern detritus from
487 the Dzirula Massif (sample TC-1, Fig. 8b).

488 The spatial distribution of source affinities within these samples has implications
489 for the Cenozoic depositional system of the Caucasus. The fact that samples on the
490 northern slope and near the western margin of the Greater Caucasus (ILN#13_700
491 and WC139/1; Fig. 9a) have a close affinity to samples of the Eurasian interior (Fig.
492 8a) suggests that detritus from the Eurasian interior was deposited to the north of the
493 Greater Caucasus and also to the south of the westernmost portion of the range (Fig. 4).
494 In contrast, the detrital zircon age distribution of sample WG66c/2 (Fig. 9a) includes
495 the major age peaks of the Greater Caucasus siliciclastic sequence (Fig. 8e), suggesting
496 that at the longitude at which it was deposited, sediment was sourced primarily from the
497 Greater Caucasus (Fig. 5b). Sample WC99/3 (Fig. 9a), deposited on the southern slope
498 of the Greater Caucasus at an intermediate longitude between WC139/1 and WG66c/2
499 (Fig. 4) shows a hybrid detrital zircon age signature suggesting mixing of the Eurasian
500 interior and Greater Caucasus siliciclastic sequence sources. Together, these samples
501 define a spatial mixing trend where the Eurasian interior is the dominant detrital zircon
502 source affinity of Neogene sediment on the north side of the Greater Caucasus and

503 in the far western portion of the basin to the south of the Greater Caucasus, whereas
504 the Greater Caucasus siliciclastic sequence is the dominant source affinity of Neogene
505 deposits on the southern margin of the central to western Greater Caucasus (Figs. 4,
506 5).

507 The spatial distribution of detrital zircon affinities to the Eurasian interior and
508 Greater Caucasus siliciclastic sequence mirrors the distribution of quartzose and lithic-
509 rich sandstones, respectively, in Oligocene to Pliocene deposits on the northeastern
510 margin of the Black Sea (Vincent et al., 2013, 2014). Quartzose sandstone is observed
511 in Neogene sedimentary rocks to the north of the Greater Caucasus and in the far
512 western portion of Neogene sedimentary rocks on the south side of the range (west
513 of 40° E; Vincent et al., 2013, 2014). The distribution of quartzose sandstone corre-
514 sponds spatially with detrital zircon age signatures of Eurasian affinity (Fig. 4; sam-
515 ples ILN#13_700 and WC139/1 in Fig. 9a). In contrast, lithic-rich sand containing
516 mudstone and volcanic fragments is observed in Neogene sedimentary rocks from the
517 western Greater Caucasus (east of 40° E; Vincent et al., 2013, 2014), in the same region
518 where Oligocene to Miocene sandstones reveal a detrital zircon age signature similar
519 to the Greater Caucasus siliciclastic sequence (Fig. 4; samples WC99/3 and WG66c/2
520 in Fig. 9a). The correspondence of quartz-rich sandstones with zircon ages of Eurasian
521 affinity and of lithic-rich sandstones with zircon of Greater Caucasus siliciclastic affin-
522 ity may reflect differing source area lithologies or the longer transport distance, and
523 thus probable greater maturity, of sediment from the Eurasian interior.

524 6.2. *Western foreland basin section*

525 Our western foreland basin section contains five samples spanning Oligocene to
526 Quaternary age (Figs. 9b, S2b) that were collected from a ~2.3 km thick sedimentary
527 section exposed along the Chanistskali River near Jvari, Georgia (Fig. 5b; Dzhanlidze
528 and Kandelaki, 1957). The section consists of organic-rich shales, marls, and tur-
529 biditic sandstones of Oligocene to Middle Miocene age (Maikopian through Badenian
530 regional stages; ~35 - 10.5 Ma; Dzhanlidze and Kandelaki, 1957, Fig. 10b) that
531 pass upward into conglomerates, sandstones, and mudstones of Late Miocene (Sar-
532 matian regional stage; 10.5 - 8.2 Ma; Jones and Simmons, 1998) to Quaternary age

(Dzhanelidze and Kandelaki, 1957, Fig. 10b). Late Miocene and younger strata in the western Greater Caucasus are interpreted as having been deposited in a largely terrestrial environment (Vincent et al., 2014). The two oldest samples from the western section, WF-1 and WF-2, were collected from Oligocene to Middle Miocene sandstones (Fig. 6a shows sample location of WF-2) and show dispersed Proterozoic to Triassic ages with wide peaks centered on 450 Ma and 300 Ma (Fig. 9b) and ~25% of ages >900 Ma. WF-1 and WF-2 have a zircon age peak at 170 Ma, as well. The age peaks of these two samples correspond well with samples of the Greater Caucasus siliciclastic sequence (Fig. 8e). The three youngest samples collected from the section, samples WF-3, WF-4, and WF-5, were collected from latest Pliocene to Quaternary terrestrial conglomerates (Fig. 6b shows sample location of WF-3). Sample WF-3 is dominated by a 170 Ma peak, along with small, wide peaks at ~300 Ma and ~450 Ma (Fig. 9b). The dominance of the 170 Ma age peak in sample WF-3 suggests affinity to the western Greater Caucasus volcanoclastic sequence (samples CT130924-9A, WGC-3 in Fig. 8f). Sample WF-3 also shows a concentration of zircon ages from 3 - 2.5 Ma, which likely originate from the eruption of the Chegem caldera in the northern Greater Caucasus at ~2.8 Ma (Lipman et al., 1993). Samples WF-4 and WF-5 have tightly clustered ~300 Ma and ~450 Ma age peaks, and have 7-8% zircon ages >900 Ma, significantly fewer than stratigraphically lower samples WF-1 and WF-2 (Fig. 9b). The tight clustering of the ~300 Ma and ~450 Ma age peaks and smaller portion of ages >900 Ma in samples WF-4 and WF-5 differentiate these samples from WF-1 and WF-2 and suggest that WF-4 and WF-5 have an affinity to the Greater Caucasus basement, rather than the Greater Caucasus siliciclastic sequence.

The three sources most similar to the age spectra observed in the western section are the Greater Caucasus siliciclastic sequence (Fig. 8e), Greater Caucasus volcanoclastic sequence (Fig. 8f), and the Greater Caucasus basement (Fig. 8b), all of which are located to the north of the section, so the observed provenance changes likely reflect changing exposure within the sediment source area. Therefore, we regard the provenance changes in the western section as recording the exposure of the volcanoclastic strata and the basement of the Greater Caucasus as a result of progressive deformation, unroofing, and erosion of the range. The age of first exposure of the Greater Caucasus

564 volcaniclastic strata is uncertain, but is bracketed by Middle Miocene sample WG66c/2
565 (Vincent et al., 2013), which is located near our sampled section and which shows no
566 evidence of derivation from the volcaniclastic strata, and late Pliocene sample WF-3,
567 which is dominated by ~170 Ma ages. Samples WF-4 and WF-5 record initial expo-
568 sure of the Greater Caucasus basement in the sedimentary source area during latest
569 Pliocene to Quaternary time. Combining the detrital zircon age data with stratigraphic
570 observations (Fig. 10b) reveals that the initial exposure of basement, and potentially the
571 initial exposure of the Greater Caucasus volcaniclastic strata, followed the transition to
572 terrestrial sedimentation within the western Caucasus.

573 An analysis of recycled palynomorphs from the same section that we sampled
574 also constrains the unroofing history of the western Greater Caucasus (Vincent et al.,
575 2014). Successively older palynomorphs are found stratigraphically higher in the sec-
576 tion, which suggests the exhumation of progressively deeper strata over time in the
577 source area (Vincent et al., 2014). In Early Oligocene time, the oldest palynomorphs
578 observed are of Eocene age. Beginning in Late Oligocene time, palynomorph assem-
579 blages imply source ages as old as Early Cretaceous, with a significant portion of
580 Eocene palynomorphs also present. In Early Miocene time, the prevalence of Eocene
581 palynomorphs decreases and recycled palynomorphs transition to predominantly Cre-
582 taceous age. A small number of palynomorphs in Early Miocene strata imply Middle
583 Jurassic source ages (Vincent et al., 2014). Though no samples younger than Early
584 Miocene were analyzed (Vincent et al., 2014), the exhumation history implied by these
585 samples is consistent with eventual exposure of basement in the sedimentary source
586 area during Pliocene to Quaternary time.

587 6.3. Central foreland basin section

588 Our central foreland basin section (Figs. 9c, S2c) contains two samples of Middle
589 and Late Miocene age, collected from a 5 - 7.5 km thick Oligocene to Quaternary
590 succession 30 km northeast of Tbilisi, Georgia (Fig. 5a; Edilashvili, 1957). In the
591 sampled section, Oligocene to Miocene mudstones, marls, and sandstone interlayers of
592 the Maykopian to middle Sarmatian regional stages (~36 - ~9 Ma; Edilashvili, 1957,
593 Fig. 10c) pass upward into sandstones, variegated mudstones, and coals of the Late

594 Miocene upper Sarmatian regional stage (~9 - 8.2 Ma; Edilashvili, 1957, Fig. 10c),
595 which are overlain by Late Miocene sandstones and conglomerates of the Meotian
596 to Pontian regional stages (8.2 - 5.3 Ma; Edilashvili, 1957, Fig. 10c). Sample CF-1
597 was taken from a Middle Miocene (pre-Sarmatian) sandstone bed within a shale-rich
598 sequence (Fig. 6c). CF-1 contains zircon ages <90 Ma and age peaks centered on
599 300 Ma and 170 Ma (Fig. 9c), a very similar age distribution to modern samples of
600 the Lesser Caucasus (Fig. 8b). Sample CF-1 also contains two ~15 Ma zircon grains,
601 which provide a maximum depositional age. Upsection, Late Miocene (Meotian to
602 Pontian) terrestrial conglomerate sample CF-2 (Fig. 6d) has dispersed Proterozoic to
603 Mesozoic zircon ages with wide peaks centered on 450-400, 300, and 170 Ma (Fig.
604 9c), indicating affinity to samples of the Greater Caucasus siliciclastic sequence (Fig.
605 8e).

606 The transition of sediment source from the Lesser Caucasus to the Greater Cauca-
607 sus observed in the central foreland basin section is most simply explained by tectonic
608 translation toward the Greater Caucasus via subduction/shortening. At the outcrop
609 from which CF-1 was collected, folding within isolated strata between undeformed
610 stratigraphic packages suggests that syn-sedimentary slumping occurred (Fig. 6c).
611 Given the Lesser Caucasus provenance of CF-1 (Fig. 9c) and the shale-rich lithology
612 and syn-sedimentary deformation of the outcrop from which it was collected, CF-1 was
613 likely deposited on the the Lesser Caucasus basin margin, in an environment similar
614 to a continental slope. The Greater Caucasus affinity of sample CF-2 indicates that at
615 least some interval of the Meotian to Pontian regional stages (8.2 - 5.3 Ma; Jones and
616 Simmons, 1998) was derived from the Greater Caucasus. The absolute minimum age
617 for this provenance switch is thus 5.3 Ma. The central section also contains a Pliocene
618 hiatus of similar timing and duration to the western section (Fig. 10).

619 6.4. Eastern foreland basin section

620 Samples from the eastern section (Figs. 9d, 10d, S2d) span almost the entire Ceno-
621 zoic, from latest Cretaceous or Paleocene time until Pliocene time, and were collected
622 from a 6 - 7.5 km thick composite section (Fig. 5c; Khain and Shardanov, 1960).
623 Within this section, a transition from marine, turbiditic sandstone, shale, and marl de-

624 position to largely terrestrial, conglomeratic deposition occurred in latest Miocene time
625 (Pontian regional stage) to earliest Pliocene time (Kimmerian regional stage; Khain and
626 Shardanov, 1960). Most samples from this section (samples EF-2 through EF-6) were
627 collected near Lahij and Shamakhi, Azerbaijan. Late Cretaceous to early Paleocene
628 sample EF-1 was collected from the north side of the Greater Caucasus, near the village
629 of Afurgha, Azerbaijan, though it was deposited prior to shortening and topographic
630 development in the Greater Caucasus (which began in late Eocene to Oligocene time;
631 Vincent et al., 2007; Adamia et al., 2011a), and is thus inferred to have been deposited
632 in the same basin as samples EF-2 through EF-6. Samples EF-1 to EF-5 were collected
633 from marine sandstone-shale sequences of Paleogene through Late Miocene age (Fig.
634 6e shows shale-rich interval from which EF-4 was collected). EF-1 through EF-5 re-
635 veal a consistent detrital zircon age signature featuring dispersed Proterozoic to Triassic
636 ages, typically with peaks centered on 400-450 Ma and 300 Ma (Fig. 9d). Age peaks
637 centered on 170 Ma are also sometimes present, and Oligocene to late Miocene sam-
638 ples in this section also show some ages from 60 to 30 Ma. Overall, samples EF-1 to
639 EF-5 show a strong similarity to modern samples of the Greater Caucasus siliciclastic
640 sequence (Fig. 8e). The Cenozoic grains in samples EF-3, EF-4, and EF-5 are likely
641 to have originated in Cenozoic volcanic centers of the Lesser Caucasus and neigh-
642 boring Talysh mountain ranges (Allen and Armstrong, 2008; Verdel et al., 2011; van
643 Der Boon et al., 2017). These Cenozoic zircon grains could have been transported by
644 turbidity currents and mixed with Greater Caucasus-derived sediment within the basin,
645 or they could have been deposited in the basin as volcanic airfall and subsequently
646 reworked. We tentatively favor the latter interpretation because samples EF-3, EF-4,
647 and EF-5 lack the major Jurassic and Cretaceous age peaks that characterize modern
648 and foreland basin samples derived from the Lesser Caucasus (samples LC-1 to LC-4,
649 Fig. 8c; CF-1, Fig. 9c). Together, samples EF-1 to EF-5 indicate derivation from the
650 Greater Caucasus siliciclastic sequence from Late Cretaceous or Paleocene time until
651 Late Miocene time.

652 Pliocene sample EF-6 (Fig. 6f) was collected from a sandstone horizon of the
653 thick, fluviolacustrine Productive Series (e.g., Hinds et al., 2004). The detrital zircon
654 U-Pb age distribution of EF-6 shows scattered Proterozoic to Cenozoic zircon ages

655 with peaks centered on ~300 Ma, 160 Ma, 105 Ma, and 85 Ma, with many additional
656 ages <85 Ma (Fig. 9d). These age peaks indicate affinity to the Lesser Caucasus arc
657 sequence (Fig. 8c). However, EF-6 also contains Precambrian zircon ages and a wide
658 ~450 Ma age peak, indicating affinity to the Greater Caucasus siliciclastic sequence
659 (Fig. 8e) in addition to the Lesser Caucasus arc sequence (Fig. 8c). Heavy mineral
660 provenance from Productive Series strata in the same region also suggest derivation
661 from the Lesser Caucasus (Morton et al., 2003), and some paleocurrents within the
662 Productive Series are oriented toward the east (Vincent et al., 2010), similar to the
663 modern Kura River (shown in Fig. 5a).

664 The contrasting provenance and lithology between the Pliocene, fluviolacustrine
665 Productive Series (sample EF-6) and pre-Pliocene underlying turbiditic marine strata
666 (samples EF-1 to EF-5) suggest a significant change in the drainage network. Detrital
667 zircon ages in the pre-Pliocene organic-rich sandstone-shale intervals (samples EF-1
668 through EF-5) suggest derivation from the Greater Caucasus to the north. In contrast,
669 the presence of Lesser Caucasus-derived material (sample EF-6; Morton et al., 2003)
670 and eastward paleocurrent directions (Vincent et al., 2010) in the Pliocene Productive
671 Series suggest deposition in a longitudinal drainage that included both Greater and
672 Lesser Caucasus sources within its catchment. The Productive Series was deposited
673 over 2-3 Myrs beginning in the earliest Pliocene (5.3 Ma) and attains thicknesses of 4-5
674 km in the Kura-South Caspian region (Green et al., 2009; Vincent et al., 2010), whereas
675 the entire Oligo-Miocene sequence attains a maximum thickness of 2.5 km (Green
676 et al., 2009), indicating an increase in sedimentation rate coincided with this change in
677 provenance. Deposition of the Productive Series, including sample EF-6, would have
678 roughly coincided with non-deposition or erosion in the western and central foreland
679 basin sections (Fig. 10b-d), suggesting that some Productive Series sediment may have
680 been eroded from the western foreland basin. The Pliocene deposition of Greater-
681 and Lesser Caucasus-derived sediment in the eastern foreland basin and erosion in the
682 western foreland basin may reflect an absence of accommodation between the Greater
683 Caucasus and Lesser Caucasus at the longitude where the continents were in closest
684 proximity to one another.

685 6.5. Far eastern foreland basin section

686 The far eastern section consists of previously published samples from the Pliocene
687 Productive Series sandstones on the Apsheron Peninsula in easternmost Azerbaijan
688 (Figs. 4, 9e, 10e, S2e; Allen et al., 2006). We note that this section covers a smaller
689 range of geologic time than the western, central, and eastern sections discussed above
690 (Fig. 10). These samples show a virtually constant detrital zircon age signature through
691 time that features a majority of ages >900 Ma, with scattered Neoproterozoic to Meso-
692 zoic ages that coalesce around ~300 Ma and 400-450 Ma age peaks in some samples
693 (Fig. 9e). A subsequent detrital zircon study with greater sampling resolution of this
694 section revealed similar age signatures (Abdullayev et al., 2018). The concentration
695 of zircon ages >900 Ma in these samples indicates affinity to the age signatures of
696 the Eurasian interior (Fig. 8a), with the wide ~300 Ma and 400-450 Ma age peaks of
697 some samples suggesting affinity to the Greater Caucasus siliciclastic strata (Fig. 8e),
698 as well.

699 7. Tectonic context of observed zircon crystallization ages

700 7.1. Cenozoic zircon ages

701 Cenozoic zircon ages are found primarily in samples derived from the Lesser Cau-
702 casus (Fig. 8c), and they are also present in small quantities in several samples that
703 otherwise appear to be derived from Greater Caucasus sources (samples WF-3, EF-3,
704 EF-4, EF-5; Fig. 9). The Lesser Caucasus was the site of a Mesozoic to Eocene arc,
705 as well as subsequent volcanism that spanned the Oligocene to Quaternary (Nalivkin,
706 1976; Dilek et al., 2010; Adamia et al., 2011b; Sahakyan et al., 2017). The western
707 Greater Caucasus hosts Pliocene to Quaternary volcanic centers (Lipman et al., 1993),
708 and also contains small, isolated intrusions of pre-Pliocene age (Nalivkin, 1976). Given
709 the close age correspondence between late Cenozoic zircon ages in sample WF-3 (2.5
710 - 3 Ma; Fig. 9b) and the eruption of Chegem caldera in the western Greater Caucasus
711 (2.8 Ma; Lipman et al., 1993), Chegem is a likely source for the young detrital zircon
712 ages of WF-3. No Cenozoic volcanic centers are known in the eastern Greater Cau-
713 casus, so we attribute Cenozoic zircon ages in eastern foreland basin samples EF-3,

714 EF-4, and EF-5 (Fig. 9d) to volcanic airfall from the Lesser Caucasus and neighboring
715 Talysh.

716 7.2. *Permian to Mesozoic zircon ages*

717 Cretaceous zircon ages are found in samples from the Lesser Caucasus (Fig. 8c),
718 the Greater Caucasus siliciclastic sequence (Fig. 8e), and the Greater Caucasus vol-
719 caniclastic sequence (Fig. 8f). In the Lesser Caucasus arc sequence (Fig. 8c), Cre-
720 taceous zircon grains are common and likely were crystallized during Mesozoic arc
721 volcanism (Sosson et al., 2010; Adamia et al., 2011b; Rolland et al., 2011). In the
722 Greater Caucasus siliciclastic sequence (Fig. 8e), Cretaceous zircon grains are likely
723 derived by volcanic airfall from the Lesser Caucasus, which is the nearest known center
724 of Cretaceous volcanism (Sosson et al., 2010; Rolland et al., 2011). Cretaceous zircon
725 ages dominate the eastern Greater Caucasus volcanoclastic sequence, which is Creta-
726 ceous in age (Nalivkin, 1976; Kopp, 1985), defining a single narrow detrital zircon age
727 peak at 105 Ma (Fig. 8f).

728 Jurassic zircon ages are observed in samples of the Lesser Caucasus arc sequence
729 (Fig. 8c), the Greater Caucasus siliciclastic sequence (Fig. 8e), and the Greater Cau-
730 casus volcanoclastic sequence (Fig. 8f). Jurassic intrusions have also been recognized
731 in all three sequences (Nalivkin, 1976; Hess et al., 1995). The Jurassic marked the
732 initiation of arc volcanism in the Lesser Caucasus and the initial rifting of the Greater
733 Caucasus basin (Zonenshain and Le Pichon, 1986; Sosson et al., 2010; Vincent et al.,
734 2016), so it is unsurprising that Jurassic zircon ages were generated in association with
735 these settings and are common throughout the region. Because Jurassic zircon ages are
736 ubiquitous in Jurassic and younger sedimentary sequences throughout the Caucasus,
737 they are not useful for differentiating between potential sediment sources.

738 Permian to Triassic zircon ages are observed in significant quantity only in the
739 Greater Caucasus siliciclastic sequence (Fig. 8e) and foreland basin sediments inferred
740 to be sourced from it. Such Permian to Triassic grains are likely derived from Per-
741 mian and Triassic volcanic and volcanoclastic rocks that overlie the Greater Caucasus
742 basement on the northern slope of the range (Belov, 1981; Nazarevich et al., 1986).

743 *7.3. Precambrian to Carboniferous zircon ages*

744 Pre-Permian zircon ages in the Caucasus reflect the crystallization history of re-
745 gional basement domains. A ~300-360 Ma age peak is ubiquitous in the Greater Cau-
746 casus basement (Fig. 8b), Transcaucasus basement massifs (Fig. 8c), and the Greater
747 Caucasus siliciclastic sequence (Fig. 8e), as well as younger sedimentary strata derived
748 from these sources (Figs. 9). The 300-360 Ma age peak reflects crystallization within
749 or simultaneous with the Variscan orogeny, when a Gondwana-derived ribbon continent
750 that may have included the Greater Caucasus and Transcaucasus basement terranes was
751 accreted to the southern margin of Eurasia (Stampfli and Borel, 2002; Stampfli et al.,
752 2013), driving high temperature—low pressure metamorphism and magmatism in the
753 Caucasus region (Belov et al., 1978; Somin, 2011). The Greater Caucasus basement
754 and Greater Caucasus siliciclastic sequence also contain a ~450 Ma age peak, typi-
755 cally in subequal proportion to the ~300 Ma age peak (Fig. 8b, e). In our samples
756 from modern rivers that drain the Greater Caucasus basement, this ~450 Ma age peak
757 is likely sourced from pre-Carboniferous metamorphic complexes that constitute part
758 of the Greater Caucasus basement (Somin, 2011). Ages of ~450 Ma correspond to
759 a period when the Greater Caucasus basement has been proposed to have undergone
760 arc volcanism during transit from Gondwana to Laurussia as part of the superterrane
761 Hunia (Stampfli et al., 2013; Stampfli, 2013). Alternatively, 450 Ma ages are observed
762 in the Nubian shield, suggesting that ~450 Ma ages observed in the Greater Caucasus
763 basement may have crystallized on the Gondwanan margin (Abdel-Rahman and Doig,
764 1987; Höhndorf et al., 1994). The presence of 300-360 and 450 Ma age peaks in sam-
765 ples of the Greater Caucasus siliciclastic sequence (Fig. 8e) indicates that the source
766 region of this sequence may have undergone a history of metamorphism and magma-
767 tism similar to that of the Greater Caucasus basement. 600-900 Ma zircon ages are
768 observed in minor proportions in many samples of Greater Caucasus basement (Fig.
769 8b), Transcaucasus basement massifs (Fig. 8c), and Greater Caucasus siliciclastic se-
770 quence (Fig. 8e). These zircon ages suggest an affinity to the Pan-African orogeny,
771 which occurred on Gondwana (e.g., Avigad et al., 2003; Johnson and Woldehaimanot,
772 2003; Horton et al., 2008; Stern and Johnson, 2010; Johnson, 2014; Vasey et al., 2020).
773 Pre-900 Ma zircon ages are present in our study mostly in the modern detritus of the

774 Eurasian interior as well as sedimentary strata likely derived in part from the Eurasian
775 interior. Zircon grains of this age are associated with the East European Craton (Allen
776 et al., 2006; Bogdanova et al., 2008).

777 **8. Implications for late Cenozoic evolution of the Caucasus and stratigraphic** 778 **records of collision**

779 *8.1. Late Cenozoic provenance and lithological changes of Caucasus foreland basin* 780 *sedimentation*

781 Dramatic changes in sediment composition and provenance occurred in the Cauca-
782 sus during late Cenozoic time (Fig. 11). Pre-Middle Miocene strata consist of organic-
783 rich, turbiditic marine sandstones and shales inferred to have been deposited in a deep
784 marine environment (Fig. 10; Hudson et al., 2008). Detrital zircon U-Pb age dis-
785 tributions from pre-Middle Miocene samples imply sourcing from either the Greater
786 Caucasus or the Lesser Caucasus, with no observed mixing of source signatures (Fig.
787 9). Detrital zircon provenance of the central section reveals that Greater Caucasus
788 detritus was deposited on the Lesser Caucasus basin margin slope sometime between
789 15 Ma and 5.3 Ma (Fig. 9c; event 1 in Fig. 11a; Fig. 11d), suggesting the subduc-
790 tion/underthrusting of the Lesser Caucasus basin margin during that time interval. The
791 western and central sampled sections, which lie broadly within the western Greater
792 Caucasus where collision has been hypothesized to have begun in latest Miocene to
793 Pliocene time (Philip et al., 1989; Avdeev and Niemi, 2011; Cowgill et al., 2016), in-
794 dicate a transition to terrestrial and largely conglomeratic sedimentation during Late
795 Miocene time, around 10.5 to 8.5 Ma (Fig. 10b, c; event 2 in Fig. 11a). At the
796 Miocene to Pliocene transition (~5.3 Ma), a hiatus began in the western and central
797 sections (10b, c; event 3 in Fig. 11a; Fig. 11d), coeval with deposition of a thick
798 package of Lesser- and Greater Caucasus-derived sediment in a longitudinal drainage
799 network in the eastern foreland basin (Figs. 9d, 10d; event 4 in Fig. 11a; Fig. 11d). Fi-
800 nally, in latest Pliocene or Quaternary time (<2.8 Ma), the first sediment derived from
801 Greater Caucasus basement was deposited in the western foreland basin (Fig. 9b; event
802 5 in Fig. 11a; Fig. 11d).

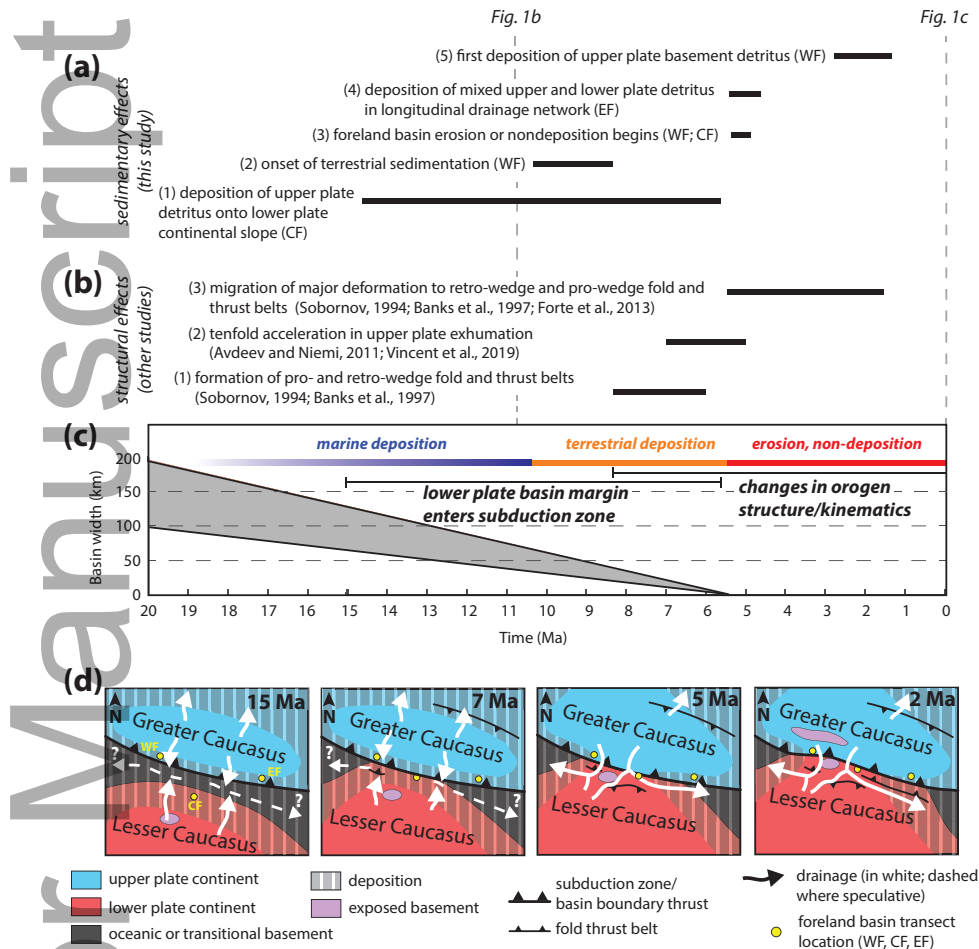


Figure 11: A timeline of sedimentary and structural effects of collision is developed from observations in the Caucasus. (a) Transitions in foreland basin sediment provenance and composition are inferred from our detrital zircon U-Pb age data and published stratigraphy. Each event is numbered for reference in the text and labeled parenthetically with the foreland basin section from which it was inferred (WF—western, CF—central, EF—eastern). (b) Structural changes in the orogen are reported from other studies, numbered and labeled with references. Vertical dashed lines indicate the ages associated with the timesteps of collision shown in Figure 1. (c) Basin width, foreland sedimentation style, and phase of collision are plotted against time. Basin width is inferred using timing estimates of Greater and Lesser Caucasus convergence and width estimates of the intervening basin (see Section 8.3 for further discussion). The gray shaded region indicates an uncertainty envelope based on variability in basin width estimates. (d) Schematic map view reconstruction of the late Cenozoic tectono-sedimentary evolution of the Caucasus.

803 *8.2. Drivers of observed lithology and provenance changes*

804 Potential drivers of late Cenozoic changes in depositional environment and prove-
805 nance across the Caucasus foreland basin include collision between the Greater and
806 Lesser Caucasus blocks, regional base level changes that occurred throughout the Paratethyan
807 system at this time (Krijgsman et al., 1999; Zubakov, 2001; Krijgsman et al., 2010;
808 Vasiliev et al., 2013; Forte and Cowgill, 2013), climatic changes, or autogenic pro-
809 cesses. To determine the effects of these potential drivers, we compare the timing of
810 observed sedimentary changes with the timing of Caucasus collision and the timing of
811 late Cenozoic regional base level changes.

812 *8.2.1. Transition from subduction to collision*

813 The Greater Caucasus underwent several structural and kinematic changes during
814 late Cenozoic time, many of which are temporally associated with the changes in sed-
815 imentary lithology and provenance outlined above. Following the initiation of defor-
816 mation in the Greater Caucasus at 35 Ma (Vincent et al., 2007; Adamia et al., 2011b),
817 the upper plate was exhumed slowly (~0.1 mm/yr) during Oligocene to Miocene time
818 as inferred from thermochronometry data (Avdeev and Niemi, 2011; Vincent et al.,
819 2011). This period of slow exhumation coincided with deposition of organic-rich, tur-
820 biditic sequences between the Greater and Lesser Caucasus, likely in a deep marine
821 setting, until Middle to Late Miocene time (Figs. 10, 11; Hudson et al., 2008). Prior
822 to Middle Miocene time, detrital zircon ages in the western and eastern foreland basin
823 sections indicate derivation exclusively from the Greater Caucasus, implying transport
824 from the north. In contrast, detrital zircon grains of the central foreland basin section
825 are derived from the Lesser Caucasus, implying transport from the south. Given the
826 compressional deformation occurring in the Greater Caucasus during this time (Vin-
827 cent et al., 2007; Adamia et al., 2011b), the slow Greater Caucasus exhumation rates
828 and presence of a marine basin between the Greater and Lesser Caucasus until at least
829 Middle Miocene time are consistent with Greater Caucasus–Lesser Caucasus conver-
830 gence was accommodated by subduction of the basin floor during Oligocene to Middle
831 Miocene time.

832 Several structural and sedimentary transitions took place in the Caucasus during

833 Middle to Late Miocene time. Between 15 and 5.3 Ma, deposition of Greater Cauca-
834 sus detritus onto the Lesser Caucasus basin margin is recorded in the central foreland
835 basin section (Fig. 10), which in other orogens has been inferred to reflect entrance
836 of the lower plate continental margin into the subduction zone (Fig. 11a; Garzanti
837 et al., 1987; Najman et al., 2010; DeCelles et al., 2014; Hu et al., 2015). During Late
838 Miocene time, deformation began within the Dagestan retro-wedge fold and thrust belt
839 (Sobornov, 1994) and the Tsaishi anticline, a pro-wedge fold-thrust structure to the
840 south of the western Greater Caucasus (Banks et al., 1997). Deformation within these
841 fold and thrust belts reflects migration of strain away from a single, dominant structure
842 that previously accommodated convergence. In models of incipient collision zones,
843 the development of fold and thrust belts corresponds with locking of the subduction
844 zone thrust due to the increasing thickness and buoyancy of lower plate material being
845 subducted (Beaumont et al., 1996; Regard et al., 2003; Toussaint et al., 2004a). Sedi-
846 mentary strata deposited between the Greater and Lesser Caucasus began to transition
847 during Middle to Late Miocene time from turbiditic sandstones and organic-rich shales
848 to conglomeratic red beds inferred to reflect terrestrial depositional environments (Figs.
849 10, 11). The timing of terrestrial deposition suggests it was caused by decreasing ac-
850 commodation space between the Greater and Lesser Caucasus as well as structural up-
851 lift above new thrust faults in some locations. The combination of deposition of Greater
852 Caucasus detritus onto the Lesser Caucasus basin margin, initiation of fold and thrust
853 belt deformation, and transition to terrestrial depositional environments is consistent
854 with incipient collision between the Lesser Caucasus arc terrane and the Greater Cau-
855 casus orogen during Late Miocene time, following subduction/underthrusting of the
856 intervening basin crust.

857 During latest Miocene to Pliocene time, structural changes within the orogen inten-
858 sified, coinciding with changes in foreland basin sediment routing. Thermochronome-
859 try data suggest that exhumation of the Greater Caucasus increased by a factor of 10, to
860 ~ 1 mm/yr, at 7-5 Ma (Avdeev and Niemi, 2011; Vincent et al., 2019), likely reflecting
861 accretion of lower plate material as predicted by some models in the early stages of
862 collision (Toussaint et al., 2004a, Fig. 11b). The Pliocene is reported as the time of
863 major activity on retro- and pro-wedge fold and thrust structures that first developed

864 in Late Miocene time (Sobornov, 1994; Banks et al., 1997, Fig. 11b). These Pliocene
865 structural changes coincide with erosion or non-deposition in the western to central
866 foreland basin (Figs. 10, 11a, d). The transition to erosion or non-deposition in the
867 western to central foreland basin coincided with longitudinal transport and mixing of
868 Greater- and Lesser Caucasus-derived sediments and their deposition in the Kura and
869 South Caspian basins (Figs. 10, 11). The coeval transition to erosive conditions in the
870 western to central foreland and longitudinal transport of Greater- and Lesser Caucasus-
871 derived sediments to the east is consistent with increasing proximity and deformation
872 between the Lesser Caucasus arc terrane and Greater Caucasus orogen.

873 The structural and sedimentary conditions of the Pliocene largely continued to the
874 Quaternary. The oldest observed foreland basin sample inferred to be derived from the
875 Greater Caucasus basement was deposited after 2.8 Ma (Figs. 10, 11a), suggesting that
876 initial exposure of basement in the sedimentary source area followed the increase in
877 exhumation rate that occurred in latest Miocene to Pliocene time (Avdeev and Niemi,
878 2011; Vincent et al., 2019). The pro-wedge fold and thrust belt of the Kura Basin
879 underwent initial deformation at ~2 - 1.5 Ma (Fig. 11b, d; Forte et al., 2013, 2014).
880 At present, deformation across most of the orogen is accommodated by fold and thrust
881 belts off the subduction zone (Forte et al., 2013; Sokhadze et al., 2018), contiguous
882 elevated topography stretches between the western Greater Caucasus and the Lesser
883 Caucasus (Fig. 2d), and longitudinal drainages are located between the two ranges
884 (Fig. 2a).

885 8.2.2. *Paratethys base level changes*

886 In addition to the late Cenozoic tectonic evolution of the Caucasus, Miocene to
887 Pliocene sedimentation may also have been affected by base level changes in the Paratethyan
888 basin system of which the Caucasus foreland basin was part (e.g., Popov et al., 2006;
889 Forte and Cowgill, 2013; van Baak et al., 2015, 2017). Base level falls of up to several
890 hundred meters may have occurred in the Black Sea and the Caspian Sea during Late
891 Miocene to Pliocene time, potentially as a result of disconnection between the Atlantic
892 Ocean and Mediterranean Sea during the Messinian Salinity Crisis (Krijgsman et al.,
893 1999; Zubakov, 2001; Krijgsman et al., 2010; Vasiliev et al., 2013; Forte and Cowgill,

2013; van Baak et al., 2017). This base level fall would have also reduced base level
in the basin between the Greater and Lesser Caucasus, which is likely to have served
as a connection between the Black and Caspian seas prior to its closure during Late
Miocene to Pliocene time (Zonenshain and Le Pichon, 1986; Popov et al., 2006). Low
Black Sea base levels lasted from 5.6 Ma until 5.4 Ma (van Baak et al., 2015), whereas
low base levels in the Caspian appear to have persisted from latest Miocene time until
4 - 2.7 Ma (Forte and Cowgill, 2013; van Baak et al., 2019). Connectivity between
the Black and Caspian Seas is inferred to have been severed in latest Miocene to earli-
est Pliocene time (Forte and Cowgill, 2013, and references therein), which our results
show may be a result of collision between the Greater and Lesser Caucasus.

The short duration of low Black Sea base levels indicates that regional Paratethyan
base level changes cannot by themselves account for the basin shallowing, terrestrial
sedimentation, and erosion/non-deposition observed in Caucasus foreland basin sec-
tions from Late Miocene time to the present. Changes in lithology and provenance ob-
served in the Cenozoic Caucasus foreland basin correspond temporally with structural
changes in the orogen that suggest basin closure and the initiation of Greater Caucasus–
Lesser Caucasus collision (Fig. 11). Many of the predicted sedimentary responses to
collision discussed in Section 2 are observed, including coarsening and shallowing of
the basin, mixing of upper and lower plate sediment in a longitudinal drainage, and
sourcing of detritus from deeper crustal levels (Fig. 1). Thus, we conclude that first
order sedimentation patterns in the late Cenozoic Caucasus foreland basin were driven
by Greater Caucasus–Lesser Caucasus collision. Changing regional base levels, along
with climate and autogenic processes, are inferred to have played a subordinate role.

8.3. *Correlating basin width with changes in sedimentary lithology and provenance*

Based on the observed correlation between structural and sedimentary changes
likely to be driven by collision in the Caucasus, we infer that convergence and col-
lision of the Greater and Lesser Caucasus is the primary driver influencing foreland
basin sediment composition and provenance. Thus, the width of the basin between
two converging continents may influence facies and provenance in pre-collisional to
collisional basins, and stratigraphic records may be able to be used to infer the width

924 of these closing basins at different points in time (e.g., Malkowski et al., 2017). We
925 use a simple calculation to estimate the width of the closing basin at the time these
926 changes occurred (Fig. 11c). Estimates of pre-convergence basin width between the
927 Lesser and Greater Caucasus range from 200 - 280 km from kinematic reconstructions
928 using paleomagnetic data (van der Boon et al., 2018) to 350 - 400 km by analogy to the
929 Black Sea basins and South Caspian basin (Cowgill et al., 2016). The basin between
930 the Greater and Lesser Caucasus is thought to have closed from 35 Ma (Adamia et al.,
931 2011a) until 5.3 Ma, when the basin became dominantly erosive and no longer accom-
932 modated sediment (Fig. 10), and for simplicity we assume a constant convergence rate
933 between 35 and 5.3 Ma. Assuming pre-convergence widths from 200 - 400 km and a
934 constant convergence rate from 35 Ma until 5.3 Ma yields convergence rates of 7 - 13
935 mm/yr. Such rates are comparable to modern convergence rates in the eastern Greater
936 Caucasus where subduction is inferred to be ongoing (Reilinger et al., 2006; Kadirov
937 et al., 2012, 2015). Using this basin width reconstruction, we find that when upper
938 plate detritus was deposited on the lower plate basin margin in the central section (15
939 - 5.3 Ma), the basin was <130 km wide (Fig. 11c). When the basin transitioned to
940 terrestrial sedimentation (10 - 8 Ma), its width was between 15 and 65 km (Fig. 11c).
941 When the basin became largely erosive (5.3 Ma), by definition the basin width was
942 reduced to zero (Fig. 11c). This reconstruction serves as a starting point for under-
943 standing the relationship between basin width, sedimentary lithology and provenance,
944 and the initiation of collision.

945 8.4. Comparison with other foreland basin systems

946 The evolution of the Caucasus foreland basin system, in addition to largely con-
947 forming to the hypothesis outlined in Section 2, shares several commonalities with the
948 evolution of other foreland basin systems in collisional and non-collisional settings.
949 The deposition of upper plate-derived detritus onto the lower plate is widely recognized
950 in the India-Asia collision zone (Garzanti et al., 1987; Najman et al., 2010; DeCelles
951 et al., 2014; Hu et al., 2015) and along the Arabia-Eurasia plate boundary (Koshnaw
952 et al., 2019). Where such deposition can be inferred to have occurred on the lower
953 plate continental margin, the age of deposition can be taken as an estimate of initial

954 continental subduction (DeCelles et al., 2014). These studies mirror observations in
955 our central section of upper plate detritus deposited stratigraphically above lower plate
956 detritus inferred to be deposited in a continental slope-type setting (Figs. 10, 11).

957 Underfilled foreland basin systems featuring longitudinal drainages close to the
958 thrust front, similar to that observed in the modern Caucasus foreland (Figs. 2, 10, 11),
959 are expected to exist in orogens undergoing active thrusting and accretion (Burbank,
960 1992; Raines et al., 2013). Given the increase in exhumation rate and activity on fold
961 and thrust belts in the Greater Caucasus since the Pliocene (Sobornov, 1994; Banks
962 et al., 1997; Avdeev and Niemi, 2011; Forte et al., 2013), we infer that significant
963 accretion has occurred recently or is ongoing, increasing the mass of the orogen and
964 driving foreland subsidence, resulting in the present drainage network. The drainage
965 network of the Caucasus is also likely to be influenced by high topography on the lower
966 plate driven by shortening in the Lesser Caucasus (Banks et al., 1997) and thermal and
967 dynamic uplift of the East Anatolian Plateau to the south (Keskin et al., 1998; Şengör
968 et al., 2003; Göğüş and Pysklywec, 2008).

969 **9. Caucasus collision evolution and comparison to other orogens and models**

970 Because natural examples of the transition from subduction to collision are rare,
971 analog and numerical modeling have been used extensively to investigate the effects
972 of collision (e.g., Beaumont et al., 1996; Chemenda et al., 1996; Regard et al., 2003;
973 Toussaint et al., 2004a,b; Faccenda et al., 2009). The late Cenozoic structural evolution
974 of the Caucasus orogen and the stratigraphic record of associated basins suggests that
975 collision between the Greater Caucasus orogen and the Lesser Caucasus arc terrane,
976 following the closure of an intervening marine basin, occurred during Late Miocene
977 time. The record of collision in the Caucasus may thus advance our understanding of
978 collision by serving as a test case for the process.

979 *9.1. Model predictions*

980 Analog and numerical models of the transition from subduction to collision reveal
981 many different possible evolutionary pathways of collisional plate boundaries that un-
982 fold over millions to tens of millions of years (Regard et al., 2003; Toussaint et al.,

2004b; Faccenda et al., 2009). In particular, the rate at which an orogen transitions
from accommodating convergence via subduction to accommodating convergence by
crustal shortening has been shown by models to depend on convergence rate, thermal
structure, and composition (Regard et al., 2003; Toussaint et al., 2004b). Stable
subduction of hundreds of kilometers of continental lithosphere is associated with convergence
rates of >25 mm/yr and cold subducting lithosphere (Moho temperature >550
 $^{\circ}\text{C}$; Regard et al., 2003; Toussaint et al., 2004b). In contrast, slower convergence rates
and hotter lithosphere is associated with convergence accommodated via lithospheric
shortening following initial subduction of the lower plate continental margin (Toussaint
et al., 2004b).

9.2. Comparison with the Caucasus and other natural systems

The Caucasus and other collisional orogens may provide insight into whether the
hypothesized relationships between convergence rate, thermal structure, and lithospheric
composition and continental subduction hold for natural systems. Collisional systems
proposed to have undergone significant continental subduction during the initiation of
collision include the Arabia-Eurasia collision (150 - 480 km; Pirouz et al., 2017; Bal-
lato et al., 2011) and the India-Asia collision (>500 km; Johnson, 2002, and references
therein). These collision zones both have cratonic lower plates (Sengupta et al., 1996;
Förster et al., 2010) and were characterized by convergence rates of 30 mm/yr (Arabia-
Eurasia; McQuarrie et al., 2003) and 200 mm/yr (India-Asia; Patriat and Achache,
1984) during the initiation of collision.

The amount of continental subduction in the Caucasus collisional system has not
been previously estimated. Given the constraints on pre-convergence width of the basin
between the Greater and Lesser Caucasus (<400 km; Cowgill et al., 2016; van der Boon
et al., 2018) and the timing of initial subduction of the Lesser Caucasus basin margin
slope (15 - 5.3 Ma), and assuming a constant convergence rate from initiation of basin
closure (35 Ma; Vincent et al., 2007; Adamia et al., 2011b) until final closure around
5.3 Ma, the amount of Lesser Caucasus continental crust subducted beneath Eurasia
following subduction of the Lesser Caucasus basin margin is <130 km. The Lesser
Caucasus was affected by Mesozoic to Paleogene arc volcanism (Sosson et al., 2010;

1013 Rolland et al., 2011; Adamia et al., 2011b) and is located on the northern margin of East
1014 Anatolia, a region inferred to have undergone lithospheric removal and/or detachment
1015 of the subducted Neotethys slab in Middle to Late Miocene time (Keskin et al., 1998;
1016 Şengör et al., 2003; Göğüş and Pysklywec, 2008). Therefore, continental lithosphere
1017 of the Lesser Caucasus is likely to be hotter and weaker than the cratonic lithosphere of
1018 Arabia and India. In addition, convergence rates during Caucasus collision are likely to
1019 have been significantly lower (7 - 13 mm/yr, assuming a constant convergence rate from
1020 Oligocene to latest Miocene time; Fig. 11) than the convergence rates inferred for the
1021 Arabia-Eurasia and India-Asia collision zones (Patriat and Achache, 1984; McQuar-
1022 rie et al., 2003). Thus, the relatively small amount of continental subduction inferred
1023 for the Caucasus compared to the Arabia-Eurasia and India-Asia collisional systems is
1024 consistent with model predictions for a system with slower convergence and a weaker,
1025 hotter lower plate continental lithosphere. The thickness and composition of base-
1026 ment initially located between the Greater and Lesser Caucasus, which may have been
1027 several kilometers thicker and/or less dense than typical oceanic crust (Mangino and
1028 Priestley, 1998; Cowgill et al., 2016), may also have reduced the amount of continental
1029 subduction compared to a system with typical oceanic lithosphere due to reduced slab
1030 pull.

1031 **10. Implications of Caucasus detrital zircon U-Pb age data for terrane boundaries** 1032 **and Tethyan tectonics**

1033 At the longitude of the Caucasus, the number and location of tectonic sutures along
1034 the southern Eurasian margin remain uncertain. Such sutures may have guided subse-
1035 quent deformation, as has been suggested in other tectonic settings (Jones and Tanner,
1036 1995; Rusmore et al., 2001; Fitzgerald et al., 2014). Scythia is thought to have under-
1037 gone a crystallization history distinct from that of the East European Craton (Saintot
1038 et al., 2006b), potentially due to a suture between Scythia and the craton. At the north-
1039 ern margin of the Greater Caucasus basement, several authors have identified an ophio-
1040 lite emplaced during Carboniferous time (Adamia et al., 1981; Somin, 2011), suggest-
1041 ing a suture between the Greater Caucasus and Scythia. However, Natal'in and Şengör

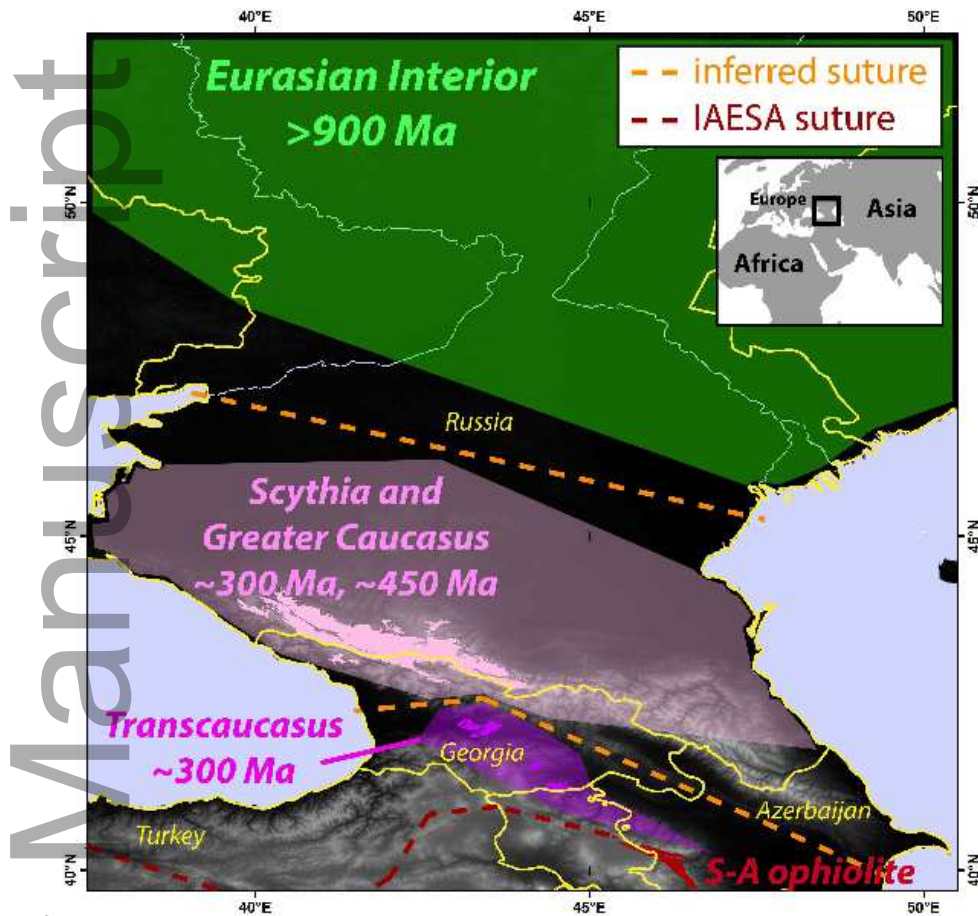


Figure 12: Crystallization ages inferred for basement domains from detrital zircon age data, and inferred suture locations between basement domains of shared crystallization history. Outcrops of crystalline basement in the Caucasus region are shown in opaque color and areas of inferred basement composition are partially transparent. IAESA stands for Izmir-Ankara-Erzincan-Sevan-Akera suture, shown in dark red dashed line. S-A ophiolite stands for Sevan-Akera ophiolite, part of the IAESA suture, exposure of which is shown in dark red. See Section 10 for further discussion.

1042 (2005) argue that the Greater Caucasus basement is part of Scythia that was displaced
 1043 by Triassic strike-slip displacement, meaning that the present location of the ophiolite
 1044 between the Greater Caucasus basement and Scythia may not reflect a true suture be-
 1045 tween the two domains. Several reconstructions place a suture between the Greater
 1046 Caucasus and Transcaucasus basement domains (Şengör, 1984; Stampfli, 2013; van

1047 Hinsbergen et al., 2019), although other authors have suggested a shared history be-
1048 tween the Greater Caucasus and the Dzirula Massif, the northernmost exposed Tran-
1049 scaucasus basement, based on petrologic and age similarities (e.g., Mayringer et al.,
1050 2011). South of the Transcaucasus, terrane boundary locations are less ambiguous be-
1051 cause of the presence of ophiolites along the Sevan-Akera suture zone (e.g., Khain,
1052 1975; Galoyan et al., 2009), the Bitlis-Zagros suture zone (e.g., Sengör and Yilmaz,
1053 1981), and between South Armenia and the easternmost Taurides (e.g., Topuz et al.,
1054 2017; van Hinsbergen et al., 2019).

1055 Our detrital zircon age data constrain the timing and significance of magmatic
1056 and metamorphic episodes affecting basement domains of the East European Cra-
1057 ton, Scythia, the Greater Caucasus, and the Transcaucasus, which add evidence for
1058 or against proposed sutures between these domains (Fig. 12). Our modern samples
1059 directly characterize the crystallization histories of the Greater Caucasus basement
1060 (Fig. 8b) and Transcaucasus basement (Fig. 8c), and published samples reflect the
1061 crystallization history of the Eurasian interior (Allen et al., 2006; Wang et al., 2011,
1062 Figs. 8a, 12). Sedimentary architecture (Sholpo, 1978) and field observations (Vincent
1063 et al., 2013) indicate that the Greater Caucasus siliciclastic sequence is derived from
1064 the north, suggesting that our samples from this sequence (Fig. 8e) constrain the crys-
1065 tallization history of the Eurasian interior and/or Scythia. Unlike the Eurasian interior,
1066 detrital zircon age signatures from the Greater Caucasus siliciclastic sequence contain
1067 a majority of ages <900 Ma, typically with peaks at 300 Ma and 450 Ma (Fig. 8e). Be-
1068 cause zircon grains of age <900 Ma are comparatively rare in samples of the Eurasian
1069 interior and do not cluster in clear age peaks at 300 Ma and 450 Ma (Fig. 8a), it is likely
1070 that the <900 Ma detrital zircon grains of the Greater Caucasus siliciclastic sequence
1071 are derived from Scythia (Fig. 12). The scattered >900 Ma ages present in Greater
1072 Caucasus siliciclastic sequence samples (Fig. 8e) may be derived from the Eurasian
1073 interior. Because the crystallization ages indicated by these detrital zircon grains con-
1074 strain the tectonic histories of the East European Craton (Allen et al., 2006; Wang et al.,
1075 2011), Scythia, the Greater Caucasus basement, and the Transcaucasus basement, they
1076 are likely to yield new insight into the locations of sutures on the southern margin of
1077 Eurasia and their role in guiding tectonic deformation on this complex plate margin.

1078 *10.1. Detrital zircon U-Pb age constraints on whether Scythia, Greater Caucasus*
1079 *basement, and Transcaucasus basement domains were formed on the Eurasian*
1080 *margin or were accreted*

1081 Central to locating terrane boundaries on the southern margin of Eurasia is de-
1082 termining whether Scythia, Greater Caucasus, and Transcaucasus basement domains
1083 formed in situ on the Eurasian margin or whether they originated on Gondwana or as
1084 intra-oceanic island arcs. The East European Craton is associated with zircon ages
1085 >900 Ma (Allen et al., 2006; Wang et al., 2011). Past work has identified zircon of age
1086 600-900 Ma as diagnostic of crystallization during the Pan-African orogeny, which
1087 occurred on Gondwana (Avigad et al., 2003; Johnson and Woldehaimanot, 2003; Hor-
1088 ton et al., 2008; Stern and Johnson, 2010; Johnson, 2014). Zircon grains of this age
1089 are virtually absent from samples containing detritus from the East European Craton
1090 (Fig. 8a). Our detrital zircon U-Pb ages from the Greater Caucasus siliciclastic se-
1091 quence (which we infer to be derived from Scythia), Greater Caucasus basement, and
1092 the Transcaucasus basement indicate that 600-900 Ma ages are present in all three
1093 domains, suggesting that they all originated on Gondwana (Fig. 8b, c, e). Whereas
1094 previously available data from Scythian basement were unable to differentiate whether
1095 Scythia was exotic to Eurasia (e.g., Saintot et al., 2006b), our data support the hypoth-
1096 esis that a suture divides Scythia from Eurasia (Fig. 12; Natal'in and Şengör, 2005).
1097 Our findings are consistent with the view that the Transcaucasus and Greater Caucasus
1098 basement domains are exotic to Eurasia (e.g., Ruban et al., 2007; Ruban, 2007, 2013;
1099 Stampfli, 2013; Vasey et al., 2020). The age of accretion of Scythia, the Greater Cau-
1100 casus, and Transcaucasus basement domains to Eurasia is bounded by the age of the
1101 Pan-African orogeny to be <600 Ma.

1102 *10.2. Detrital zircon age constraints on the similarities and differences between Greater*
1103 *Caucasus basement and Scythia*

1104 A suture between the Greater Caucasus basement and Scythia is suggested by ophi-
1105 olites and eclogite-bearing blueschists in the northern Greater Caucasus that divide the
1106 two domains and that were emplaced in the Carboniferous (e.g., Adamia et al., 1981;
1107 Perchuk and Philippot, 1997; Philippot et al., 2001; Somin, 2011), although the Greater

1108 Caucasus and Scythia have also been proposed to constitute a single terrane disrupted
1109 and duplexed by Triassic strike-slip faulting (Natal'in and Şengör, 2005). If the Greater
1110 Caucasus basement and Scythia constitute a single terrane, the two domains would be
1111 expected to share a common crystallization and metamorphic history. If the Greater
1112 Caucasus basement is a separate terrane from Scythia, it is unlikely (though possible)
1113 that the Greater Caucasus basement would share the crystallization history of Scythia.
1114 Detrital zircon ages from the Greater Caucasus basement cluster around age peaks at
1115 300 Ma and 450 Ma (Fig. 8b). Detrital zircon ages from the Greater Caucasus silici-
1116 clastic sequence, which we infer to be derived largely from Scythia, also cluster around
1117 age peaks at 300 Ma and 450 Ma (Fig. 8e). The major difference between the age sig-
1118 natures of the Greater Caucasus basement and Greater Caucasus siliciclastic sequence
1119 is that the 300 Ma and 450 Ma age peaks are wider in the siliciclastic sequence samples
1120 (Fig. 8e) than in the basement samples (Fig. 8b). Assuming that the Greater Caucasus
1121 siliciclastic sequence was derived from a large region or regions of Scythia, this differ-
1122 ence may reflect somewhat diachronous crystallization across Scythia, of which only a
1123 small portion is exposed in the Greater Caucasus basement. Pb loss or other complex-
1124 ities in preserved zircon U-Pb dates could exacerbate the difference in age peak width
1125 between the Greater Caucasus basement and Greater Caucasus siliciclastic sequence,
1126 but the lack of a systematic difference in discordance between the two sources (Fig. S3)
1127 suggests that such complexities are not likely to be responsible for the entire observed
1128 difference in age peak width. Overall, our detrital zircon ages suggest that the Greater
1129 Caucasus basement has a similar crystallization history to Scythia, lending support to
1130 the hypothesis that the Greater Caucasus basement is part of Scythia, and suggesting
1131 that there is not a major terrane boundary between the Greater Caucasus basement and
1132 Scythia (Fig. 12; Natal'in and Şengör, 2005). The presence of ophiolites in the north-
1133 ern Greater Caucasus may be attributable to strike slip duplexing of a single terrane
1134 (Natal'in and Şengör, 2005).

1135 *10.3. Detrital zircon age constraints on the similarities and differences between Greater*
1136 *Caucasus basement and Transcaucasus basement*

1137 While several authors have proposed the existence of a suture between the Greater
1138 Caucasus and Transcaucasus basement (Şengör, 1984; Adamia et al., 2011b; Stampfli,
1139 2013; van Hinsbergen et al., 2019), others have noted age and compositional similarity
1140 between the Transcaucasus and Greater Caucasus (Zakariadze et al., 2007; Mayringer
1141 et al., 2011) and suggested a shared tectonic history between the two domains. The
1142 presence or absence of a suture here is important because the Greater Caucasus basin
1143 opened between the Greater Caucasus basement and Transcaucasus basement (Zonen-
1144 shain and Le Pichon, 1986; Vincent et al., 2016). Thus, the opening of the Greater
1145 Caucasus basin may have been guided by a pre-existing structure between the Greater
1146 Caucasus and Transcaucasus. Our detrital zircon ages show that while the Greater
1147 Caucasus basement contains subequal age peaks at 300 Ma and 450 Ma (Fig. 8b), the
1148 Transcaucasus basement contains a 300 Ma age peak but does not contain a 450 Ma
1149 age peak (Figs. 8c, 12). Our samples of the Greater Caucasus siliciclastic sequence
1150 (representing Scythia) indicate that 300 Ma and 450 Ma age peaks are subequal in size
1151 across much of Scythia (Figs. 8e), in addition to within the Greater Caucasus basement
1152 (Figs. 8b, 12). The fact that the Transcaucasus basement lacks such a pervasive and
1153 significant age peak compared to the Greater Caucasus and Scythia lends support to the
1154 hypothesis that a suture separates the Greater Caucasus and Transcaucasus (Fig. 12).

1155 *10.4. Suture locations*

1156 The basement domain ages inferred from our detrital zircon data are consistent
1157 with the presence of two sutures between the Eurasian interior and the Transcaucasus,
1158 one between Eurasia and Scythia/Greater Caucasus and one between Scythia/Greater
1159 Caucasus and the Transcaucasus (Fig. 12). These sutures were generated by the suc-
1160 cessive transit of terranes from Gondwana to the Eurasian margin (e.g., Şengör, 1984;
1161 Stampfli et al., 2013) and thus the sutures decrease in age from north to south (Yıl-
1162 maz et al., 2014). The ophiolites located to the south of the Transcaucasus, including
1163 the Sevan-Akera ophiolites (Fig. 12) reflect sutures associated with Neotethys and are
1164 though to have closed in Late Cretaceous time or later (Sosson et al., 2010; Rolland

1165 et al., 2012), indicating that the sutures we infer to the north of the Transcaucasus must
1166 predate Neotethys.

1167 Up to three ocean basins have been proposed to exist between Gondwana/Africa
1168 and the Eurasian margin prior to Neotethys, termed the Qaidam, Rheic, and Paleotethys
1169 oceans (e.g., Şengör, 1984; Stampfli et al., 2013), and our inferred suture locations (Fig.
1170 12) are broadly consistent with multiple hypothesized locations of these sutures. Sev-
1171 eral authors infer that at the longitude of the Caucasus, the Paleotethys suture coincides
1172 spatially with the Neotethys suture along the Sevan-Akera suture zone (Fig. 12; e.g.,
1173 Adamia et al., 2011b; Stampfli, 2013). If this is the case, then the two sutures we infer
1174 between Eurasia and the Transcaucasus would represent the Qaidam and Rheic ocean
1175 sutures (Stampfli, 2013). However, other authors prefer to place the Paleotethys suture
1176 between the Greater Caucasus and the Transcaucasus due to the lack of any pre-Triassic
1177 rocks, which would be expected for Paleotethys, within the Sevan-Akera suture zone
1178 (e.g., Şengör, 1984; Natal'in and Şengör, 2005; van Hinsbergen et al., 2019). In this
1179 case, the inferred suture between Scythia and the Eurasian interior may correspond with
1180 the Qaidam ocean and the Rheic ocean suture may correspond spatially with either the
1181 Qaidam or Paleotethys sutures. The opening and subsequent closure of the Greater
1182 Caucasus basin following the formation of these sutures is likely to have obscured evi-
1183 dence of any of these sutures located between the Greater Caucasus and Transcaucasus
1184 (Cowgill et al., 2016; van der Boon et al., 2018; van Hinsbergen et al., 2019).

1185 **11. Conclusions**

1186 We present new detrital zircon U-Pb age data from the Caucasus that reveal tempo-
1187 rally correlated changes in orogen structure and sediment provenance consistent with
1188 a Middle Miocene to Pliocene initiation of collision between the Greater and Lesser
1189 Caucasus. Oligocene to Miocene strata record deposition in a deep marine environ-
1190 ment between the Greater and Lesser Caucasus, while the Greater Caucasus was al-
1191 ready undergoing deformation (Vincent et al., 2007), potentially as an accretionary
1192 prism. Upper plate (Greater Caucasus) detritus was deposited onto the lower plate
1193 (Lesser Caucasus) margin at 15 - 5.3 Ma, implying subduction/underthrusting of the

1194 lower plate basin margin at this time, approximately coeval with a Late Miocene tran-
1195 sition to terrestrial sedimentation. Accelerated upper plate exhumation and migration
1196 of significant shortening to fold and thrust belt systems occurred around 5.3 Ma, coeval
1197 with a transition to erosive conditions in the foreland basin at the locus of collision and
1198 deposition of a thick package of upper- and lower plate-derived detritus transported
1199 longitudinally. These structural changes and the initiation of erosive foreland condi-
1200 tions suggest a transition in the mode of convergence accommodation from subduction
1201 to crustal shortening by 5.3 Ma.

1202 Our results suggest that the lower plate basin margin was subducted at most ~9 Myr
1203 prior to the initiation of major crustal shortening associated with the Greater Caucasus-
1204 Lesser Caucasus collision, during which <130 km of Lesser Caucasus continental litho-
1205 sphere could have been subducted. This amount of continental subduction is less than
1206 has been proposed for the India-Asia and Arabia-Eurasia collision systems (Johnson,
1207 2002; Ballato et al., 2011; Pirouz et al., 2017). However, the amount inferred for the
1208 Caucasus is qualitatively consistent with geodynamic models of collision systems with
1209 moderate convergence rates (~7 - 13 mm/yr) and hot, weak lower plate lithosphere, as
1210 inferred in the Caucasus.

1211 Our detrital zircon U-Pb age data also reveal crystallization histories of regional
1212 basement terranes, constraining the locations of tectonic sutures. The East European
1213 Craton is characterized by zircon ages >900 Ma, while Scythia and the Greater Cau-
1214 casus basement share sub-equal zircon age peaks centered on 450 Ma and 300 Ma,
1215 and the Transcaucasus basement is dominated by a 300 Ma age peak and lacks 450
1216 Ma zircon ages. These age distributions suggest sutures between Scythia and the East
1217 European Craton and between the Greater Caucasus basement and the Transcaucasus.
1218 Scythia, the Greater Caucasus basement, and the Transcaucasus basement all contain
1219 zircon grains of 900-600 Ma, characteristic of the Pan-African orogeny on Gondwana.
1220 Thus, all three domains likely originated on Gondwana.

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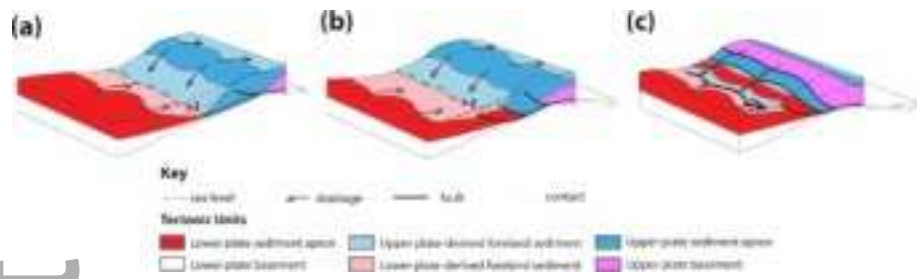
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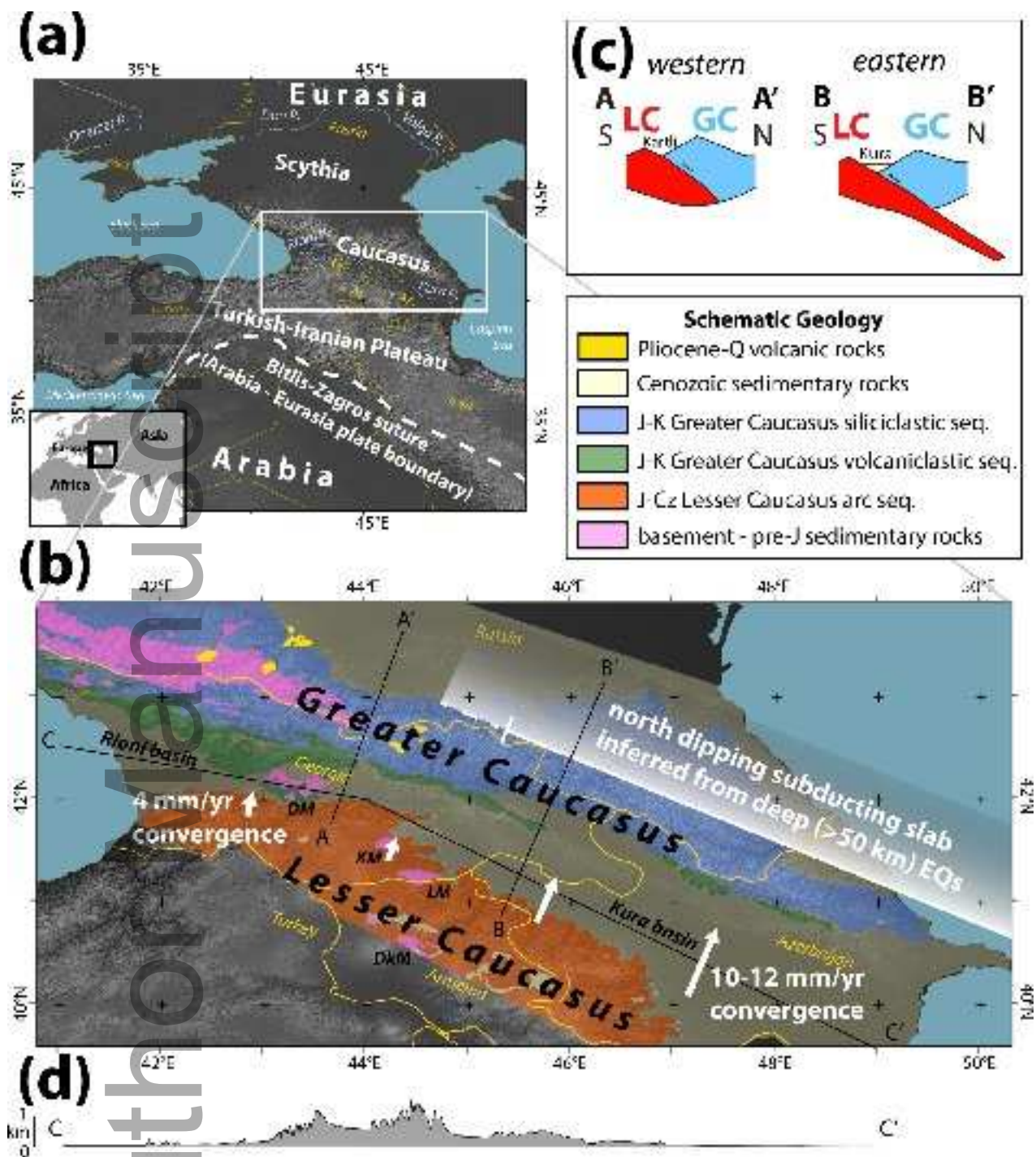
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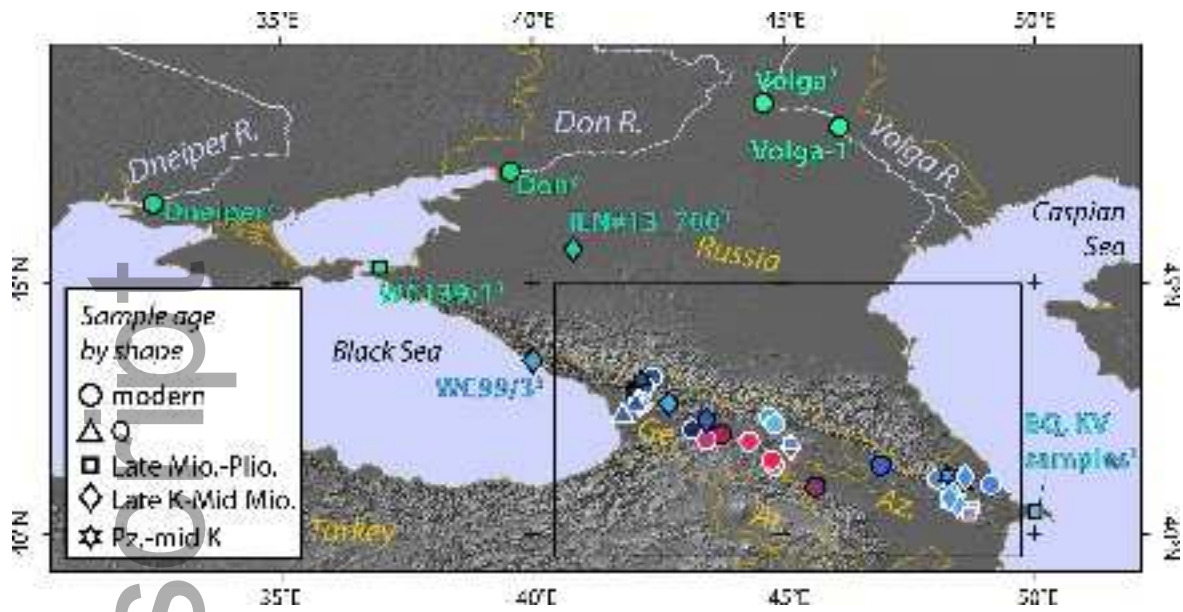


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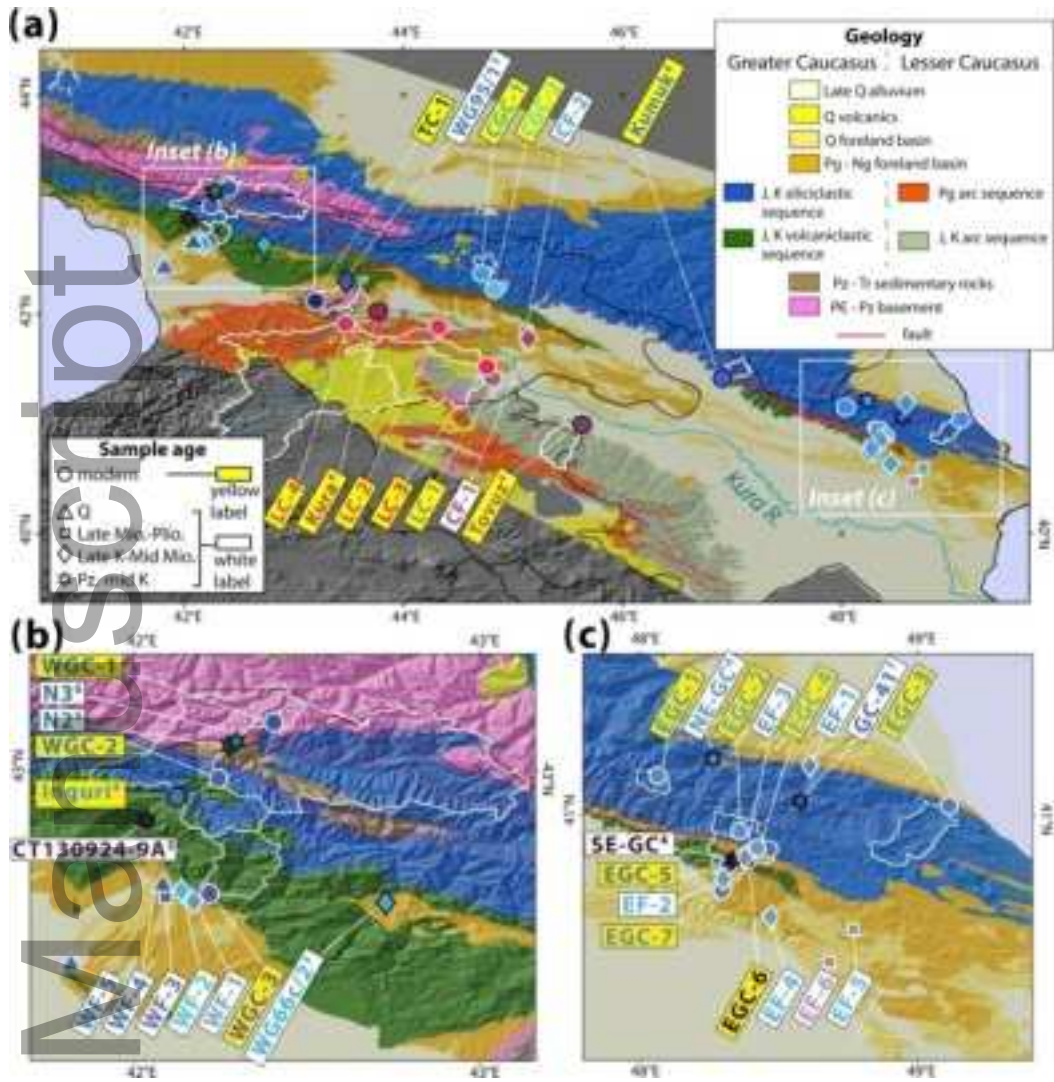
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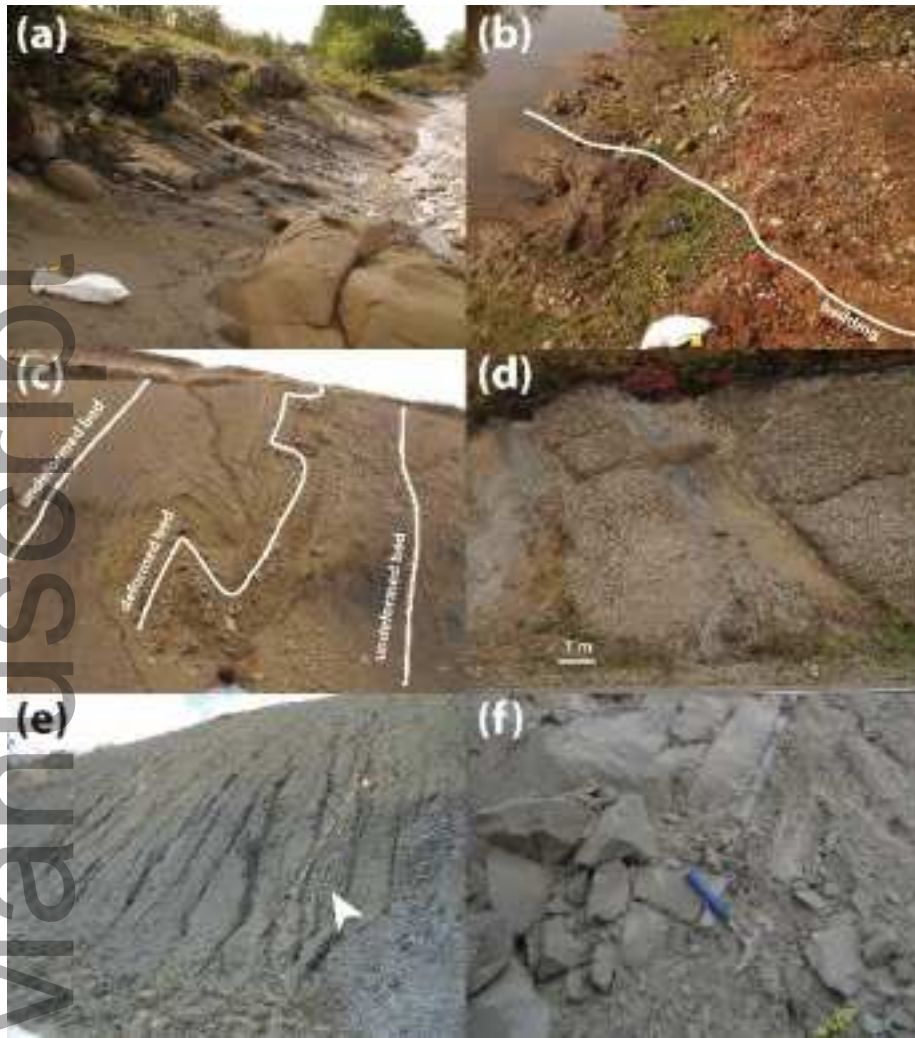


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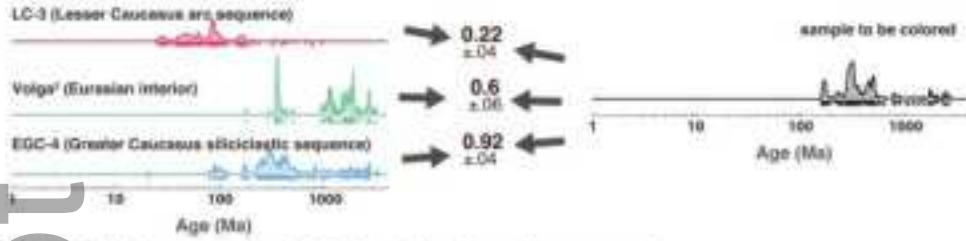


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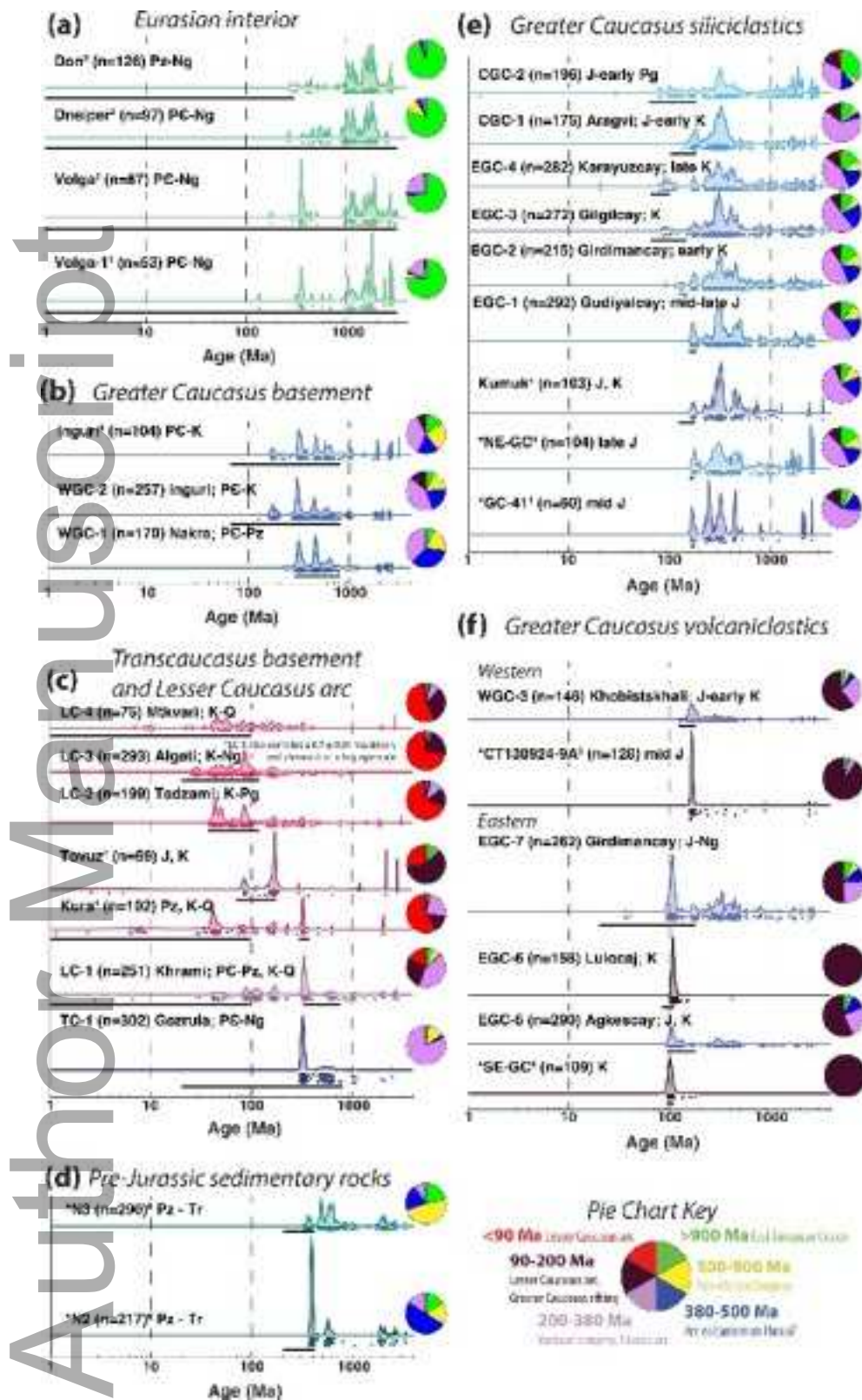
(a) BPC calculated relative to endmembers



(b) BPC values used as R, G, B values to color sample

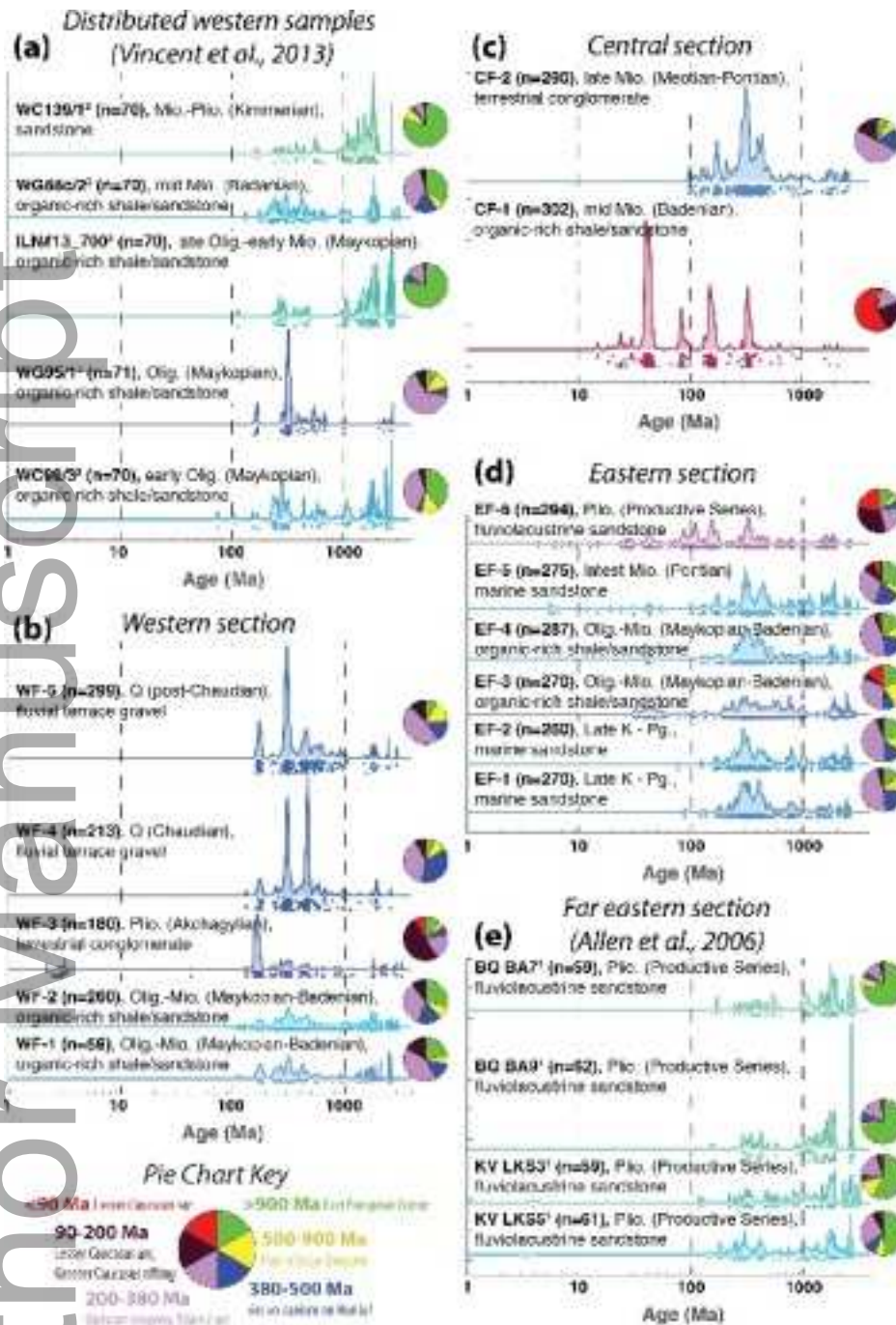


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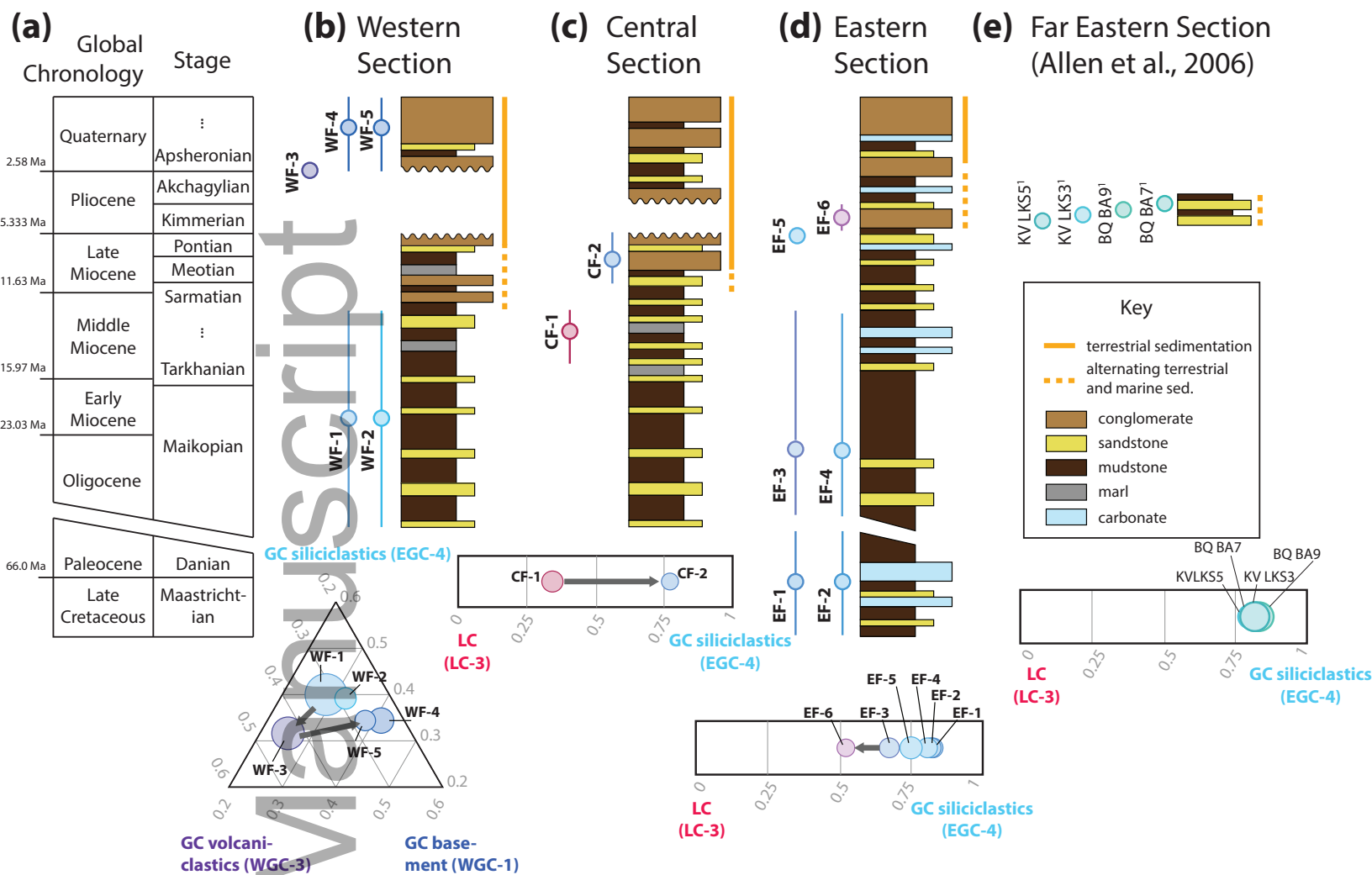


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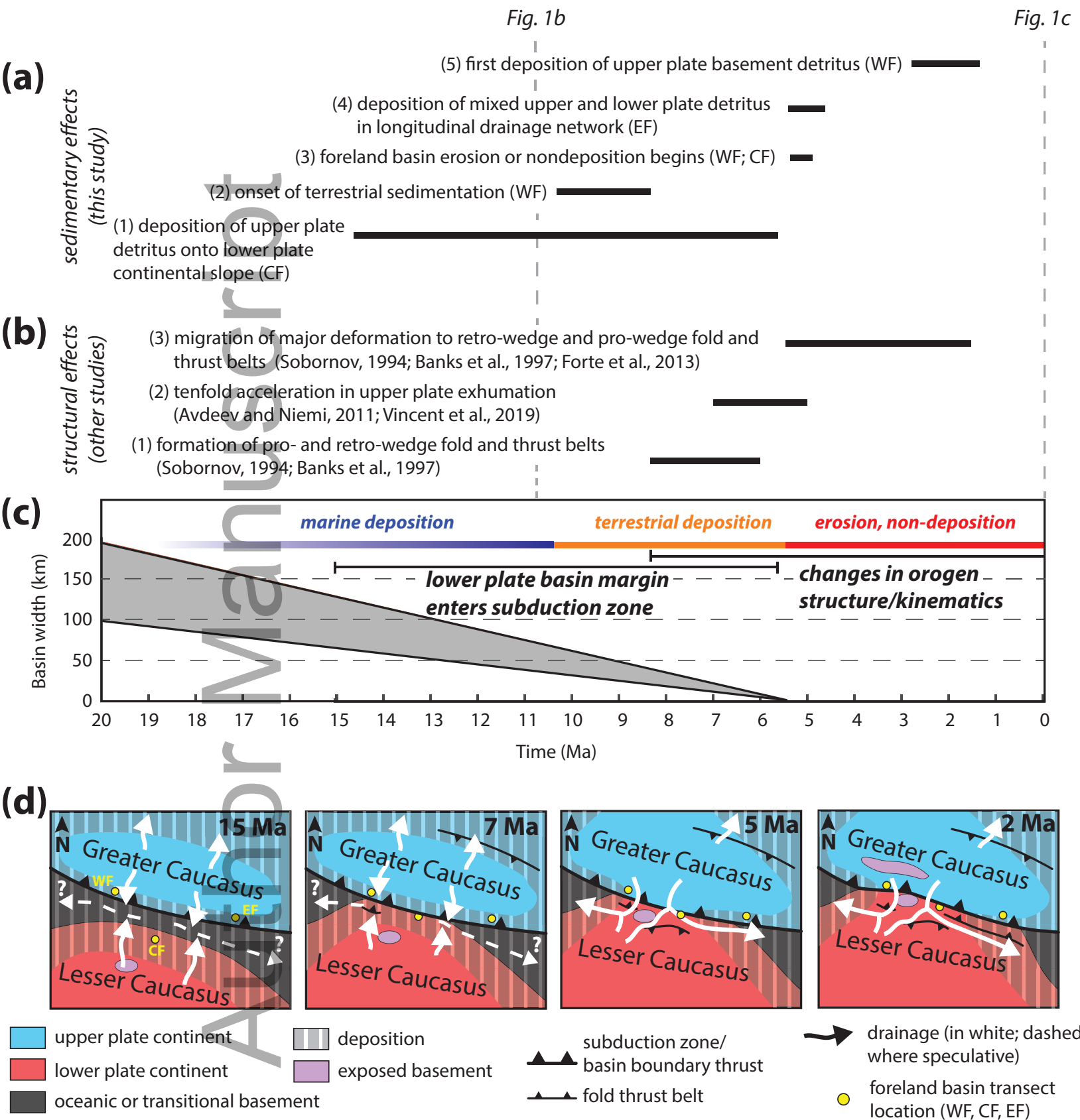
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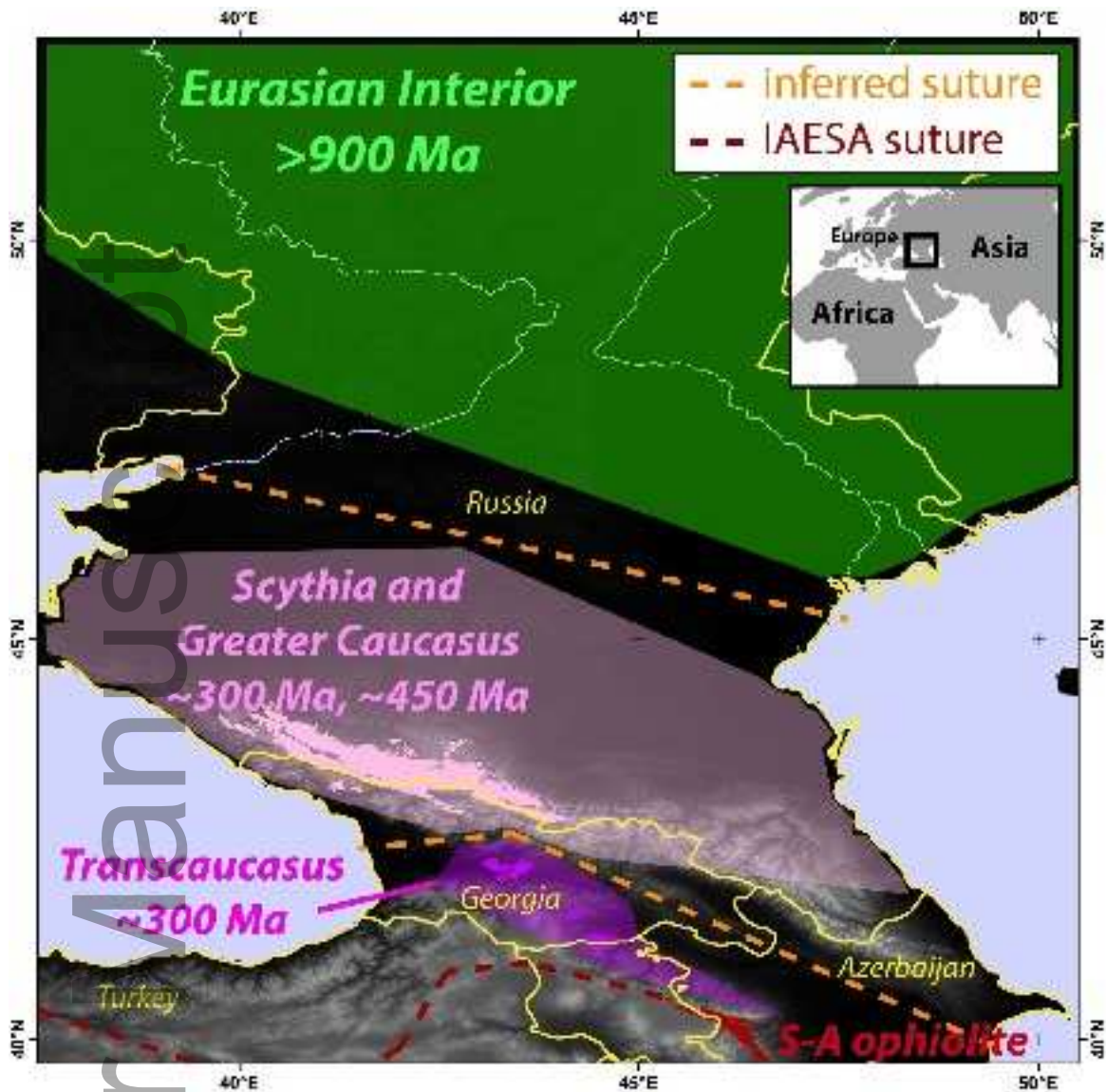
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