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FROM GONDWANA RIFTING TO ALPINE OROGENY: DETRITAL ZIRCON GEOCHRONOLOGIC AND PROVENANCE SIGNALS FROM THE KOPET DAGH BASIN (NE IRAN)

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ABSTRACT. The Kopet Dagh mountains in NE Iran exhibit a 7-km-thick continuous 14 15 sedimentary sequence recording detritus from exposed surrounding terranes from the last 175 Myr. This work presents a multi-disciplinary geochronologic and provenance analysis in an 16 17 attempt to identify and date major geologic events along the northern segment of the Tethys and reconstruct the regional tectonic history from Gondwana-related rifting until Alpine 18 19 orogeny. Sandstone framework, heavy mineral analysis, U-Pb dating of detrital zircons, and 20 Hf-isotope ratio measurements on dated zircons from Triassic to Paleocene sandstones indicate three main tectonic events that include early Silurian intracontinental rifting (opening of Paleo-21 22 Tethys), early Carboniferous rifting of a back-arc basin (Aghdarband Basin), and Late Triassic collisional to post-collisional magmatism(Paleo-Tethys collision). Mineralogical and age peak 23 considerations indicate that detritus was supplied from the south into the extensional Kopet 24 Dagh Basin during Middle Jurassic, while Cretaceous to Paleocene sandstones show signs of 25 26 increasing cannibalism. Keywords: Gondwana, Kopet Dagh Basin, Paleo-Tethys, Neo-Tethys, Rifting, Collision, 27

- 28 Detrital zircon, Hf isotopes, Provenance anlysis
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INTRODUCTION

The long-standing tectonic history between Gondwana and Eurasia includes several phases of rifting and collision, recorded by magmatic and metamorphic episodes in various domains of related orogenic belts; from the Alps in the west to the Himalayas in the east (for example

Sengör, 1990a; 1990b; Stampfli, 2000; Stampfli and Borel, 2002). The timing of main tectonic 34 35 episodes including rifting, subduction initiation, and collision are crucial for the reconstruction of plate tectonics (for example Stampfli and Borel, 2002). Geochronology and geochemistry of 36 magmatic and metamorphic rocks are widely applied in geodynamic studies to constrain the 37 timing and other characteristics of tectonic events (for example Barbarin, 1999; Brown, 2014). 38 However, the occurrence and surface exposure of such igneous and metamorphic terranes may 39 be limited in some tectono-sedimentary domains because of complete erosion or coverage by 40 younger sediments. In addition, the application of different dating methods on igneous and 41 metamorphic rocks provides a wide range of ages, which makes the exact timing of tectonic 42 43 events difficult. In fact, timing constraints for tectonic events based only on magmatic or 44 metamorphic rocks may be misleading. A more accurate detection of large-scale tectonic events can be found in clastic sedimentary successions, where the detritus source rapidly responds to 45 46 any plate tectonic reorganization (Cawood and others, 2012; Fedo and others, 2003). Therefore, provenance analysis, zircon geochronology, and zircon isotopic composition on detrital 47 sandstones may help to improve the precision when constraining the timing of tectonic events 48 (for example Cawood and others, 2012; Weltje and von Eynatten, 2004). 49

Several episodes of continental rifting and collision took place between Gondwana and Eurasia 50 51 since the Palaeozoic until the Cenozoic that ultimately climaxed in the Alpine-Himalayan orogeny (for example Sengör and others, 1984; Stampfli, 2000; Stampfli and Borel, 2002; 52 Stöcklin, 1974). Separation of several continental slivers and microcontinental blocks from 53 Gondwana occurred in two phases, coeval with the opening of two ocean basins, the Paleo-54 Tethys and Neo-Tethys oceans (for example Alavi, 1996; Stampfli, 2000; Şengör and others, 55 1984; Stöcklin, 1974). The opening of the Iranian segment of the Paleo-Tethys between the 56 Iranian continent (Gondwana) and the Turan platform (Eurasia) was suggested to have occurred 57 in the Early Palaeozoic (Ordovician-Silurian) based on ages of oceanic basalts (Boulin, 1988; 58 Stampfli, 2000). The subsequent northwards subduction of the Paleo-Tethys beneath Eurasia 59

initiated during the Late Devonian and continued until the Late Triassic as shown from dating 60 of the Darrehanjir-Mashhad ophiolites (for example Moghadam and others, 2015a; Stampfli, 61 2000; Wilmsen and others, 2009). The opening of a new ocean (Neo-Tethys Ocean) between 62 the Arabian plate and the Central Iran block at the end of the Permian went hand in hand with 63 the northward movement of the Central Iran block, resulting in the closure of the Paleo-Tethys 64 Ocean in the Late Triassic, and the consequent collision of the Iranian plate with the Turan 65 66 block; the Cimmerian orogeny (for example Berberian and King, 1981; Muttoni and others, 2009; Sengör, 1979; Zanchetta and others, 2013). This Cimmerian orogeny prevented further 67 shortening along the northern segment of the Tethys and triggered the onset of subduction of 68 69 the southern Neo-Tethys Ocean (for example Zanchetta and others, 2013).

70 The northward subduction of the Neo-Tethys Ocean beneath the Central Iran block was accompanied by the opening of the Greater Caucasus-South Caspian-Kopet Dagh basins 71 72 during the Middle Mesozoic, potentially in form of a larger back-arc basin (Alavi et al, 1997; Brunet and Cloetingh, 2003; Zanchi and others, 2006). The Neo-Tethys Ocean finally closed 73 74 during the collision of the Arabian plate and Central Iran block in late Oligocene, which resulted in the Alpine orogeny (Pirouz and others, 2017). This collision related to the closure of the Neo-75 76 Tethys resulted in tectonic inversion of previously extensional domains (Greater Caucasus, 77 South Caspian, and Kopet Dagh basins) along the Paleo-Tethys suture (for example Robert and others, 2014; Ruh and Vergés, 2018; Ruh, 2019). 78

The Kopet Dagh Basin contains a stratigraphic record covering the last ~175 Myr (Middle Jurassic to Neogene sedimentary succession; Afshar Harb, 1982a; Moussavi-Harami and Brenner, 1990). Detailed provenance analysis of this shallowing-upward (deep marine–marine– deltaic–continental) sequence allows i) to reconstruct the Mesozoic-Tertiary sedimentary evolution of the Kopet Dagh Basin, ii) to clarify the general tectonic setting associated with the Turan and Iran plates, and finally iii) to precisely constrain the timing of rifting and collision events related to the opening and closure of the Paleo-Tethys and Neo-Tethys oceans.

86	In this study, we present combined results from sandstone modal framework, heavy mineral
87	assemblage analysis, detrital zircon U-Pb age, and in-situ Hf isotope composition of Triassic
88	(pre-rift basement), and Middle Jurassic to Paleocene detrital sediments from the Kopet Dagh.
89	Results are interpreted to identify and characterize the respective source rocks in terms of ages
90	and genesis. Based on these data, we determine the provenance of sedimentary detritus and
91	reconstruct the variety of rift-related, continental arc, and post-collisional tectonic settings from
92	Gondwana-related rifting (opening of Paleo-Tethys) to Alpine orogeny (closure of Neo-
93	Tethys).
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95	GOLOGY OF THE KOPET DAGH
96	Tectonic Setting
97	The Kopet Dagh mountains in NE Iran strike over 700 km along the Iran–Turkmenistan border
98	and form the northern limit of the Alpine-Himalayan orogeny (Golonka, 2004: Robert and
99	others, 2014; fig. 1). The Main Kopet Dagh Fault (MKF) and the East Alborz and Binalud
100	mountains mark the northern and the southern boundary of the Kopet Dagh Basin, respectively
101	(fig. 1). Tectonically, the Kopet Dagh Basin formed as an extensional back-arc to the Neo-
102	Tethys subduction system farther to the south (Mangino and Priestley, 1998). The onset of
103	extension is marked by a major nonconformity at the base, and rapid sediment facies and
104	thickness variations in the Middle Jurassic (165 Ma to 170 Ma) deep-marine turbiditic
105	sediments (Kavoosi and others, 2009; Robert and others, 2014; Thomas and others, 1999). In
106	contrast to the Caspian Sea Basin in the west, there is no evidence for the formation of oceanic
107	crust beneath the Kopet Dagh Basin (Brunet and others, 2003). After this rifting event, the basin
108	experienced a phase of tectonic quiescence for at least 130 Myr until Oligocene-Miocene, when
109	the extensional basins along the Paleo-Tethys suture experienced tectonic inversion as a
110	consequence of the collision of Arabian and Eurasian plates to form the Zagros mountain range,

and due to newly arranged relative plate motions (Hollingsworth and others, 2010; Lyberis and 111 Manby, 1999). This tectonic inversion is synchronous with the onset of deposition of 112 continental sedimentary facies in the Kopet Dagh Basin (Robertand others, 2014) and tectonic 113 uplift of the Alborz mountains (Razaeian and others, 2012). Uplift of the Kopet Dagh mountains 114 resulted in widespread folding and faulting of the Jurassic to Tertiary stratigraphy and an 115 estimated N-S shortening of ~75 km (Allen and others, 2003; Lyberis and Manby, 1999). The 116 compressional phase continued for about 15 Myr, with an increased component of dextral 117 strike-slip movement since ~10 Ma (Hollingsworth and others, 2006). 118

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Stratigraphy

121 The study area is located in the eastern part of the Kopet Dagh Basin covered by the Darreh Gaz, Mashhad, Sarakhs, Torbat-e-Heydarieh, and Torbat-e-Jam 1:250,000 geological maps 122 published by the Geological Survey of Iran (figs. 1, 2; Afshar 1982a, 1982b; Afshar and others, 123 1986; Behrouzi and others, 1993; Vaezi Pour and others, 1992). An up to seven-kilometers-124 thick terrigenous and carbonate s edimentary succession was deposited into the Kopet Dagh 125 Basin between Middle Jurassic to Neogene (figs. 2, 3; Moussavi-Harami and Brenner, 1990). 126 The Middle Jurassic (late Bajocian to Bathonian) intervals of deep marine turbiditic 127 conglomerate, thin- and thick-bedded sandstones, siltstone, and mudstone (Kashafrud 128 Formation) are the oldest sediments deposited into the Kopet Dagh Basin (for example 129 Poursoltani and Gibling, 2011; Poursoltani, and others, 2007). The Middle Jurassic sediments 130 rest unconformably on Triassic basic intrusions and volcanogenic sedimentary rocks and 131 exhibit variations in thickness across major extensional basement faults, indicating a syn-rifting 132 depositional environment (Robert and others, 2014; Thomas and others, 1999). Thickness of 133 the turbiditic sediments vary between 280 m to over 2000 m in the East Kopet Dagh Basin (fig. 134 3). The Middle Jurassic turbiditic succession is unconformably overlain by Upper Jurassic 135 (Callovian-Kimmeridgian) thick (>1200 m) marine carbonates and shallow marine, coastal 136

plain silisiclastic sediments (Mozduran Formation; Kavoosi and others, 2009; Lasemi, 1995; 137 fig. 3). Thick fluvial siliciclastic sediments of the Early Cretaceous (Surijeh Formation) record 138 epicontinental sea retogradation in the eastern part of the Kopet Dagh Basin (fig. 3; Moussavi-139 Harami and Brenner, 1990). Contemporaneous deposition of marine carbonate indicates the 140 141 presence of a marine environment in the western Kopet Dagh Basin. Further transgression established a marine depositional environment throughout the entire Kopet Dagh Basin until 142 Late Cretaceous (Turonian). This transgressions system is characterized by intervals of oolitic 143 limestone (Tirgan Formation), marl and shale (Sarcheshmeh Formation), shale (Sanganeh 144 Formation), and glauconitic-carbonatic sandstone and shale (Aitamir Formation; fig. 3). This 145 146 wide range of lithologies indicates sea level fluctuation and depositional environments 147 represented by tidal flats, lagoons, carbonate ramps, continetal shelfs, and shallow marineshorefaces. A short-lived sea retogradation during the Late Cretaceous (Turonian) is responsible 148 for an unconformity between the shallow marine glauconitic shale (Aitamir Formation) and 149 shallow marine marl and limestones (Abderaz Formation; fig. 3). Between the Turonian until 150 151 the Paleocene, the East Kopet Dagh Basin experienced other transgressional episodes characterized by marine environments including the deposition of shallow marine marl and 152 153 limestones (Abderaz Formation), shelf and open marine marl and shale (Abtalkh Formation), 154 shallow marine and shoreface glauconitic sandstone (Neyzar Formation), and shallow carbonate platform sandy limestones (Kalat Formation; fig. 3). Basin-scale sea retrogradation 155 and surface exposure of strata is recorded by erosional surfaces within the uppermost part of 156 157 Late Cretaceous sandy limestones (Kalat Formation; Mahboubi and others, 2006). In early Paleocene, reddish fluvial and lacustrine mudstones and sandstones (Pestehleigh Formation) 158 159 were unconformably deposited onto uppermost Cretaceous marine sediments. During the late Paleocene, the Kopet Dagh Basin gradualy changed from a shallow continental sedimentary 160 environment to a shallow carbonate paltform with marl and limestone sediments (Chehel 161 Kaman Formation). The basin subsidence continued until Eocene-early Oligocene, when 162

marine shale and sandstones (Khangiran Formation) conformably overlayed late Paleocene
sediments. Repeated minor phases of transgressions and regressions followed into the Miocene,
until the basin was tectonically inverted duing Alpine orogeny, accompanied by deposition of
continental conglomerates, sandstones, and shale (Neogene red beds; fig. 3).

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METHODS

Nineteen sandstone samples including one sandstone sample of a lens within Triassic 169 polymictic conglomerate, and eighteen well-cemented medium-grained Jurassic to Paleocene 170 turbiditic, fluvial-deltaic, and continental sandstone samples were investigated (with prefix KD-171 01–20 in the ETH collection). Studied sandstones represent five different litho-stratigraphic 172 formations (Qara-Gheitan, Kashafrud, Shurijeh, Aitamir, and Pestehleigh formations) of the 173 Kopet Dagh Basin (figs 1, 2, 3 and table S1). We combined sandstone framework composition 174 and heavy mineral analysis, detrital zircon U-Pb geochronology, and in-situ Hf-isotopic 175 composition to reconstruct the lithological composition of the source-rocks and the tectonic 176 evolution of the source area. 177

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Sandstone thin section

Sandstones modal framework analysis was performed in thin sections stained for carbonates 179 and feldspars with the Gazzi-Dickinson method (Dickson, 1966; Norman, 1974). For statistical 180 reliability, at least 300 lithic fragments were counted in each thin section (Folk, 1980). The 181 Zuffa method was applied to count detrital grains (Zuffa, 1985). Sand size lithic fragments 182 183 (>63 µm) within rock fragments were counted as monomineralic grains. Results were converted to percentages for compositional comparison (Weltje and von Eynatten, 2004; table 184 S2). Five standard complementary triangular diagrams were used to display the data (QFL, 185 186 QmFLt, LvhLsLm, QmPK, and QpLvLsm; Folk, 1980; Dickinson, 1985).

Approximately 2-3 kg of fresh rock were collected at each sample site for heavy mineral 189 analysis. To obtain transparent heavy mineral fractions, sandstones were crushed with the 190 SelFrag apparatus batch equipment using high voltage (130-150 kV) pulse power technology, 191 which liberates morphologically intact minerals. From the <2 mm sieved fraction, carbonate 192 was dissolved in warm (60-70°C), 10%-acetic acid. Heavy minerals were pulled out in 193 separation funnels (Mange and Maurer, 1992) from 63 µm to 400 µm sieve fractions using 194 bromoform (density 2.88 g/cm³). The bulk heavy mineral fractions were mounted in piperine 195 (Martens, 1932) between a glass slab and a cover. Identification and quantification were 196 carried out under a petrographic microscope by applying the mid-point ribbon and fleet 197 198 counting methods. At least 200 grains were counted per sample (Mange and Maurer, 1992; table

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SI3).

Zircon geochronology

Zircons were extracted from additional 2-3 kg of fresh sandstones crushed with the SelFrag 201 202 apparatus. Standard Holman-Wilfley shaking tables and magnetic separation methods (Frantz 203 magnetic separator) delivered mineral fractions enriched in zircons. Zircons of grain fraction 60-250 µm (non-magnetic) were handpicked under a binocular microscope. Selected grains 204 were mounted in U- and Pb-free resin, polished to expose the internal parts of the minerals, and 205 coated with carbon. Suitable zircon domains and inclusions were identified by 206 cathodoluminescence (CL) and backscattered-electron (BSE) images obtained by split screen 207 208 on a CamScan CS44 Scandunk Viermalning electron microscope (SEM; Tescan a.s., Brno) at ETH Zurich, and analyzed with laser ablation inductively coupled mass spectrometry (LA-ICP-209 210 MS) at ETH Zurich with an excimer laser (ArF 193 nm). Zircon characteristics were interpreted following the approach by Corfu and others (2003). 211

212 The laser ablation ICP-MS analyses were performed with a RESOlution (Australian Scientific

213 Instruments/Applied Spectra) laser ablation system at ETH Zurich equipped with a dual-volume

S-155 ablation cell (Laurin Technic, Australia), coupled to an Element XR (Thermo Scientific, 214 215 Germany) sector-field ICP-MS. A laser repetition rate of 5 Hz, energy density of ca. 2.5 J/cm², and spot diameter of 30 µm were applied. The carrier gas consisted of high-purity He (ca. 0.7 216 L/min) and make-up Ar (ca. 1 L/min) from the ICP-MS. Data acquisition time per spot was 217 218 about 1 minute (30 s gas blank + 30 s ablation). The data were processed with the Igor Pro Iolite v2.5 software (Hellstrom and others, 2008), using the VizualAge data reduction scheme (Petrus 219 220 and Kamber, 2012). Laser-induced element fractionation (after Paton and others, 2010), instrumental mass discrimination, and drift through the analytical session were corrected by 221 standard bracketing against zircon reference material "GJ-1" (using isotope ratios 222 223 recommended by Horstwood and others, 2016). The quoted uncertainties for each individual 224 analysis correspond to the internal error and propagated uncertainty based on the scatter of the primary reference material (see Paton and others, 2010). The accuracy and reproducibility 225 226 within each run of analysis were monitored by periodic measurements of zircon reference materials "Plešovice" (337 Ma; Sláma and others 2008) and "91500" (1065 Ma; Wiedenbeck 227 228 and others 1995). More details about the analytical conditions, the data from unknowns and reference materials are provided in table S5. Concordia diagrams were obtained using 229 ISOPLOT v.3.0 (Ludwig, 2003). A concordant age is given by the overlap of the error ellipse 230 with the Concordia age curve. In this study, ages with discordance >5% are not included for 231 further interpretation (table S4). The frequency U-Pb age distribution diagram or probability 232 233 density plot described by Ludwig (2003) includes a histogram representing the number of individual zircon grains within a short age range, and the probability curve depicts the mean 234 age peaks of the contained age populations in one sample. 235

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Zircon Lu-Hf isotopic analyses

Lu–Hf analyses were obtained by laser ablation multi-collection inductively coupled plasma
mass spectrometry (LA-MC-ICP-MS) at ETH Zurich, using a RESOlution (ASI/Applied
Spectra) excimer (ArF) 193 nm laser ablation system attached to Nu Plasma II (Nu Instruments)

multi-collector ICP-MS. A laser repetition rate of 5 Hz, a spot diameter of 50 µm, and a laser 240 energy density of ca. 4 J/cm² was applied. Ablation was performed in a dual-volume, fast-241 washout S-155 ablation cell (Laurin Technic) fluxed with carrier gas consisting of ca. 0.37 242 L/min He and make-up gas consisting of ca. 0.92 L/min Ar. A typical run consisted of 40 s of 243 background measurement followed by 40 s of sample ablation with an integration time of 0.5 244 s. The ICP-MS instrument was optimized for maximum sensitivity on ¹⁸⁰Hf, and optimal peak 245 shape and coincidence for Yb, Lu, and Hf isotopes. The list of acquired masses and 246 corresponding Faraday cups are provided in table S7. 247

The resulting intensities were processed offline with the Igor Pro Iolite v2.5 software, using an 248 in-house data reduction scheme (Hellstrom and others, 2008). Background-subtracted 249 intensities were used to calculate isotope ratios, which were corrected for instrumental mass 250 bias by normalization to the natural abundance ratios ¹⁷³Yb/¹⁷¹Yb (for both Yb and Lu isotope 251 ratios) and ¹⁷⁹Hf/¹⁷⁷Hf (for Hf isotope ratios) of Chu and others (2002). The isobaric 252 interferences of ¹⁷⁶Yb and ¹⁷⁶Lu on ¹⁷⁶Hf were subsequently corrected using the natural 253 abundance ratios of 176 Yb/ 173 Yb = 0.79502 and 176 Lu/ 175 Lu = 0.02656. Accuracy and external 254 reproducibility of the method were controlled by repeated analyses of reference zircon 255 standards "GJ" (Morel and others, 2008), "Plešovice" (Sláma and others, 2008), "Mud Tank" 256 and "Temora" (Woodhead and Hergt, 2005), and "QGNG" (Woodhead and others, 2004). The 257 ¹⁷⁶Hf/¹⁷⁷Hf ratios obtained on all reference materials are within uncertainties identical to the 258 recommended values. Calculations of initial ¹⁷⁶Hf/¹⁷⁷Hf ratios and ϵ -Hf were performed 259 applying the ¹⁷⁶Lu decay constant of Scherer and others (2001) and ¹⁷⁶Hf/¹⁷⁷Hf and ¹⁷⁶Lu/¹⁷⁷Hf 260 values for the chondritic uniform reservoir (CHUR) recommended by Bouvier and others 261 (2008). The quoted uncertainties on initial Hf isotopic composition correspond to the analytical 262 uncertainty and the average 2 S.D (table S6). Reproducibility of the initial Hf isotopic 263 compositions of all measured zircon reference materials (table S7), were propagated by 264

quadratic addition. More details about the analytical conditions, the data from unknowns, andreference materials are provided in table S7.

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RESULTS

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Sandstone frame work

The studied Triassic and Jurassic sandstones are mainly classified as feldspathic litharenite and 270 lithic arkose (fig. 4A). Cretaceous sandstones are classified as subarkose and sublitharenite. 271 272 The Paleocene sandstone sample is a litharenite. Quartz grains of all samples are mostly monocrystalline (~50% in Triassic, 50-75% in Middle Jurassic, and >80% in Cretaceous and 273 Paleocene sandstones; figs. 4A, 4B, 4C and table S2). K-feldspars are dominant in feldspar 274 grains (~73% in the Jurassic, and ~80% in the Cretaceous and Paleocene sandstones; fig. 4B 275 and table S2). From Jurassic to Cretaceous and Paleocene sandstones, the percentage of 276 277 monocrystalline quartz and K-feldspar grains increases, while the percentage of polycrystalline quartz and plagioclase grains decreases (figs. 4C and 4d). The Triassic sandstone is rich in 278 volcanics (~55%), with less sedimentary and metamorphic lithics (~45%). Jurassic sandstones 279 280 are composed of sedimentary (~60%), volcanic (~30%), and metamorphic (~10%) rock fragments. In Cretaceous and Paleocene sandstones, sedimentary and metamorphic rock 281 fragments (together >95%) are dominant, while volcanic lithics are less common (<5%). In 282 both Jurassic and Cretaceous sandstones, sedimentary lithic grains mainly comprise siltstone, 283 limestone, and dolomite, whereas metamorphic lithic grains generally consist of low-grade to 284 285 medium-grade slate, phyllites, and schists. Volcanic lithic grains mainly represent andesite and volcanic glass (figs. 4C, 4D and table S2). Dominantly volcanic lithics (figs. 4C and 4D) and 286 pyroxene minerals (fig. 5) point toward a transitional arc as the source terrane of the Triassic 287 288 sandstone, whereas Jurassic samples point to a dissected arc and mixed nature of the source terranes (figs. 4E and 4F). Cretaceous and Paleocene sandstones show a recycled orogenic 289

source based on their rock fragment distributions, indicating mineralogical maturity (figs. 4E

291 and 4F).

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Heavy mineral study

Heavy mineral assemblages of sandstones from the Kopet Dagh are characterized as follows: 294 (1) A group of ultra-stable heavy minerals including zircon, tourmaline, and rutile (ZTR), 295 sphene, monazite, and less stable apatite that are typically derived from acidic to intermediate 296 magmatic rocks and recycled siliciclastic sandstones. The amount of ZTR ranges from 22% of 297 total grain count in the Triassic sandstone and reaches up to 53% of total grain count in Jurassic 298 and up to 74.5% in Cretaceous sandstones (Fig. 5). The percentage of apatite grains ranges 299 between 9% in Triassic and 17.5% in Jurassic sandstones (fig. 5 and table S3). (2) A group of 300 meta-stable heavy minerals including pyroxene and amphibole attributed to mafic magmatic 301 source rocks. The amount of pyroxene in Triassic sandstones reaches up to 64% of the total 302 heavy mineral grains, whereas Jurassic and Cretaceous sandstones contain 24% and 1.5% 303 pyroxene, respectively (fig. 5 and table S3). (3) A third group describes meta-stable minerals, 304 305 including the epidote group, garnet, chloritoid, andalusite, and staurolite, that are common heavy minerals related to medium- to high-grade metamorphic rocks. The percentage of these 306 minerals varies from 1% of total counted grains in Triassic sandstones to 17% of total counted 307 grains in Paleocene sandstones (fig. 5 and table S3). (4) Fourth, Cr-spinel (up to 4% of total 308 grain count; fig. 5 and table S3) indicates mafic-ultramafic rocks as source of detritus. 309

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Detrital zircon U-Pb ages

Laser ablation ICP-MS U-Pb dating on 2478 detrital zircon grains was conducted on one Lower
Triassic low-grade meta-sandstone (Qara-Gheitan Formation), five Jurassic turbiditic
sandstones (Kashafrud Formation), three Lower Cretaceous fluvial-deltaic sandstones (Shurijeh

Formation), and one Paleocene continental sandstone (Pestehleigh Formation; figs. 2, 3, 6, 7, 8,

316 9, and tables S1 and S4).

Late Triassic (217 Ma) and early Paleoproterozoic (2374 Ma) zircons are the youngest and 317 oldest zircons of 232 analysed detrital zircons from the Lower Triassic sandstone (fig. 7; sample 318 KD-08). This sandstone reveals a single probability peak at 337 Ma (early Carboniferous). Five 319 Jurassic sandstones show zircon age distributions between 154 Ma (Late Jurassic) and 2966 Ma 320 321 (Meso-Archean) based on a total of 1191 dated grains (Fig. 7; samples KD-01, KD-03, KD-06, KD-10, KD-15). Detrital zircons of the Jurassic sandstones are peaking at 232 Ma (Early 322 Triassic), 343 Ma (early Carboniferous), 434 Ma (early Silurian), 643 Ma (middle 323 324 Neoproterozoic), and 1833 Ma (late Paleoproterozoic; Fig. 7). A total of 865 zircons from three Lower Cretaceous sandstones revealed ages between Early Cretaceous (116 Ma) to 325 Mesoarchean (2861 Ma) (fig. 8; samples KD-02, KD-09, KD-18). The Lower Cretaceous 326 327 sandstones show a similar zircon age distribution as the Jurassic sandstones, but with an additional zircon probability peak at 132 Ma (Early Cretaceous; fig. 8). The Paleocene 328 329 sandstone shows ages ranging between 119 Ma (Early Cretaceous) and 2781 Ma (Neoarchean) from 190 dated zircon grains (fig. 8; sample KD-14). The probability peaks of Paleocene 330 sandstone zircon ages are similar to the ones of Cretaceous sandstones (fig. 8). The combined 331 332 zircon ages call attention to late Paleoproterozoic, middle Neoproterozoic, early Silurian, early Carboniferous, Late Triassic, and Early Cretaceous protoliths in the potential source areas (figs. 333 9 and 10). 334

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Hf isotopic composition

Hafnium isotopic composition analysis was conducted on 151 concordantly dated zircon grains with internal growth structures that were large and thick enough to be examined with laser beams. The overall Hf isotopic compositions (epsilon hafnium values: ϵ -Hf_(t)) of Triassic to Paleocene sandstones range from -23 to +15 (fig. 11). Late Paleoproterozoic zircon grains demonstrate evolved ε -Hf_(t) values from -13 to +1. Early Silurian zircons show a mixed positive and negative pattern of ε -Hf_(t) from -2 to +7 (fig. 11). Early Carboniferous zircons show positive ε -Hf_(t) values of -1 to +12 with four grains between -17 and -4 (fig. 11). Similar to early Silurian zircons, the Late Triassic zircons show mixed positive and negative ε -Hf_(t) values from -3 to +7 with one outlier of -15. The only analysed Early Cretaceous zircon shows a ε -Hf_(t) value of +7.6 (fig. 11).

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DISCUSSION

The presented multi-proxy study of Triassic to Palaeocene sandstones from the Kopet Dagh mountains refine our understanding of the corresponding sediment sources and allows for an improved reconstruction of the tectono-sedimentary evolution of the tectonically inverted Kopet Dagh Basin.

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Evidence from sandstone frame work

The sandstone modal framework analysis of the sandstone lens within a Triassic 354 Formation) polymathic conglomerate (Qara-Gheitan stipulate transitional 355 a magmatic arc provenance (figs. 4E and 4F). However, petrographic analyses on the 356 Qara-Gheitan sandstones by Ghaemi (2009) specify provenance from a dissected 357 Additionally, geochemical magmatic arc. analysis of large granodiorites 358 and tonalities pebbles of the Qara-Gheitan conglomerate by Zanchetta and others, (2013) 359 suggests an active margin setting with prevailing of I-type plutonic and high-K 360 volcanic rocks. The discrepancy between our results and previously published data 361 be explained sampling of different within 362 can by sandstone lenses the inhomogeneous alluvial Qara-Gheitan Formation. 363

364 The mineralogical composition of Middle Jurassic sandstones of the Kashafrud 365 Formation composed of sedimentary and volcanic lithic fragments with subordinate

metamorphic lithics (figs. 4C and 4D) suggest a provenance from a dissected 366 magmatic arc and mixed-recycled orogeny (figs. 4E and 4F). Modal framework and 367 paleocurrent analysis along with stratigraphic studies of Middle Jurassic sandstones 368 by Poursoltani and others, (2007) and Poursoltani and Gibling (2011) are in 369 agreement with our results. In addition, they suggested that the detritus was supplied 370 from an active margin in the southeast to the extensional Kopet Dagh Basin in the 371 372 northwest through channel systems to slope and basin-floor fans forming fluvial deltaic (sandstone), distal 373 (basal conglomerate), and proximal to turbiditic (sandstone and mudstone) facies. 374

375 Lower Cretaceous sandstones (Shurijeh Formation) show higher mineralogical and textural maturity compared to Middle Jurassic sandstones (figs. 4A, 4B, 4C and 4D) 376 and indicate a recycled orogenic provenance (figs. 4E and 4F). In addition, modal 377 378 framework analysis by Mortazavi and others (2013) on Lower Cretaceous sandstones supports a provenance characterized by recycled orogenic belts and 379 craton interior signature. Paleocurrent directions of Lower Cretaceous sandstones 380 suggest that sediments were supplied from the south and southwest into the Kopet 381 Dagh Basin (Moussavi-Harami and Brenner, 1990). Therefore, Jurassic sandstones 382 383 represent a potential source for second-cycle Lower Cretaceous sandstones. Paleocene continental sandstones (Pestehleigh Formation) show relativly higher 384 mineralogical maturity compared to Triassic-Jurassic sandstones (figs. 4A, 4B, 4C 385 and 4D), indicating a recycled orogenic provenance (figs. 4E and 4F). This 386 interpretation agrees with findings of Moussavi-Harami (1993), who suggested that 387 northward and northwestward thinning of coarse-grained Paleocene sandstone layers 388 indicates that the detritus was supplied from the recycling of older sediments in the 389 390 south and southeast.

Evidence from heavy minerals assemblage

The heavy mineral assemblage of the Triassic sandstone with dominant pyroxene minerals (fig. 392 5 and table 3) indicates a source from mafic magmatic rocks (probably a magmatic arc), which 393 is in concordance with sandstone modal framework results (figs. 4E and 4F). In addition, the 394 internal textures (CL images) and Th/U ratios of detrital zircons from pebbles of the Triassic 395 conglomerate suggest a magmatic origin for these zircons (Zanchetta and others, 2013). An 396 increase in ultra-stable minerals (ZTR) along with a decreasing percentage of pyroxene 397 minerals in the heavy mineral fraction of Jurassic sandstones (fig. 5 and table 3) imply first-398 cycle sandstones that were transported over a long distance, or second-cycle sandstone formed 399 by erosion and deposition of pyroclastic-volcanoclastic rocks in a deep-marine basin 400 401 (Poursoltani and others, 2007). A significant increase of clastic sedimentary lithics compared to the Triassic sandstone and first appearance of garnets in Jurassic sandstones (fig. 5 and table 402 3) point toward metamorphic rocks as a minor additional source of detritus. Poursoltani and 403 Gibling (2011) reported ultra-stable heavy minerals (ZTR), along with apatite and garnet as the 404 main heavy mineral spectrum in the Jurassic sandstones, coinciding with our findings. Early 405 406 Cretaceous sandstones indicate a high percentage of ultra-stable minerals (ZTR) and a reduced quantity of pyroxene minerals in the heavy mineral assemblages with a much significant 407 intensity. Vice versa, the amount of pyroxene (20%) in Jurassic sandstones decreases to 1.5% 408 in Cretaceous sandstones (fig. 5 and table 3). This indicates that the heavy mineral content was 409 partly controlled by cannibalism and recycling of older sandstones. An increase of 410 monocrystalline quartz content along with a decrease of less stable volcanic lithics in Early 411 Cretaceous sandstones compared to older lithologies provides further evidence for recycling of 412 Jurassic sandstones (table 3). In addition, based on geochemical analysis of Early Cretaceous 413 sandstones, Mortazavi and others, (2013) suggested moderate to high weathering of the 414 sediments during transportation before deposition, which is in agreement with recycling of 415 older sediments. However, the effect of diagenetic processes on the decrease of metastable 416

heavy minerals (mostly pyroxene group) and volcanic lithics, and partial alteration of feldspars
to clay minerals should not be ignored (Mortazavi and others, 2014; Moussavi-Harami and
Brenner, 1993).

Paleocene sandstones indicate a heavy mineral spectrum similar to the one of Early Cretaceous sandstones. However, the amount of garnet increases from 3.5% in Early Cretaceous sandstones to 15.5% in Paleocene sandstones. This implies that erosion of garnet-bearing metamorphic rocks, along with recycling of the older sandstones, were the main sources of detritus in the Palaeocene. The most probable source of garnet and metamorphic lithic grains are Carbo-Permian metamorphic rocks to the southeast of the Kopet Dagh Basin (figs. 2 and 3).

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Evidence from Detrital Zircon U-Pb Ages and Hf Isotopes

The 2477 analyzed detrital zircon grains from the Lower Triassic, Middle Jurassic, Lower Cretaceous, and Paleocene turbiditic, deltaic-fluvial, and continental sandstones yielded apparent ages ranging from ca. 119 Ma to ca. 2966 Ma (figs. 7, 8, 9 and 10). Six major U-Pb age groups are identified in the Kopet Dagh detrital zircon.

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Mesoarchean to Paleoproterozoic Zircons

The oldest zircons create a broad heterogeneous age spectrum from Mesoarchean to 433 434 Paleoproterozoic (2966-1600 Ma) with a main peak in the Paleoproterozoic (1833 Ma; fig. 9). Zircons of this population contain ~11% of total dated zircons and are characterized by anhedral 435 436 to rounded grains without internal zoning (>80%), and a wide range of Th/U ratios (between 0.01 and 2.39; fig. 6 and table S4). The ε -Hf_(t) values of Mesoarchean to early Paleoproterozoic 437 zircons represent a mixture of positive and negative values (-6.9 to +10.3), while late 438 Paleoproterozoic zircons show dominantly negative values (-13.6 to -2.3) with only one grain 439 of positive value (+ 1.4; fig. 11 and table S6). Possibly, these are inherited magmatic zircons or 440 fragments of older reworked zircons from crystalline rocks of a craton. Because of the 441 442 probability of repetitive reworking of detrital zircons through several tectonic cycles since the

Archean, the possible metamorphic/igneous source area remains undefined. However, similar 443 444 age spectra (with a major peak at 2.5 Ga and a minor peak at 1.8 Ga) were reported in Neoproterozoic to Cambrian, lower Permian and Middle Jurassic, and Devonian sandstones of 445 the Alborz mountains (Horton and others, 2008; Moghadam and others, 2017). Also, 446 447 Mesoarchean to Paleoproterozoic zircons were reported in Cretaceous to Eocene sandstones of Azerbaijan Basin (Mohammadi and others, 2020a), Eocene to Oligocene sandstones from the 448 449 Sistan Basin (Mohammadi and others, 2016a), and Cretaceous to Miocene sandstones of the Makran Basin in Iran (Mohammadi and others, 2016b; 2017). In addition, similar age spectra 450 were reported in Paleozoic to Triassic sandstones of the Taurides mountains in Turkey (Abbo 451 452 and others, 2015). There are no reported occurrences of Archean to Paleoproterozoic zircons in 453 the Iranian or Anatolian continental crust (for example Ramezani and Tucker, 2003; Moghadam, and others, 2017; Moghadam, and others, 2020). However, Archean to 454 455 Paleoproterozoic zircons with main peaks at 2.5 Ga and 1.8 Ga were identified from metamorphic basement of the Afghan block (Faryad and others, 2016), the Tibetan terranes 456 457 including Songpan Ganzi (Weislogel and others, 2006) and Nan Shan-Qilian Shan-Altun Shan (Gehrels and others, 2011), and the Tarim craton (Gehrels and others, 2011). The lack of a 1.8 458 459 Ga and presence of a 1.9 Ga zircon age peak in the Archean to Paleoproterozoic age spectrum 460 of Iberia and African Neoproterozoic to Cambrian sandstones (Abdelsalam and others, 2002) 461 excludes the Saharan meta-craton as a potential source area. In fact, the source of Archean to Paleoproterozoic zircons in the Kopet Dagh strata remains enigmatic. Restricting conjecture to 462 463 the closest cratons points to the nearby Afghan Block with Archean to Paleoproterozoic metamorphic and igneous rocks (Faryad and others, 2016) as an original source area, which 464 supplied detritus to the Kopet Dagh Basin through repetitive recycling of sedimentary 465 successions (fig. 10). 466

Neoproterozoic Zircons

468 The second age population includes Neoproterozoic detrital zircons (1000-542 Ma) with a main peak at 643 Ma (fig. 9). This population represents ~18% of the total dated zircons. 469 Neoproterozoic zircons are mostly rounded or anhedral grains with sporadic magmatic or 470 metamorphic growth zoning or lack zoning (fig. 6). Similar to Archean to Paleoproterozoic 471 zircons, Neoproterozoic zircons show Th/U ratios with a wide range between 0.01 and 2.92 472 473 (table S4), which indicate both magmatic and metamorphic sources. Due to different grain shapes and internal growth structures, Neoproterozoic zircons are probably remnants of older 474 recycled grains or inherited cores from continental crystalline rocks. The E-Hf(t) values of 475 Neoproterozoic zircons range between -19.3 and +11.3 (fig. 11 and table S6). Neoproterozoic 476 (Pan-African) detrital zircons were reported from several Gondwana-related tectono-477 stratigraphic domains such as Iran (for example Hassanzadeh and others, 2008; Hosseini and 478 479 others, 2015; Nutman and others, 2014; Ramezani and Tucker, 2003), the Taurides (Abbo and others, 2015), Libya (Meinhold and others, 2013), the Arabian-Nubian Shield (Morag and 480 481 others, 2012), and north Africa (Abati and others, 2010; Avigad and others, 2012). Neoproterozoic detrital zircons with a main peak at 559-537 Ma (Cadomian age) were 482 483 furthermore reported from Alborz and Zagros Neoproterozoic to Paleozoic strata (Horton and 484 others, 2008; Moghadam and others, 2017). In addition, the Neoproterozoic detrital zircons without internal zoning with a main peak at 640-610 Ma were reported from Central Iranian 485 Neoproterozoic to Cambrian sandstones (Honarmand and others, 2016). Nutman and others 486 487 (2014) reported 637 Ma U-Pb ages of magmatic zircons from the Central Iran crystalline basement (Chadegan orthogneiss). Therefore, repetitive recycling of the Cryogenian to 488 Ediacaran crystalline basement of Central Iran is a plausible source of the Neoproterozoic 489 detrital zircons of Kopet Dagh strata (fig. 10). 490

Ordovician to Silurian Zircons

493 The third age group denotes Ordovician to Silurian zircons (465-422 Ma) with a peak at 437 Ma with subhedral to euhedral shapes, dominantly magmatic zoning, and Th/U ratios above 0.5 494 (fig. 6 and table S4), demonstrating an abundant magmatic source. This population contains 495 ~9.3% of the total dated zircons (table S4). ε -Hf_(t) values (-1.9 to +6.7, only one grain with -496 8.9; fig. 11 and table S6) suggest rift-related magmatic rocks as detritus source (for example 497 Vervoort and Blichert-Toft, 1999). Presented zircon ages correspond to major rifting episodes 498 that occurred from late Ordovician to Silurian along the northern Gondwana margins 499 (Moghadam and others, 2020; Ruban, 2007; Stampfli, 2000), which correspond to the 500 501 separation of the Hun terranes from Gondwana along with the opening of Paleo-Tethys Ocean 502 (Moghadam and others, 2020). Late Ordovician to middle Silurian rift-related magmatism was furthermore reported from northern Central Iran (for example Nowrouzi and others, 2014), 503 504 eastern Alborz (Derakhshi and Ghasemi, 2015; Moghadam and others, 2020), and Anatolia (for example Topuz and others, 2020; Topuz and others, 2021). In the eastern Alborz, basaltic and 505 basaltic-andesitic magmatism during the Silurian were reported (local name: Soltan Maidan 506 Complex; Derakhshi and Ghasemi, 2015; Ghavidel-Syooki and others, 2011). Geochemistry of 507 this volcanic units shows alkaline affinity with an intra-plate and continental-rift setting 508 509 (Derakhshi and Ghasemi, 2015). The zircon U-Pb ages of rounded granitic pebbles from conglomerate layers at the base and in middle of the afore-mentioned volcanic unit indicate an 510 early Silurian genesis (434 ± 6.4 Ma; Ghavidel-Syooki and others, 2011). Furthermore, zircon 511 512 U-Pb ages of A-type granitic pebbles from Devonian sandstones (Padeha Formation) in the eastern Alborz indicate early Silurian ages (441 \pm 3.1 Ma) with ϵ Hf_(t) values ranging from +8.2 513 to +2.5 (Moghadam and others, 2017). The zircon U-Pb age of granitic pebbles from both 514 Silurian and Devonian successions fit well with the main peak (437 Ma) of the late Ordovician 515 to Silurian detrital zircon population of the Kopet Dagh sandstones (fig. 9). Furthermore, 516 Ordovician to Silurian detrital zircons with a main peak at 435 Ma and ϵ Hf_(t) values of -3.6 to 517

+6.3 from late Ordovician (Qelli Formation) sandstones from eastern Alborz were reported by
Moghadam and others (2017). Taking into consideration all above-mentioned criteria as well
as a short transport distance between eastern Alborz and Kopet Dagh, Ordovician to Silurian
detritus of the Kopet Dagh Basin was most probably supplied through recycling of Ordovician
to Devonian sandstones from the eastern Alborz.

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Devonian to Carboniferous Zircons

The forth age population (385-305 Ma) with a main peak at 340 Ma (fig. 9) exhibits dominantly 525 euhedral zircon grains (~75%) with magmatic internal texture and high Th/U ratios (> 0.5; fig. 526 6 and table S4). This age spectrum comprises $\sim 17\%$ of the total analysed detrital zircons (table 527 S4). Devonian to Carboniferous zircons display general ε -Hf_(t) values between -1.4 and +15.3 528 and three grains between -12.1 and -17.7 (fig. 11 and table S6). Dominant positive ε -Hf_(t) values 529 suggest magmatic zircons with a depleted mantle signature as observed in mafic-ultramafic 530 oceanic crustal and ophiolitic rocks (Patchett, 1983; Blichert-Toft and Albarède, 1997). 531 Carboniferous detrital zircons with a main peak at 344-347 Ma from Cretaceous turbiditic 532 sandstones of Azerbaijan (NW Iran) were reported by Mohammadi and others (2020a). 533 However, a further potential source area includes the northeast of Iran, where Devonian to 534 Carboniferous magmatic rocks were reported from the Aghdarband (Darrehanjir) ophiolite 535 complex (Moghadam and others, 2015a; Zanchetta and others, 2013). The Aghdarband window 536 in the southeast of the Kopet Dagh Basin contains Permo-Triassic undifferentiated sedimentary, 537 538 low-grade metamorphic, and igneous rocks. Igneous rocks include diabase, gabbro, ultramafic, and volcanoclastic sediments (figs. 2 and 3; Afshar-Harb 1982a; Behrouzi and others, 1993). 539 540 In the southern part of the Aghdarband complex, an upper Permian to Lower Triassic polymictic conglomerate (Qara-Gheitan Formation; Behrouzi and others, 1993) overthrusts the Triassic 541 unit. The composition and age of the Aghdarband basement is generally poorly constrained. 542 However, the exposed Devonian to Carboniferous meta-sediments in the north of the 543

Aghdarband complex (Behrouzi and others, 1993; Zanchetta and others, 2013) indirectly 544 indicates the basement age of this complex. Besides this, zircon U-Pb ages of plagio-granitic 545 dikes and ferro-diorites (382-380 Ma) from the Darrehanjir ophiolites also indicate a Late 546 Devonian age for this basement complex (Moghadam and others, 2015a). Furthermore, the 547 zircon U-Pb ages of granitoid cobbles in the Qara-Gheitan conglomerate indicate a late 548 Carboniferous (314 Ma) age (Zanchetta and others, 2013). The wide range of Devonian to 549 Carboniferous rocks in the Aghdarband complex is in concordance with detrital zircon ages in 550 the Kopet Dagh sandstones (fig. 9). Supportively, positive zircon $\varepsilon Hf_{(t)}$ values (+13.8 and 551 +14.9) of dated plagio-granites and ferro-diorites suggest a depleted mantel signature 552 553 (Moghadam and others, 2015a). Devonian and Carboniferous granites and granitioids were 554 reported from the Karakum-Mangyshlak and Karabogaz micro-blocks in Central Asia (for example Natalin and Sengör, 2005). Widespread late Paleozoic to early Mesozoic volcanism in 555 the Turan Block reported by Natalin and Sengör (2005) could represent another possible source 556 for early Carboniferous detrital zircons from the Kopet Dagh. However, the calc-alkaline 557 affinity of these rocks indicates a magmatic arc setting, which is not in agreement with the Hf 558 isotopic composition of detrital zircons. In addition, the reported general SE-NW paleocurrent 559 direction in the Kopet Dagh sandstones (Poursoltani and others, 2007) strengthen the case that 560 561 the Devonian to Carboniferous detritus was supplied from the Aghdarband Complex rather than from the Turan Block. 562

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Permian to Triassic Zircons

The fifth zircon age spectrum in the Kopet Dagh sandstones (300-200 Ma) with a peak at 233 Ma (fig. 9) covers ~25.4% of the total dated zircons (table S4). Zircon grains of this population exhibit euhedral shapes (>80%) with magmatic zoning (fig. 6). Furthermore, over 90% of zircons in this spectrum indicate a high Th/U ratio (>0.55; table S4), which denotes magmatic origin. ϵ -Hf_(t) values of Permo-Triassic zircons range between -5.9 and +10.3 (>80%) with two

grains with values of -15 and -15.2 (fig. 11 and table S6). Similar to Devonian to Carboniferous 570 571 zircons, dominant positive ɛ-Hf_(t) values of the Permo-Triassic zircons propose a depleted mantle signature related to mafic-ultramafic rocks from oceanic crust and ophiolite complexes 572 (Patchett, 1983; Blichert-Toft and Albarède, 1997). Both Permian and Triassic ages were 573 reported from the Fariman and Mashhad complexes in the south of the Kopet Dagh Basin (for 574 example Chiu and others, 2017; Ghazi and others, 2001; Karimpour and others, 2010; Kozur 575 576 and Mostler, 1991; Mirnejad and others, 2013; Moghadam and others, 2015a; Topuz and others, 2018; Zanchetta and others, 2013). 577

The Fariman complex consists of two units: i) The lower metamorphosed unit composed of 578 579 green schists, metabasites, mafic-ultramafic rocks, and serpentinites, and ii) the upper volcanic-580 sedimentary unit. Conodonts and radiolarian fossils reveal a late early Permian to middle Permian age for the Fariman complex (Zanchetta and others, 2013). In addition, ⁴⁰Ar-³⁹Ar 581 582 dating of magmatic hornblende from olivine cumulates yielded early Permian (276 ±4 Ma) ages for the Fariman complex (Topuz and others, 2018). Furthermore, and esitic dykes dated at 211 583 Ma indicate Late Triassic magmatic activity in the Fariman complex (Zanchetta and others, 584 2013). The Fariman complex has been interpreted as a continental magmatic arc with an intra-585 arc basin (Zanchetta and others, 2013), a subduction- and plume-related Paleo-Tethyan 586 587 ophiolite (Moghadam and others, 2015a), as well as a fragment of an oceanic plateau (Topuz and others, 2018). 588

The Mashhad Complex in the south of the Kopet Dagh Basin represents a remnant 589 of the Paleo-Tethys oceanic crust and includes three members: i) The lower member 590 ultramafics. gabbro-diorites, pillow 591 with lavas. marbles, and meta-sediments (Moghadam and others, 2015a). An early Permian age was suggest for the Mashhad 592 ophiolite based on the study of pelagic conodonts (Kozur and Mostler, 1991) and 593 ⁴⁰Ar-³⁹Ar ages of hornblende from gabbros (287.6 Ma and 281.7; Ghazi and others, 594 consists of Permo-Triassic deep-water 595 2001). ii) The middle member clastic

turbidites and metamorphosed sedimentary rocks (accretionary prism). iii) The 596 intermediate pyroclastic rocks 597 upper member exhibits arc-related acidic to 2015a). Post-collisional intrusive bodies crosscut (Moghadam and others, 598 the Mashhad complex that contain several granitoid and monzogranite suites with zircon 599 U-Pb ages between 200 to 217 Ma (Karimpour and others, 2010; Mirnejad and 600 others, 2013). In addition, the Mashhad granodiorite and Mashhad diorite were dated 601 602 to 203 Ma and 219 Ma, respectively, with ε -Hf_(t) values between -3 to +3 (Chiu and others, 2017). The complex is unconformably is covered by Jurassic terrigenous 603 sediments of the Kashafrud Formation. The southeastward continuation of the 604 Mashhad Complex, which is known as the Torbat-e-Jam Complex, was also 605 intruded by post-collisional I-type granitites (217 Ma; Zanchetta and others, 2013) 606 and gabbro-quartz diorite (215 Ma; Ghavi and others, 2018) of similar ages. 607 608 According to Zanchetta and others (2013), the age range of post-collisional intrusions indicates that the obduction of the Mashhad ophiolites occurred before the 609 610 Late Triassic (Norian-Rhaetian).

The age spectrum and dominant positive ε -Hf_(t) values of Permian to Triassic 611 zircons, together with the presence of chromium spinel in the heavy mineral 612 assemblage, indicates an exposure of mafic-ultramafic rocks in the source area. This 613 interpretation is in agreement with unroofing and consequent deposition of the 614 Mashhad and Fariman Complexes, and a remnant of Triassic continental arc in the 615 Kopet Dagh Basin. Northward directed paleocurrents and the presence of Norian 616 granodiorite pebbles (211 Ma) in the conglomerate underlying Jurassic sandstones 617 indicate erosion and unroofing of post-collisional intrusive bodies and ophiolitic 618 host rocks in Middle Jurassic time (for example Zanchetta and others, 2013; Zanchi 619 and others, 2009; Zanchi and others, 2016). 620

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Early Cretaceous Zircons

623 The sixth and youngest age population (146-119 Ma) with a main peak at 132 Ma (fig. 9) contains only 1.5% of the total dated zircon grains (table S4). Zircons grains of this population 624 are euhedral in shape, exhibit magmatic zoning, and show Th/U ratios >0.3 (fig. 6 and table 625 S4). ε -Hf_(t) values (+7.6) of this population indicate non-depleted mantle signatures (fig. 11 and 626 table S6). Occurrence of Early Cretaceous zircons in Lower Cretaceous sandstones (samples 627 628 KD-09 and KD-18) and the Paleocene sandstone (sample KD-14; figs. 8 and 10) indicate synsedimentary magmatic activity in the source area. Furthermore, the presence of detrital zircons 629 with early Aptian ages (~124 Ma; average of 8 youngest ages) in Lower Cretaceous sandstones 630 631 of the Shurijeh Formation indicate maximum depositional ages younger than the previously 632 reported Neocomian stratigraphic age (for example Afshar-Harb 1982a, 1982b; Afshar-Harb and others, 1986; Behrouzi and others, 1993; Mortazavi and others, 2014; Moussavi-Harami 633 634 and Brenner, 1993). The potential source area of Early Cretaceous zircons could be the Cheste Sharif pluton in north Afghanistan. The Cheste Sharif pluton is located in the Band-e-Bayan 635 Zone (a narrow W-E trending crustal block bounded by the large-scale Hari Rod fault) and is 636 dated at 138 Ma by K-Ar biotite (Debon and others 1987). The presence of Early Cretaceous 637 zircons in Lower Cretaceous sandstones in the eastern Kopt Dagh (fig. 2) is in agreement with 638 639 a detritus supply from the Band-e-Bayan Zone during Early Cretaceous time. Most probably, Early Cretaceous zircons in the Paleocene sandstones were recycled from Lower Cretaceous 640 sandstones (fig. 10). 641

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REGIONAL TECTONIC IMPLICATIONS

Integrated provenance analysis including zircon ages and isotopic compositions of the
Mesozoic to Paleocene detrital material in the Kopet Dagh Basin recorded three main tectonic
events in the Phanerozoic (figs. 9 and 11). These events took place in late Ordovician to early
Silurian (main peak at 437 Ma), Late Devonian to early Carboniferous (main peak at 340 Ma),

and late Permo-Triassic (main peak at 233 Ma). The first event (late Ordovician to early
Silurian) corresponds to the separation of Eurasia from Gondwana and the consequent opening
of the Paleo-Tethys Ocean (for example Stampfli and Borel, 2002; Stampfli and others, 2013)
(fig. 12; 440 Ma). This rifting event is marked by mafic magmatism along the northern margin
of Gondwana (Alborz Mountains; Derakhshi and Ghasemi, 2015). Hf-isotopic composition of
Kopet Dagh Ordovician-Early Silurian detrital zircons is in agreement with magmatism in a
rifting setting.

The second recorded event in late Devonian to early Carboniferous coincides with a plutonic 655 episode with A-type (intra-continental) magmatism in NW Iran (Azerbaijan area) and the 656 657 Sanandaj-Sirjan Zone (for example Mohammadi and others, 2020b). In addition, late Devonian 658 to early Carboniferous ages are represented by a series of mafic and ultramafic rocks (ophiolites and oceanic igneous complexes) from Turkey in the west to Tibet to the east (for example 659 660 Moghadam and others, 2015b). This event records the first stages of Paleo-Tethys subduction beneath the Turan Block that initiated in Late Devonian (~380 Ma) and continued until Late 661 Triassic (Moghadam and others, 2015a) (fig. 12; 380 Ma). According to Zanchetta and others 662 (2013), the exposed Devonian to Carboniferous Aghdarband and Permo-Triassic Fariman 663 complexes in NE Iran formed as a back-arc and an intra-arc basin, respectively, along the 664 665 southern margin of the Turan Block, and related volcanoclastic rocks were supplied from an active magmatic arc in the south of the Kopet Dagh Basin (fig. 12; 340 Ma). Contrariwise, 666 Moghadam and others (2015a) argued that the Aghdarband and Mashhad ophiolites in NE Iran 667 668 were part of the Paleo-Tethys oceanic crust, where related volcanoclastic rocks were supplied from an active magmatic arc (Silk Road Arc) in the north of the Kopet Dagh Basin. New zircon 669 670 data presented in this study with depleted mantle composition and a main peak at 340 Ma support the hypothesis of Zanchetta and others (2013) and point toward intensified rifting and 671 spreading of a back-arc basin ~40 Myr after the initiation of the Paleo-Tethys subduction (fig. 672

673 12; 380 Ma).

The third event (late Permian to late Triassic) coincides with a continental magmatic arc and 674 675 the final collision of the Paleo-Tethys (fig. 12; 230 Ma). Rifting of the Iran block from Gondwana in Late Paleozoic forced to northward subduction of the Paleo-Tethys Ocean 676 beneath the Turan (Eurasia) plate, which resulted in the formation of a Permo-Triassic 677 continental magmatic arc. The Mashhad Ophiolite Complex was abducted in early Late Triassic 678 (Carnian-Norian; Moghadam and others, 2015b). Finally, collision between the Iran and Turan 679 680 plates occurred in Late Triassic (Norian-Rhaetian) and was accompanied by the intrusion of several post collisional plutonic bodies into the magmatic arc and the Mashhad Ophiolite 681 Complex (fig. 12; 230 Ma). Finally, unroofing of the magmatic arc, the Mashhad Ophiolite 682 683 Complex, and related post-collisional intrusions in Middle Jurassic led to deposition of Middle Jurassic turbiditic sandstones in the Kopet Dagh extensional basin (fig. 12; 165 Ma). 684

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CONCLUSIONS

The present study assesses the provenance of detrital sediments in Triassic to Paleocene 687 turbiditic, fluvial-deltaic and continental sandstones of the Kopet Dagh Basin in northeast Iran. 688 Over 2470 detrital zircons U-Pb ages yield a spectrum of variable protoliths from Archean to 689 Early Cretaceous (2966 Ma - 119 Ma) with main peaks in Paleoproterozoic (1883 Ma), 690 691 Neoproterozoic (643 Ma), early Silurian (437 Ma), early Carboniferous (340 Ma), Late Triassic 692 (233 Ma), and a minor peak in Early Cretaceous (132 Ma). The combined U-Pb ages and Hf 693 isotope data indicate that protolith rocks belonged to an early Silurian intracontinental rift 694 (opening of Paleo-Tethys), an early Carboniferous rifting of a back-arc (Aghdarband Basin), and a Late Triassic collisional to post-collisional (Paleo-Tethys collision) magmatism. Our 695 results highlight the timing of Aghdarband Basin opening 40 Ma after subduction initiation of 696 697 the Paleo-Tethys. Cr-spinel and heavy mineral assemblages imply additional detritus sourcing from ophiolites (Mashhad and Fariman complexes). Our analytical results furthermore suggest 698 that the majority of Jurassic sedimentary detritus sourced from protoliths in the south of the 699

Kopet Dagh Basin and deposited in a submarine fan-delta complex. Early Cretaceous detritus
was supplied from similar protoliths and furthermore from recycled Jurassic turbiditic
sandstones. These sediments were deposited in a fluvial-deltaic complex. The Paleocene
continental sediments were mainly supplied from recycling of exposed Jurassic to Early
Cretaceous sediments.

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- 708

709 Supplementary Table Captions

Table S1: Geographical location (degree, minute, second), lithology, local formation name, sedimentary
environment, age and characteristics of the analyzed samples of eastern Kopet Dagh sandstones.
Analysis key: MFA: modal framework analysis, HM: heavy minerals, DZA: detrital zircon age and
Hafnium isotopes.

- 714
- Table S2: Sandstone framework data and calculated percentages for eastern Kopet Dagh sandstones.
- 716

717 Table S3: Heavy mineral data, calculated percentages and heavy mineral indexes of eastern Kopet Dagh718 sandstones.

719

720 Table S4: Zircon U-Pb isotopic data and calculated ages for eastern Kopet Dagh sandstones. C= Coarse

721 grain (>200 μ) and M= Medium grain (100-200 μ).

722

723 Table S5: Zircon U-Pb dating details including sample preparation, analytical conditions, laser ablation

- system, ICP-MS instrument, data from unknowns and reference materials, and data processing.
- 725
- Table S6: Lu-Hf isotopic data and calculated ratios for eastern Kopet Dagh sandstones.
- 727 Table S7: Zircon Lu-Hf isotopic analyses details including sample preparation, analytical conditions,
- 728 laser ablation system, ICP-MS instrument, data from unknowns and reference materials, and data
- 729 processing.

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1092	Fig. 2. Simplified geologic map of the East Kopet Dagh Basin and samples location.
1093	Stratigraphic ages correspond to the 1:250,000 geological maps of Sarakhs (Afshar-Harb,
1094	1982a), Darreh Gaz (Afshar-Harb, 1982b), Mashhad (Afshar-Harb and others, 1986), and
1095	Torbat-e-Jam (Behrouzi and others, 1993).
1096	
1097	



Fig. 3. Composite stratigraphic section of the East Kopet Dagh Basin with local formation
names, sampled formations, and comments on lithology and sedimentary environment.
Stratigraphic ages correspond to the 1:250,000 geological maps of Sarakhs (Afshar-Harb,
1982a), Darreh Gaz (Afshar-Harb, 1982b), Mashhad (Afshar-Harb and others, 1986), and
Torbat-e-Jam (Behrouzi and others, 1993). Black stars: sampled sedimentary sequences.





Fig. 4. Detrital composition of Kopet Dagh sandstones in ternary classification schemes and
provenance discrimination diagrams. (A) Q-F-L sandstone classification diagram (Folk, 1980);
(B) Qm-P-K diagram (after Dickinson and Suczek, 1979); (C) Qp-Lvh-Lsm diagram (after



Staurolite

Chloritoid Andalusite

Others<1%



displaying the relative abundance of heavy mineral groups. 1119

1120

KD-06

KD-03



1123 Fig. 6. Representative cathodoluminescence images of dated detrital zircons of the Kopet Dagh

sandstones. White circles show laser spot positions for U-Pb age analyses.



Fig. 7. Detrital zircon ²⁰⁶Pb/²³⁸U age probability density diagrams with corresponding
Concordia plots for the Lower Triassic and Jurassic samples KD-01, KD-03, KD-06, KD-08,
KD-10, and KD-15. Ages with discordance greater than 5% are excluded. Time scale after
Gradstein and others (2012).



Fig. 8. Detrital zircon ²⁰⁶Pb/²³⁸U age population probability density diagrams with corresponding Concordia plots for Early Cretaceous and Paleocene samples KD-02, KD-09, KD-14 and KD-18. Ages with discordance greater than 5% are excluded. Time scale after Gradstein and others (2012).



1142 Fig. 9. Combined zircon U-Pb age distribution pattern of Kopet Dagh sandstone samples.



1148 Fig. 10. Detrital zircon U-Pb mean age populations versus stratigraphic age of the Kopet Dagh

- sandstones. Time scale after Gradstein and others (2012).



Fig. 11. Time-corrected ε-Hf_(t) values versus ²⁰⁶U/²³⁸Pb ages of Kopet Dagh detrital zircons.
Depleted mantle evolution trend (dashed line) from Griffin and others (2000), and Chondritic
Uniform Reservoir values (CHUR) from Bouvier and others (2008).



1159 Fig. 12. Tectonic reconstruction along N-S profiles of the Kopet Dagh Basin through time.