1 **Provenance information recorded by mineral inclusions in**

- 2 detrital garnet
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12 Abstract

Garnet single-grain analysis is an often used and well 13 14 established tool in sedimentary provenance studies, especially 15 when metamorphic source rocks are involved. So far, however, 16 solely the geochemical composition of detrital garnets is considered to draw conclusions concerning probable source 17 18 rocks. The gained information is often limited by (i) 19 geochemical overlap of garnets derived from different lithology 20 and metamorphic grade, (ii) similar probabilities of belonging 21 to more than one source rock type, and (iii) the limitations of 22 discriminating different protolith compositions. Here we present the first attempt of using mineral inclusions in detrital 23

24 garnet as a provenance tool. We analyzed the inclusions of 25 ~300 fine to medium sand-sized detrital garnets from two 26 proximal modern sand samples taken in the HP/UHP Western 27 Gneiss Region of SW Norway. All mineral inclusions $\geq 2 \ \mu m$ 28 were identified by Raman spectroscopy, showing that (i) most 29 garnets from HP/UHP metamorphic source rocks contain 30 mineral inclusions $\geq 2 \mu m$, (ii) Raman spectroscopy is a very 31 powerful tool to characterize the inclusion types, and (iii) less 32 stable mineral phases like kyanite, omphacite, diopside, 33 enstatite, coesite, amphibole group, and epidote group minerals 34 occur as inclusions in garnet. These minerals, which are 35 important for provenance studies, can thus be preserved in the 36 sedimentary record as long as garnet is stable. The combination 37 of inclusion types in garnet and geochemical garnet 38 classification shows that (i) inclusions well reflect the 39 geological characteristics of the sampled catchments, implying 40 that they are useful indicators for HP/UHP provenance, and (ii) 41 inclusions in garnet can be used to support and enhance the 42 provenance information obtained by garnet geochemistry.

43 Keywords: Sediment provenance; Garnet; Mineral inclusions;
44 Raman spectroscopy; Western Gneiss Region

45 **1. Introduction**

46 Provenance studies often deal with changes in the source area47 arising from the temporal geodynamic evolution of the region.

48 In particular, the initiation of high-pressure (HP) and ultrahigh-49 pressure (UHP) metamorphic rocks as a sedimentary source is 50 of specific interest, displaying the exposure of deep crustal 51 levels in the source area, which typically has significant 52 geologic and geodynamic implications. Characteristic HP/UHP 53 metamorphic index minerals like glaucophane, lawsonite, 54 omphacite, kyanite, or coesite are either mechanically and/or 55 geochemically unstable. Therefore. these minerals 56 progressively disappear when subjected to processes of the 57 sedimentary cycle, and thus important provenance information 58 gets lost. Additionally, HP/UHP rocks are often overprinted 59 under lower-grade metamorphic conditions of the amphibolite 60 and granulite facies during exhumation, whereby HP/UHP 61 phases get usually replaced by lower-pressure phases. This 62 leads to the fact that the mineral assemblages typically reflect 63 retrograde stages, whereas the prograde and peak metamorphic 64 history is obscured. Consequently, the transfer of characteristic 65 HP/UHP index minerals into the sedimentary system may be 66 hampered.

In contrast, information about prograde and peak metamorphic stages can be recorded and preserved in garnet, a heavy mineral which can grow during multiple metamorphic stages. Garnet is a very abundant metamorphic mineral covering a broad range of pressure-temperature (P-T) conditions and protolith compositions (e.g., Krippner et al., 2014). Because garnet 73 composition is a function of these parameters, and garnet is 74 comparatively stable during surface weathering, transport, and 75 deep burial conditions (e.g., Morton and Hallsworth, 1999), 76 several geochemical discrimination schemes have been 77 developed and applied as a tool in sedimentary provenance 78 analysis (see Krippner et al., 2014, for a review and evaluation). A recently published discrimination scheme by 79 80 Tolosana-Delgado et al. (2017) operates with a multivariate 81 statistical model to assign garnet grains with a certain 82 probability to major host rock groups. Until now, however, all 83 garnet provenance tools solely consider geochemical and 84 isotopic garnet composition. Although it is well known from 85 crystalline rocks that metamorphic garnet usually contains 86 inclusions and that the occurrence and distribution of these 87 inclusions can be used to deduce the metamorphic history of a 88 rock (e.g., Thompson et al., 1977; Krogh, 1982; Faryad et al., 89 2010), this concept has not yet been thoroughly applied in 90 provenance studies.

91 Here we present the distribution of mineral inclusions in 92 detrital garnets from two proximal modern sand samples taken 93 in the HP/UHP Western Gneiss Region of SW Norway. All 94 mineral inclusions $\geq 2 \ \mu m$ of ~300 garnets (i.e., ~150 per 95 sample) in the grain-size range 63–500 μm were identified by 96 Raman spectroscopy. This technique has the advantages of (i) 97 not being restricted to a specific sample preparation, allowing

98 straightforward combination with other analytical techniques 99 like the electron microprobe, (ii) being contact-free and having 100 an excellent volume resolution due to the usage of a confocal 101 microscope system adapted for the Raman spectrometer, 102 enabling the identification of even small inclusions within the 103 entire garnet volume, (iii) getting structural and implicitly 104 geochemical information in a single step, so that even 105 polymorphs like quartz and coesite can be distinguished, and 106 (iv) being comparatively quick, which is important when 107 analyzing a large number of garnet grains and mineral 108 inclusions (e.g., Nasdala et al., 2004; Andò and Garzanti, 2014; 109 Neuville et al., 2014). We compared the obtained inclusion data 110 with the geochemical composition of the host garnets and show 111 that (i) less stable but for provenance studies important mineral 112 phases occur as inclusions in garnet, suggesting that these can 113 be preserved in the sedimentary record as long as garnet is 114 stable, (ii) inclusions in garnet well reflect the geological 115 characteristics of the sampled catchments, implying that they 116 are useful indicators for HP/UHP provenance, and (iii) 117 inclusions in garnet can be used to support and enhance the 118 provenance information given by geochemical garnet 119 composition.

120 **2. Geological setting**

121 The two studied areas (Flatraket and Runde) are located in the 122 Western Gneiss Region of SW Norway, which extends over an area of ~50,000 km² (Wain, 1997) between Bergen in the South 123 124 and Trondheim in the North (Fig. 1). The Western Gneiss 125 Region is composed of the autochthonous Western Gneiss 126 Complex, overlain by a series of allochthonous Caledonian 127 thrust nappes (Hacker et al., 2010). It is believed that the 128 Western Gneiss Complex represents the westward continuation 129 of the Baltic basement. Thus, the Western Gneiss Region is a 130 large tectonic window through the allochthonous nappe pile, in 131 which the Western Gneiss Complex is exposed as a large 132 segment of the former craton Baltica (Roberts and Gee, 1985; 133 Krabbendam and Wain, 1997; Beyer et al., 2012).

134 The Western Gneiss Complex mainly consists of orthogneisses, 135 which were initially generated during the Gothian Orogeny 136 (~1.7 to 1.5 Ga) and subsequently overprinted during the Sveconorwegian Orogeny (~1.3 to 0.9 Ga) (e.g., Beyer et al., 137 138 2012). The resulting metamorphic conditions ranged between 139 amphibolite and granulite facies (Bingen et al., 2001; Røhr et 140 al., 2004). Relicts of this event are rare and comprise ~1% of the Western Gneiss Complex (Wain et al., 2001; Peterman et 141 142 al., 2009).

143 The Caledonian Orogeny in Paleozoic times is commonly 144 subdivided into the pre-Scandian (not recognized in the 145 Western Gneiss Complex; Hacker et al., 2010), the Scandian, 146 and the post-Scandian metamorphic phases. The single phases 147 can chronologically overlap and coexist in the broad area of the 148 Western Gneiss Region.

149 The Scandian phase encompasses the time interval between 150 early Silurian and early Devonian times (~435 to 400 Ma). During this phase, the two continental blocks Laurentia 151 152 (western block) and Baltica (eastern block) collided under 153 oblique plate convergence and final closure of the Iapetus 154 Ocean (e.g., Roberts and Gee, 1985; Spengler et al., 2009; 155 Hacker et al., 2010). This was accompanied by overthrusting 156 and arrangement of the allochthonous nappes (e.g., Roberts and 157 Gee, 1985; Andersen et al., 1990), forming the ~1000 km-long 158 mountain system called the Scandinavian Caledonides (Beyer 159 et al., 2012). The progressive convergence led to the subduction 160 of Baltica's western continental margin beneath Laurentia (e.g., 161 Torsvik 2005) and Cocks. and coherent HP/UHP 162 metamorphism (e.g., Smith, 1984; Carswell and Cuthbert, 163 2003). The metamorphic grade and depth of subduction 164 increased from SE towards the NW, as well as the intensity of deformation (e.g., Hacker et al., 2010). Temperature conditions 165 166 ranged between ~550°C in the SE (Krogh, 1977; Griffin et al., 1985; Carswell and Cuthbert, 2003) and 850-950°C in the 167

northernmost sector (Kylander-Clark et al., 2008). In the
northernmost part also the highest pressure conditions were
reached with up to 5.5–6.5 GPa (Scambelluri et al., 2008;
Spengler et al., 2009), which were equalized to subduction
depths of 180–200 km (van Roermund, 2009). Although the
rocks were subjected to UHP conditions, they reacted only
partially and locally (Corfu et al., 2014).

175 The post-Scandian metamorphic phase in Devonian times (~400 to 385 Ma) included the decompression and exhumation 176 177 of the Western Gneiss Region to shallow crustal levels (e.g., 178 Hacker et al., 2010). The exhumation was accomplished by 179 coaxial E-W extension, vertical thinning, and minor N-S 180 shortening. This led to a progressive exhumation of the 181 Western Gneiss Region from SE to NW, which is contrary to 182 the direction of the subduction during the Scandian phase 183 (Krabbendam and Wain, 1997; Hacker et al., 2010; Spencer et 184 al., 2013). Exhumation was accompanied by an extensive 185 retrograde amphibolite to granulite facies metamorphic 186 deformation and recrystallization (e.g., overprint with 187 Krabbendam and Wain, 1997; Walsh and Hacker, 2004). This extensive and pervasive metamorphic overprint eliminated 188 189 almost all records of HP/UHP metamorphism (e.g., Root et al., 190 2005). Solely dispersed eclogite bodies and eclogite facies 191 rocks persisted (composing only ~1vol.% of the HP/UHP 192 terrane) and record nearly all information about the HP/UHP metamorphism in the Western Gneiss Region (e.g., Wain et al.,2000).

195 The post-Scandian metamorphic phase was followed by a 196 progressive exhumation of the UHP domains into the mid-197 upper crust by late folding, which continued through the late 198 Devonian up to ~374 Ma (Hacker et al., 2010; Walsh et al., 199 2013). Today, three discrete UHP domains crop out within the 200 Western Gneiss Region, which probably represent the cores of 201 east-plunging antiforms. The distinct areas with UHP eclogites 202 are separated by areas with HP eclogites (e.g., Root et al., 203 2005).

204 **3. Sampling areas**

The sampling area at Flatraket is located ~6 km north of the Nordfjord and ~17 km WSW of Åheim. The island of Runde is located ~55 km NW from Flatraket (Fig. 1). Both sampling areas are situated at the margins of an UHP domain, Flatraket at the western margin of the Nordfjord-Stadlandet UHP domain, and Runde at the western margin of the Sorøyana UHP domain (Root et al., 2005).

The stream-sediment sample at Flatraket (AK-N13-1: 61°58.554'N, 5°13.845'E) was taken a few meters upstream to the mouth of a northward draining stream so that mixing with sediments from coastal currents can be excluded (Krippner et al., 2016). In contrast, the sediment sample at Runde (AK-N37: 217 62°23.341'N, 5°38.252'E) represents a beach-sediment sample. 218 However, because the sample was taken directly at the mouth 219 of a small modern stream, and any morphological features 220 indicating influences of coastal currents are absent, mixing with 221 material transported along the shore can also be neglected for 2.2.2 the sample from Runde (Schönig et al., 2018). Thus, the 223 material in the two analyzed samples exclusively derived from 224 the catchments marked by the watersheds in Fig. 1.

225 It is important to note that older sedimentary deposits could 226 also be present in the catchments, which could scale up the area 227 where the initial source rocks of the analyzed garnets might 228 have been located (Schönig et al., 2018). In particular, glacial 229 deposits in the Western Gneiss Region should be considered, 230 which mainly originate from the Late Weichselian glaciation 231 (Rye et al., 1987; Mangerud, 2004; Hughes et al., 2016). In the 232 studied catchments, glacial deposits are thin but present 233 (Goksøyr, 1938; Undås, 1942; Thoresen, 2013). However, even 234 if the thin glacial deposits in the catchments could have 235 provided small amounts of garnets to the sampled sediments, 236 the ice flow direction coming exclusively from inland (e.g., 237 Mangerud, 2004) only permit local sources within the Western 238 Gneiss Region for the glacial deposits. Thus, the general 239 HP/UHP signal from the Western Gneiss Region is not biased.

240 **3.1 Flatraket**

241 Within the catchment of the stream-sediment sample AK-N13-242 1, layered micaceous quartzo-feldspathic gneiss constitutes the 243 major portion (Fig. 1). This gneiss was pervasively deformed 244 during the post-Scandian phase of the Caledonian Orogeny, 245 recrystallized under amphibolite facies conditions, and locally 246 contains garnet (Krabbendam and Wain, 1997; Krabbendam et 247 al., 2000). Eclogite pods <50 m in size are embedded in the 248 micaceous gneiss. The cores of the pods are least affected by 249 the amphibolite facies retrogression, and mafic bodies >10 m 250 partially exhibit fresh eclogite within the core. The amphibolite 251 facies overprint increases towards the pod margins and the 252 outermost parts consist of amphibolite (Krabbendam et al., 253 2000). UHP metamorphism in this region is recorded by two 254 known UHP eclogites, which contain bimineralic quartz/coesite 255 inclusions in garnet and omphacite (Smith, 1984, 1985; Wain, 256 1997; Root et al., 2004; Smith and Godard, 2013; Schönig et 257 al., 2018). However, both do not contribute material to the 258 sediment sample AK-N13-1 because erosional material from 259 the Flatraket harbor UHP eclogite (AK-N12) will be directly 260 fed into the sea, and the UHP eclogite at Straumen is located 261 outside the catchment in western direction (Fig. 1).

262 Besides the micaceous quartzo-feldspathic gneiss, the Flatraket 263 complex is the next largest unit in the catchment, which 264 consists of megacrystic felsic gneiss with a quartz-monzonitic 265 composition and layered garnetiferous felsic gneiss, both 266 showing also mafic parts (Krabbendam et al., 2000; Wain et al., 2001). In general, the complex represents a $\sim 2 \text{ km}^2$ granulite 267 268 facies low-strain enclave within the pervasively deformed and 269 recrystallized amphibolite facies gneisses of the Western 270 Gneiss Region (Wain, 1997; Wain et al., 2001). U-Pb ages of 271 zircon and monazite indicate magmatic crystallization of the 272 complex during the Gothian Orogeny at ~1680 to 1640 Ma and 273 constrain timing of the granulite facies overprint to the 274 Sveconorwegian Orogeny at ~1100 Ma (Corfu et al., 2014). 275 During the Scandian phase of the Caledonian Orogeny, the 276 majority of the complex remained dry and undeformed, 277 whereby the Proterozoic granulite facies mineral assemblages 278 preserved metastable. Eclogite facies rocks are restricted to 279 zones of fluid infiltration and/or deformation and constitute ~5% of the complex (Wain et al., 2001; Corfu et al., 2014). 280 281 During the subsequent post-Scandian metamorphic phase, the 282 fluid infiltration was more extensive and leads to a pervasive 283 amphibolite facies metamorphism on the retrograde part of the 284 Caledonian orogenic cycle. More than 50% of the Flatraket 285 Complex was affected by amphibolite facies hydration in 286 different degrees, but granulite facies relicts are still present 287 (Krabbendam et al., 2000; Wain et al., 2001). Several dykes and pods occur within the complex, including mafic to 288 289 intermediate rocks (granulites, eclogites, locally retrogressed to

amphibolite), dioritic to gabbroic rocks (primary granulite
facies, strong amphibolite facies overprint, sometimes eclogitic
assemblages at the rims, locally relictic magmatic texture in the
core), and meta-anorthosites (mainly granulite facies).

294 Table 1 summarizes the mineralogical composition of the 295 metamorphic rocks occurring in the sampled catchment of the 296 stream-sediment sample AK-N13-1 and the close vicinity as 297 outlined above. The heavy mineral spectra of the sample in the 298 63–125 µm grain-size fraction consists mainly of epidote group 299 minerals and pyroxenes, followed by garnet, and much smaller 300 amounts of amphibole group minerals, apatite, tourmaline, 301 kyanite, olivine, rutile, and titanite (Krippner et al., 2016). In 302 Fig. 1, also pebble samples (eclogites AK-N13-2a and AK-303 N13-2b and felsic gneisses AK-N13-2c and AK-N13-2d: 304 61°58.554'N, 5°13.845'E) and a crystalline rock sample (UHP 305 eclogite AK-N12: 61°58.710'N, 5°14.063'E) from Krippner et 306 al. (2016) were marked, which were used for geochemical 307 comparison with the analyzed detrital garnets in this study.

308 3.2 Island of Runde

Within the catchment of the beach-sediment sample AK-N37, the largest unit is a micaceous quartzo-feldspathic orthogneiss (Fig. 1), which is also the main rock type of the whole region (northern Sorøyane). Its similarity to the common orthogneisses of the Western Gneiss Complex suggests an affiliation to the basement of Baltica (Root et al., 2005). Because of their similarity, the micaceous quartzo-feldspathic gneisses at
Flatraket and Runde are not differentiated in Fig. 1, but note
that the region around Runde experienced higher peak
metamorphic temperatures (e.g., Kylander-Clark et al., 2008).

The next smallest rock unit on the island is a mylonitic quartzofeldspathic gneiss, whereby only small divisions crop out at the outermost part of the sampled catchment. The mylonitic gneiss is locally garnetiferous (Krippner et al., 2016), contains amphibole (hornblende), and is of amphibolite facies. U–Pb ages in titanite ~384 Ma underline the pervasive retrogression during the post-Scandian phase (Spencer et al., 2013).

326 In the gneisses, numerous eclogite pods occur on the island of 327 Runde, which – with a few exceptions – have not been mapped 328 so far (Dahl, 1954; Root et al., 2004, 2005; Spencer et al., 329 2013; Krippner et al., 2016). Therefore, it can be assumed that 330 various eclogite pods occur in the sampled catchment, although 331 these are not shown in Fig. 1. For the shown eclogites, it is 332 known that the eclogite at Langenes (AK-N38), ~1 km NE of 333 the sampled catchment, contains garnet, sodic diopside (13% 334 jadeite), magnesiohornblende, and rutile (Root et al., 2004). 335 This eclogite is indicated as HP eclogite because no 336 mineralogical evidence for UHP metamorphism is found so far, 337 which might be related to the absence of silica. An adjacent 338 silica-bearing eclogite a few hundred meters in the southern 339 direction, in contrast, contains bimineralic coesite/quartz

340 inclusions, displaying that the area underwent UHP conditions 341 (Root et al., 2005). UHP metamorphism is also indicated by 342 intact, monomineralic coesite inclusions found in detrital 343 garnets of the same sample as studied here (AK-N37). Due to 344 the geochemical variation of the detrital coesite-bearing host 345 garnets and the variation in mineral inclusion assemblages, not 346 only eclogites but also felsic rocks are considered as UHP 347 source rocks (Schönig et al., 2018).

348 The heavy mineral spectra of the sample AK-N37 in the 63-349 125 µm grain-size fraction is dominated by pyroxenes and 350 garnet, followed by significant amounts of amphibole group minerals, epidote group minerals, and apatite. Tourmaline, 351 352 zircon, kyanite, olivine, rutile, and titanite occur in much 353 smaller proportions (Krippner et al., 2016). In Fig. 1 also 354 crystalline rock samples (eclogite AK-N38: 62°24.212'N, 355 5°39.230'E; and garnetiferous felsic gneiss AK-N39-1: 62°24.012'N, 5°39.459'E) from Krippner et al. (2016) were 356 357 marked, which were used for geochemical comparison with the 358 analyzed detrital garnets in this study.

359 4. Methods

360 **4.1 Mineral separation and sample preparation**

361 Mineral separation and sample preparation were performed at 362 the University of Göttingen (Geosciences Center, Department 363 of Sedimentology and Environmental Geology). About 300 g

364	of both modern sand samples from Flatraket (AK-N13-1) and
365	Runde (AK-N37) were wet-sieved to separate grain-size
366	fractions, treated with acetic acid, split by quartering into
367	amounts of 15-20 g, and the heavy mineral fraction was
368	separated using sodium polytungstate with a density of 2.89 g
369	cm^{-3} . Garnets were handpicked under the binocular microscope
370	from three grain-size fractions (63–125 $\mu m,$ 125–250 $\mu m,$ 250–
371	500 μ m) and embedded in synthetic mounts using a bonding
372	epoxy composed of a mixture of Araldite® resin and hardener
373	at a ratio of 5:1. Mounts with the picked garnet crystals were
374	ground with silicon carbide abrasive paper and polished in two
375	steps with 3 μm and 1 μm Al_2O_3 abrasives in suspension.
376	Overall ~350 grains per grain-size fraction and sample were
377	picked (i.e., ~1050 per sample) and numbered in ascending
378	order. For this study, the first 50 grains from each grain-size
379	fraction were selected for inclusion analysis by Raman
380	spectroscopy and geochemical analysis by the electron
381	microprobe. This means grain numbers 1–50 (63–125 μ m),
382	351–400 (125–250 $\mu m),$ and 701–750 (250–500 $\mu m)$ from
383	sample AK-N13-1, and grain numbers 1–50 (63–125 $\mu m),$
384	351–400 (125–250 $\mu m),$ and 706–755 (250–500 $\mu m)$ from
385	sample AK-N37.

386 4.2 Raman spectroscopy

387 Raman spectroscopy was performed at the University of388 Göttingen (Geosciences Center, Department of Sedimentology

389 and Environmental Geology) using a Horiba Jobin Yvon 390 XploRA Plus spectrometer equipped with an Olympus BX41 391 microscope, a 532 nm diode laser (25 mW maximum output 392 power), and a motorized x-y-z stage. The confocal microscope 393 is coupled to a 200 mm focal length spectrograph equipped with a four-grating turret $(2400 \ 1 \ mm^{-1}, 1800 \ 1 \ mm^{-1}, 1200 \ 1$ 394 mm^{-1} , and 600 l mm^{-1}). All measurements were performed 395 396 with maximum laser power, a confocal hole diameter and slit set to 100 μ m, the 1800 1 mm⁻¹ grating, and a 100× objective 397 398 with a numerical aperture of 0.9, where ~50% of the signal 399 derive from an excitation volume depth of ~1.8 µm and ~95% 400 derive from a volume depth of ~7.0 µm. For mineral 401 identification of all inclusions $\geq 2 \mu m$ within the garnets, the spectrometer was calibrated on the 520.7 cm⁻¹ line of Si, and 402 the recorded spectrum was centered at 1000 cm⁻¹, covering a 403 spectral field between $\sim 100 \text{ cm}^{-1}$ and $\sim 1800 \text{ cm}^{-1}$. 404

405 Raman spectra of inclusions, in particular if they are small, are 406 often masked by a strong signal from the garnet hosts. 407 Therefore, most of the time the spectra of the garnet hosts were 408 also captured directly next to the mineral inclusions to be 409 identified, and subsequently the host spectra were subtracted 410 from the inclusion/host mixed spectra, isolating the inclusion 411 spectra (Fig. 2). In this way, sometimes inclusions $<2 \mu m$ were 412 also identified if they had a good Raman response (e.g., rutile), 413 but only an identification of all inclusions $>2 \mu m$ can be 414 guaranteed. Captured spectra were exported to the software 415 CrystalSleuth (Laetsch and Downs, 2006), the background was 416 automatically subtracted via the software, and the 417 corresponding mineral was identified by comparison with the 418 RRUFF database (Lafuente et al., 2015) also via the 419 CrystalSleuth software. For the obtained inclusion spectra in 420 this study, mineral identification was made by comparison with 421 the database and classification into inclusion types. Overall, all 422 inclusions $\geq 2 \,\mu m$ in ~50 handpicked garnets from all three 423 analyzed grain-size fractions were identified by Raman 424 spectroscopy, i.e., ~150 garnets per sample and a total of ~300 garnets. For this, ~120 hours were spent in the Raman 425 426 laboratory, which equates to ~ 2.5 garnets per hour.

427 **4.3 Electron microprobe**

428 Electron microprobe measurements were performed at the 429 University of Göttingen (Geosciences Center, Department of 430 Geochemistry) using a JEOL JXA 8900 RL electron 431 microprobe equipped with five wavelength dispersive 432 spectrometers. Before analysis, all samples were coated with 433 carbon to ensure conductivity. Measurement conditions include 434 an accelerating voltage of 15 kV and a beam current of 20 nA. 435 Counting times were 15 s for Si, Mg, Ca, Fe, and Al, and 30 s 436 for Ti, Cr, and Mn. The compositions of the ~50 detrital garnet 437 grains from each of the three grain-size fractions (63–125 µm; 438 125–250 µm; 250–500 µm) were determined. Preferentially,

the centers of the detrital grains were analyzed. Only when
inclusions or fractures are located in the center, the
measurement spot was shifted towards the rim.

442 **5. Results**

443 **5.1 Mineral inclusions**

444 5.1.1 Classification of inclusion types

445 By comparing the collected inclusion spectra with the database, 446 a large number of mineral inclusions were identified. This 447 includes oxides (coesite, corundum, quartz, rutile), feldspar 448 tectosilicates (albite, anorthoclase, labradorite. group 449 microcline, oligoclase, orthoclase, sanidine), mica group 450 phyllosilicates (muscovite, phlogopite), pyroxene group 451 (diopside, omphacite) and enstatite, amphibole group 452 (actinolite, gedrite, tremolite) inosilicates, epidote group 453 sorosilicates (clinozoisite, epidote, zoisite), nesosilicates 454 (kyanite, zircon), the phosphate apatite, carbonate minerals 455 (calcite, dolomite, magnesite, siderite), sulfate minerals 456 (anhydrite, celestine, gypsum), and opaque minerals. The 457 identification of specific mineral phases by simply comparing 458 their Raman spectra with the database is sometimes unreliable, 459 especially within individual mineral groups. However, the 460 approach of using mineral inclusions identified by Raman 461 spectroscopy as a metamorphic source rock indicator, as 462 introduced here, should be straightforward without detailed

analysis of every single spectrum. Therefore, the identification
of the detected mineral phases by the RRUFF database was
evaluated by validating specific spectral patterns. This results
in a grouping of some phases into inclusion types to ensure
their correct assignment and to focus on the most important
information.

469 Due to their unique spectral Raman pattern, all detected oxides 470 (coesite, corundum, quartz, rutile), nesosilicates (kyanite, 471 zircon), and the phosphate apatite can be clearly identified and 472 differentiated from other mineral phases (Fig. 3a-e), even if 473 they are chemically similar and only differ in crystal structure 474 like coesite and quartz (Fig. 3a). Therefore, all of them are 475 indicated as single inclusion types, except corundum. 476 Corundum was only found as small slices on the polished garnet surfaces of one garnet (<1%) from AK-N13-1 477 478 (Flatraket), and five garnets (~3%) from AK-N37 (Runde). 479 Because Al₂O₃ (corundum) abrasives are used for polishing, 480 these slices are considered to be contaminants and were thus 481 excluded from further analysis.

Based on the Raman spectral pattern, feldspars can be readily identified by the presence of two or three peaks in the 450–515 cm^{-1} region, with the strongest peak between 505–515 cm^{-1} (Freeman et al., 2008). Several different members of the feldspar group tectosilicates were identified from the database as inclusions in the analyzed garnets. However, often similar 488 high match probabilities are given for more than one member, 489 in particular for the series orthoclase-sanidine-microcline and 490 labrodorite-oligoclase-albite-anorthoclase, respectively. In this 491 study, inclusions identified as orthoclase-sanidine-microcline 492 were assigned to the alkali feldspar inclusion type, and 493 inclusions identified as labrodorite-oligoclase-albite-494 anorthoclase to the plagioclase inclusion type. Alkali feldspar 495 and plagioclase can be distinguished by the position of the 496 strongest peak. For alkali feldspars this is located between 513-515 cm⁻¹, whereas all plagioclase main bands are located at 497 <510 cm⁻¹ (Mernagh, 1991; Freeman et al., 2008; Bersani et 498 499 al., 2018). Because the tested inclusions which were assigned to 500 the alkali feldspar type show the main band position of ≥ 514 cm^{-1} and those assigned to the plagioclase type $\leq 511 cm^{-1}$ (Fig. 501 502 3f), the classification of these two feldspar types based on the 503 database seems reliable.

504 Phyllosilicates are easily distinguishable from the other major 505 structural types of silicates. Within the group, however, the 506 discrimination of specific types is more complicated, and the H₂O/OH spectral region (3500-3800 cm⁻¹) should be also 507 508 considered (Wang et al., 2015). The phyllosilicate inclusion 509 types identified by the database comprise the mica group 510 minerals muscovite (also high match probabilities for 511 trilithionite and paragonite) and phlogopite (also high match 512 probabilities for fluorophlogopite). Collected spectra of both

show a broad pattern in the $3500-3800 \text{ cm}^{-1}$ region (Fig. 4), 513 514 indicating the presence of H₂O, and thus, a complicated 515 stacking sequence of tetrahedral and octahedral layers so that 516 kaolinite-serpentine and pyrophyllite-talc group phyllosilicates 517 can be excluded. The combined spectral pattern of the $<1200 \text{ cm}^{-1}$ and the 3500–3800 cm⁻¹ region, in turn, makes an 518 assignment to the mica group reliable. Furthermore, the 519 strongest peak in the 600–800 cm⁻¹ region of inclusions 520 identified as muscovite by the database is located at $>700 \text{ cm}^{-1}$, 521 522 verifying an affiliation to the dioctahedral group of 523 phyllosilicates like muscovite-paragonite. This is further supported by the presence of a peak at $\sim 430 \text{ cm}^{-1}$, which is 524 typical in Al-rich phyllosilicates. In contrast, the strongest peak 525 in the 600-800 cm⁻¹ region of inclusions identified as 526 phlogopite by the database is located at $<700 \text{ cm}^{-1}$, verifying an 527 affiliation to the trioctahedral group of phyllosilicates like 528 529 phlogopite-biotite. Furthermore the presence of a peak at ~350 cm^{-1} (present when Mg-rich), the absence of a peak at ~550 530 cm⁻¹ (present when Fe-rich), and the position of the OH-bands 531 between 3650–3750 cm⁻¹ call for a more phlogopitic 532 533 composition (spectrum evaluation based on Wang et al., 2015). 534 However, because discrimination within the solid solution 535 series is difficult and cannot be generalized, for this study, we assigned the mica group mineral inclusions to the muscovite-536 537 paragonite and phlogopite-biotite series, respectively.

538 Raman spectra of pyroxenes can be unambiguously 539 distinguished from other minerals, and the spectral pattern also 540 allows distinct discrimination between clinopyroxenes and 541 orthopyroxenes, because clinopyroxenes have one intense band in the 650–700 cm^{-1} region (Fig. 5a), whereas orthopyroxenes 542 543 show a doublet (Fig. 5b) (Mernagh and Hoatson, 1997; Wang 544 et al., 2001; Buzatu and Buzgar, 2010).

545 In the analyzed garnet grains, some clinopyroxenes were 546 identified as diopside by the database (sometimes also high 547 match probabilities for augite), others as omphacite, and some 548 give similar match probabilities for diopside and omphacite. In 549 Fig. 5a, spectra of nine clinopyroxene inclusions are plotted in 550 comparison with two diopside (ID 040009: 551 $(Ca_{0.97}Fe_{0.02}Na_{0.01})_{\Sigma=1.00}(Mg_{0.97}Fe_{0.02}Al_{0.01})_{\Sigma=1.00}Si_{2.00}O_{6.00},$ ID 552 060171: $(Mg_{0.98}Mn_{0.02})_{\Sigma=1.00}Si_{2.00}O_{6.00})$ $Ca_{1.00}$ and one 553 omphacite (ID 061129.2: $(Ca_{0.51}Na_{0.48})_{\Sigma=0.99}$ spectra $(Mg_{0.44}Al_{0.44}Fe^{2+}_{0.14}Fe^{3+}_{0.02})_{\Sigma=1.04}Si_{2.00}O_{6.00})$ from the RRUFF 554 555 database, and one omphacite spectrum from the UHP eclogite 556 exposed at Flatraket harbor (for location see Fig. 1). All spectra 557 are ordered regarding the position of the main band, which ranges from $\sim 665 \text{ cm}^{-1}$ for the diopside spectra from the 558 database to ~ 684 cm⁻¹ for the spectrum of the omphacite 559 inclusion in garnet grain number 702 of sample AK-N13-1 560 561 (Flatraket). The same trend can be observed for the bands of the triplet in the 300-420 cm⁻¹ region, which positions 562

563 continuously change from $\sim 323 \text{ cm}^{-1}$ (diopsides from database) 564 to $\sim 344 \text{ cm}^{-1}$ (omphacite in grain number 702 of AK-N13-1) 565 for the lowest frequency band, $\sim 356 \text{ cm}^{-1}$ to $\sim 381 \text{ cm}^{-1}$ for the 566 intermediate band, and $\sim 389 \text{ cm}^{-1}$ to $\sim 411 \text{ cm}^{-1}$ for the highest 567 frequency band.

568 Because all inclusions with high match probabilities for 569 omphacite are located above the omphacite spectrum from the 570 RRUFF database in Fig. 5a, and thus show higher Raman shifts 571 for the Raman bands mentioned above, this inclusion type was 572 classified as omphacite. Inclusions which show highest match 573 probabilities for diopside or similar probabilities for diopside 574 and omphacite, in contrast, are located below the omphacite 575 spectrum but above the diopside spectra from the database. 576 These inclusions were classified as diopside. Note that these 577 diopsides may have variable amounts of an omphacite (jadeite) 578 component which, however, cannot be quantified more 579 precisely here.

580 Contrary to the clinopyroxenes, the orthopyroxenes give a clear 581 picture, with all inclusion spectra giving the highest match 582 probabilities for enstatite (example in Fig. 5b). Therefore, this 583 inclusion type was classified as enstatite.

584 The identified amphiboles can be definitely assigned to the 585 amphibole group, but discrimination within the group is not 586 straightforward. Comparison with the database of different

587 spectra from amphibole inclusions (Fig. 5c) gives similar high 588 match probabilities for different amphibole group minerals like 589 actinolite, gedrite, and tremolite. For a more precise 590 discrimination the spectral region of H₂O/OH (3400-3800 cm⁻ ¹), spectra in different orientations from the same amphibole 591 592 with polarized incident light (not appropriate for inclusions in 593 mounted host mineral grains), and a reference amphibole with 594 known chemistry should be considered (Leissner et al., 2015). 595 This is not applicable in this study, and even if it was, 596 uncertainties would remain due to high fluorescence, low 597 signal-to-noise ratios, and possible impurities (Apopei and 598 Buzgar, 2010). For these reasons, amphibole inclusions are 599 classified as the amphibole group inclusion type, without any 600 subdivision. The presence of glaucophane, however, can be 601 excluded because the characteristic glaucophane band at ~385 cm^{-1} was not obtained in any of the amphibole group spectra. 602

603 Similar to the amphibole group inclusion type, also the epidote 604 group minerals, the carbonate minerals, and the sulfate minerals 605 were grouped. Epidote group mineral inclusions only occur in 606 ~5% of the analyzed garnets in the Flatraket sample, and 607 comparison with the database gives similar high match 608 probabilities for clinozoisite, epidote, and zoisite, whereas in 609 particular clinozoisite and epidote cannot be discriminated by their Raman spectra in the $<1200 \text{ cm}^{-1}$ region. Carbonate and 610 611 sulfate minerals are more frequent in the samples and the 612 database identified most of them as dolomites and anhydrites, 613 respectively. However, in case of the carbonate minerals often 614 high match probabilities are also given for magnesite, calcite 615 and siderite. In the case of sulfate minerals, some inclusions are 616 identified as gypsum and celestine, and others cannot be 617 assigned to a specific sulfate mineral. Overall, the partially 618 uncertain assignment to a specific mineral and the low 619 significance of specific minerals of this group as a source rock 620 indicator suggests that grouping as carbonate or sulfate 621 minerals is sufficient for this study. Note, however, that most 622 carbonate mineral inclusions are most likely dolomite and most 623 sulfate mineral inclusions are most likely anhydrite.

Opaque minerals with a weak Raman response are present in ~5–7% of the analyzed garnets. They were not identified in this study and are excluded from further discussion. Identification requires long accumulation times and strong corrections to isolate the inclusion spectra from the inclusion/host mixed spectra, which is beyond the scope of this study.

Besides the mineral inclusions in the detrital garnets, CO_2 fluid inclusions were also detected. Fluid inclusions can provide information about different stages of rock formation, but fluid inclusion studies are very time consuming (van den Kerkhof and Hein, 2001) and are also beyond the scope of this study.

635 **5.1.2** Inclusions in detrital garnets from Flatraket

636 Overall, 148 grains of the 150 analyzed grains (~99%) from the 637 stream-sediment sample of Flatraket (AK-N13-1) are garnet. 638 The two other mineral grains (grain number 15 = titanite; grain 639 number 364 = enstatite) were confounded with garnet during 640 picking. Of the 148 grains, 130 (~88%) exhibit inclusions 641 which are sufficient in size (i.e., $\geq 2 \mu m$) to be analyzed by 642 Raman spectroscopy. The amount of garnets with analyzable 643 inclusions increases with increasing grain size from ~84% (41 644 of 49) in the fine fraction (63–125 μ m), over ~88% (43 of 49) 645 in the medium fraction (125–250 μ m), to ~92% (46 of 50) in 646 the coarse fraction (250–500 µm). The detected inclusion types 647 for each of the analyzed garnets from Flatraket are listed in 648 Supplementary Table 1. In Supplementary Table 2 the 649 proportion of the analyzed garnet grains with specific inclusion 650 types is summarized. These results are shown in Fig. 6. In 651 general, when inclusions of a specific type occur within the 652 analyzed garnets, on average 1 to 4 inclusions of the same type 653 are present in the garnet host. Only quartz inclusions are more 654 frequent, with on average 5 to 20 inclusions per quartz-bearing 655 garnet.

Quartz, rutile, and mica group minerals represent the main
mineral inclusions, which are present in the bulk of the garnet
grains. Quartz is the most frequent inclusion type, and is
present in ~63% (82 of 130) of the garnets. The proportion of

660 grains which exhibit quartz inclusions is almost constant over 661 the three grain-size fractions (~61 to 65%). Rutile inclusions are also rather frequent and occur in ~43% (56 of 130) of the 662 663 garnets. The coarser the grain-size fraction, the more frequent 664 are rutile inclusions. Starting from ~29% of the garnet grains in 665 the fine fraction, they are more frequent in the medium fraction 666 (~44%), and the highest amount of rutile inclusions is found in 667 the coarse fraction (~54%). Besides quartz and rutile, mica 668 group minerals are abundant inclusions in garnet grains from 669 Flatraket. They are composed of the muscovite-paragonite and 670 phlogopite-biotite types. Phlogopite-biotite (~36%; 47 of 130) 671 is more abundant than muscovite-paragonite (~23%; 30 of 672 130). Both together are present in ~52% (67 of 130) of the 673 garnets. Generally, mica inclusions are more frequent in the 674 fine grain-size fraction compared to medium and coarse 675 fractions (Fig. 6).

676 The main mineral inclusions are followed by inclusions of 677 apatite, sulfate minerals, and feldspars, which are present in a 678 significant amount of the host garnets (~20 to 30%). Apatite 679 inclusions occur in ~28% (37 of 130) of the garnet grains. 680 While apatite inclusions in the fine fraction are only present in 681 ~15% of the grains, they are more frequent in the medium and coarse fractions (~37% and ~33%, respectively). Inclusions of 682 683 sulfate minerals are present in $\sim 24\%$ (31 of 130) of the garnets. 684 Sulfate mineral inclusions are highest in the medium grain-size

685 fraction with \sim 30%, and less abundant in the fine and coarse 686 fraction (~20% and ~22%, respectively). Feldspar inclusions, 687 including alkali feldspar and plagioclase, are present in ~22% 688 (29 of 130) of the garnets. They are less frequent in the medium 689 grain-size fraction with ~12%, but more abundant in the fine 690 and coarse fractions (~24% and ~30%, respectively). The 691 number of garnets with alkali feldspar inclusions is 692 considerably smaller in the medium fraction ($\sim 2\%$) than in the 693 fine (~15%) and coarse (~20%) fractions, while plagioclase 694 inclusions are rather evenly distributed over all grain-size 695 fractions (~9 to 13%; Fig. 6).

696 Carbonate minerals, clinopyroxenes, zircon, kyanite, epidote 697 group minerals, and amphibole group minerals are less 698 abundant and occur as inclusions in <20% of the garnets. 699 Carbonates are present in ~12% (16 of 130) of the garnets. 700 These are least frequent in the medium grain-size fraction 701 $(\sim 7\%)$ and more abundant in the fine and coarse fractions 702 (~12% and ~17%, respectively). Overall, clinopyroxene 703 inclusions are present in ~10% (13 of 130) of the garnets, with 704 omphacite dominating over diopside (Fig. 6). While in the fine 705 fraction only ~5% of the garnets contain inclusions of 706 clinopyroxene, this number increases with increasing grains 707 size to $\sim 7\%$ in the medium fraction and $\sim 17\%$ in the coarse 708 fraction. Zircon and kyanite inclusions occur in ~7% (9 of 130) 709 of the garnet grains. Both are most frequent in the coarse

fraction (~15% and 13%, respectively), while in the two finer fractions they are rare (~2 to 5%). Inclusions of the epidote group and amphibole group minerals occur with ~5% (7 and 6 out of 130, respectively) of the garnets. Both are slightly more frequent in the fine fraction (~7%) and continuously decrease in the coarser fractions (Fig. 6).

716 **5.1.3 Inclusions in detrital garnets from Runde**

717 Overall, 150 grains (50 of every grain-size fraction) were 718 analyzed from the beach-sediment sample of Runde (AK-N37), 719 whereby 148 grains of it (~99%) are garnets, and two are other 720 mineral grains which were confounded with garnet during 721 picking (grain number 351 = plagioclase; grain number 395 =722 alkali feldspar). Of the 148 grains, 119 (~80%) exhibit 723 inclusions $\geq 2 \mu m$. The amount of garnet grains with inclusions 724 $\geq 2 \mu m$ is almost constant and only slightly increases from 725 ~78% (39 of 50) in the fine fraction (63–125 μ m), over ~81% 726 (39 of 48) in the medium fraction (125–250 μ m), to ~82% (41 727 of 50) in the coarse fraction (250-500 µm). The detected 728 inclusion types for each of the analyzed garnets from Runde are 729 listed in Supplementary Table 3. In Supplementary Table 4 the 730 proportion of the analyzed garnet grains with specific inclusion 731 types is summarized. These results are shown in Fig. 7. In 732 general, when inclusions of a specific type occur within the 733 analyzed garnets, on average 1 to 4 inclusions of the same type 734 are present in one garnet host. Only rutile inclusions are more frequent with on average 5 to 20 inclusions per rutile-bearinggarnet.

737 The most frequent inclusion types within the garnets of the 738 beach-sediment sample are quartz and rutile. Quartz inclusions 739 are present in ~42% (50 of 119) of the grains. They are 740 widespread in the medium grain-size fraction (~49%) and less 741 frequent in the fine and coarse fractions (~38% and ~39%, 742 respectively). Rutile inclusions occur in ~40% (48 of 119). 743 They are also most common in the medium grain-size fraction 744 (~51%) and less frequent in the coarse fraction (~44%). A 745 much smaller amount (~26%) contain rutile inclusions in the 746 fine grain-size fraction.

747 Inclusions of mica and feldspar group minerals are also present 748 in significant amounts of the garnet grains. Mica group mineral 749 inclusions are solely composed of phlogopite-biotite. They 750 occur in ~24% (29 of 119) of the host garnets and show only 751 minor fluctuations across the three grain-size fractions with a 752 slight increase with increasing grain-size fraction (~23 to 27%). 753 Feldspar inclusions are present in ~21% (25 of 119) of the 754 garnets. They are composed of alkali feldspar and plagioclase 755 inclusions, for which alkali feldspar is less frequent. Together, 756 the amount of garnets which exhibit feldspar inclusions is 757 almost constant over the three grain-size fractions (~20 to 758 23%). By considering alkali feldspar and plagioclase

separately, an enrichment of both in the medium fractioncompared to the fine and coarse fraction is indicated (Fig. 7).

761 Lower but also significant amounts of garnet grains carry 762 inclusions of zircon, apatite, kyanite, and pyroxene. Zircon 763 inclusions occur in ~18% (22 of 119) of the garnets. While the 764 proportion of grains with zircon inclusions in the fine fraction 765 is ~15%, and in the coarse fraction ~17%, they are slightly 766 more frequent in the medium fraction where they were 767 identified in ~23% of the garnets. Inclusions of apatite are 768 present in ~18% (21 of 119) of the garnets. Both the fine and 769 medium fraction, show ~15% grains with apatite inclusions. In 770 the coarse fraction, apatite inclusions are more frequent and 771 occur in ~22% of the grains. Kyanite inclusions are present in 772 ~17% (20 of 119) of the studied garnet grains. With increasing 773 grain-size fraction, the amount of garnets with kyanite 774 inclusions increases from ~10 to 22%. Overall, ~15% (18 of 775 119) of the garnet grains exhibit pyroxene inclusions, whereas 776 the amount decreases with increasing grain-size fraction. In the 777 fine fraction, pyroxene inclusions occur in ~21%, in the 778 medium fraction in $\sim 13\%$, and in the coarse fraction in $\sim 12\%$ 779 of the grains. Pyroxene inclusions include the two 780 clinopyroxene types, omphacite and diopside, and the 781 orthopyroxene type, enstatite. Clinopyroxene inclusions consist 782 predominantly of diopside, which occurs in ~11% (13 of 119) of the garnets. Diopside is more frequent in the fine fraction 783

784 $(\sim 18\%)$ than in the medium and coarse fraction ($\sim 8\%$ and $\sim 7\%$, 785 respectively). Only 2 out of the 119 garnets (~2%) contain 786 clinopyroxene inclusions which were classified as omphacite. 787 Orthopyroxene (i.e., enstatite) inclusions are much less frequent 788 than clinopyroxene inclusions. They are present in ~5% (6 of 789 119) of the garnets and in one-third of the cases the 790 orthopyroxene inclusions are located in garnet grains which 791 also carry clinopyroxene inclusions. With ~8%, the proportion 792 of garnets that exhibit orthopyroxene inclusions is the highest 793 in the medium grain-size fraction. In the coarse and fine 794 fraction the proportion is lower (~5% and ~3%, respectively).

795 Furthermore, also carbonate mineral, amphibole group, and 796 sulfate mineral inclusions are present in a smaller proportion of 797 host garnets. Over the three grain-size fractions, carbonate 798 mineral inclusions occur in ~9% (11 of 119) of the garnets. The 799 amount of garnets with carbonate mineral inclusions decreases 800 with increasing grain-size fraction (~18 to 5%). Amphibole 801 inclusions occur in ~7% (8 of 119) of the grains. In the fine and 802 medium fraction, ~8% of the garnets exhibit amphibole 803 inclusions and in the coarse fraction they are slightly less 804 frequent (~5%). Sulfate mineral inclusions occur in ~6% (7 of 805 119) of the garnets, and these are only present in the fine and medium fraction ($\sim 8\%$ and $\sim 10\%$, respectively). 806

807 In addition, three inclusions of coesite were identified in the808 analyzed grains. One in grain number 24 of the fine grain-size

fraction and two in grain number 378 of the medium fraction.
That means ~2% (2 of 119) of the analyzed garnet grains
contain coesite inclusions. A detailed description of the coesitebearing garnets, four more coesite-bearing garnets from the
same sample, the characteristics of the coesite inclusions, and
the resulting implications are given in Schönig et al. (2018).

815 **5.2 Garnet geochemistry**

816 The garnet compositions measured by the electron microprobe are given in Supplementary Table 5. Four garnet grains (grain 817 818 number 22 from AK-N13-1, and grains 40, 395, and 732 from 819 AK-N37) were excluded because the coating was damaged or 820 composition indicates contamination the measured bv 821 inclusions. The compositions of the coesite-bearing garnets 822 number 24 and 378 from sample AK-N37 (Runde) were 823 measured at several spots (Schönig et al., 2018) and averaged 824 for this study.

825 The multivariate statistical discrimination scheme of Tolosana-826 Delgado et al. (2017) was used to discriminate between garnet 827 host rocks. Because the sampled catchments are almost 828 exclusively composed of metamorphic rocks, we choose the 829 prior probability 'equal-M' with equal probabilities of 30% for 830 all metamorphic host-rock groups (A - eclogite facies; B -831 amphibolite facies; C – granulite facies), and significantly 832 lower probabilities of 5% for ultramafic (D) and felsic plutonic 833 rocks (E1). However, we also tested the prior probability 'global', and all analyzed garnets were still classified asmetamorphic garnets.

836 Probabilities for the detrital host garnets of belonging to the 837 major host-rock groups are given in Supplementary Table 5. 838 These are plotted for the major metamorphic host-rock groups 839 in Fig. 8a (Flatraket) and Fig. 9a (Runde). In Figs. 8b-i and 9b-840 i, these host-rock probabilities are shown separately for garnets 841 with specific inclusion types. All of these ternary diagrams are 842 complemented with pie charts indicating the proportion of 843 grains belonging with highest probability to one of the 844 metamorphic host-rock groups A, B, and C (for detailed 845 numbers see Supplementary Table 6).

846 Overall, both catchments show similar characteristics. The 847 major proportion of garnets (~59% for Flatraket, ~47% for 848 Runde) show highest probability of belonging to the granulite 849 facies group C, followed by garnets assigned to the eclogite 850 facies group A (~39% and ~40%, respectively). Garnets 851 assigned to the amphibolite facies group are subordinate (~3% 852 and ~12%, respectively). This pattern is similar for all grain-853 size fractions, but garnets assigned to the amphibolite facies 854 from the Flatraket sample are only present in the fine fraction.

Furthermore, quartz, rutile, kyanite, apatite, and zircon
inclusions occur – with minimal deviation – in garnets of all
geochemical compositions determined as the detrital overall

858 composition (as illustrated in Figs. 8a, 9a). There are, however, 859 variations in their proportions (see Supplementary Table 6 for 860 exact proportions). Whereas kyanite inclusions in AK-N37 861 (Runde) show a similar distribution like the detrital overall 862 composition (Fig. 9c), they are slightly less frequent in type A 863 (eclogite facies) garnets of AK-N13-1 (Flatraket) (Fig. 8c). 864 Quartz and apatite inclusions in both catchments are less 865 frequent in type A garnets, but instead more abundant in type C 866 (granulite facies) and/or type B (amphibolite facies) garnets 867 (Figs. 8b, 8h, 9b, 9h). While rutile and zircon inclusions at 868 Flatraket are more frequent in type A and less frequent in type 869 C garnets (Figs. 8c, 8h), rutile inclusions are less frequent in 870 type A garnets from Runde (Fig. 9c), and zircon inclusions 871 from Runde show a similar pattern like the overall composition 872 (Fig. 9h).

873 Inclusions of mica group minerals (muscovite-paragonite and 874 phlogopite-biotite) and alkali feldspar are enriched in garnets 875 of type C, and sometimes also type B, compared to type A 876 garnets (Figs. 8f, 8g, 9f, 9g). In contrast, omphacite and epidote 877 group inclusions are much more frequent in type A garnets, and 878 do not occur, or only subordinated, in type B and C garnets 879 (Figs. 8d, 8e, 9d). The same applies to diopside inclusions in 880 the Flatraket sample (Fig. 8d), but in the sample from Runde 881 diopsides are more abundant in type C garnets (Fig. 9d). 882 However, these diopsides are mainly close to the transition
zone between type A and C, and thus, have also high
probabilities of belonging to type A. Enstatite and amphibole
group inclusions show a very similar pattern to these diopside
inclusions of the transition zone. They are more frequent in
type C garnets, but the type C garnets often also have high
probabilities of belonging to type A (Figs. 8e, 9d, 9e).

889 Inclusions of plagioclase, carbonate minerals, and sulfate 890 minerals show a less distinct pattern. They are present in 891 garnets of a large part of the overall composition, but not to the 892 same extent like quartz, rutile, kyanite, apatite, and zircon. 893 Plagioclase inclusions are slightly enriched in type A 894 (Flatraket) or type B (Runde) garnets, compared to the overall 895 composition (Figs. 8g, 9g). While inclusions of carbonate and 896 sulfate minerals in the Flatraket sample are present in similar 897 proportions of garnet types like the overall composition (Fig. 898 8i), carbonate mineral inclusions are more abundant in type C 899 garnets and less abundant in type A garnets in the sample from 900 Runde, which is contrary to the sulfate mineral inclusions in the 901 Runde sample (Fig. 9i).

902 Coesite inclusions in the analyzed garnets from Runde are only 903 present in type C garnets. However, this is an effect of the 904 small number of garnets containing coesite. Schönig et al. 905 (2018) analyzed an about five times larger number of garnets 906 from the same sample and found six garnets with coesite 907 inclusions, whereby three of the garnets are clearly classified as 908 type C, one as type A, and two have almost equal probabilities909 of belonging to type A and C.

910 6. Discussion

911 6.1 Provenance information value of the obtained912 inclusion types

913 The high proportion (\geq 80%) of detrital garnet grains containing 914 mineral inclusions $\geq 2 \mu m$ indicates that inclusions in garnets of 915 metamorphic origin are potentially useful to gain provenance 916 information. The ultimate aim is to identify specific 917 metamorphic source rock types by analyzing these inclusions. 918 When analyzing crystalline rocks, metamorphic facies types are 919 differentiated by identification of diagnostic mineral 920 assemblages, i.e., associations of mineral species in mutual 921 grain contact (e.g., Bucher and Frey, 2002). In classical 922 provenance studies, this information is not available due to the 923 consideration of individual mineral grains which are not in 924 contact with other mineral species. However, by considering 925 mineral inclusions in the detrital single grains, mineral 926 assemblage information of the source rocks can be obtained 927 because the inclusions were entrapped during growth of the 928 host mineral. Thus, an inclusion and its host mineral are 929 coexisting since entrapment occurred during the same 930 metamorphic stage. Caution should be taken when considering 931 more than one inclusion in a single grain. Garnet, in particular, 932 can record multiple metamorphic stages and isolate entrapped 933 mineral inclusions from subsequent events. Thus, adjacent 934 inclusions in garnet do not necessarily represent the same 935 metamorphic stage. Therefore, mineral assemblages derived 936 from more than one inclusion give hints but are no evidence for 937 coexistence during entrapment. In contrast, when considering 938 only single inclusions in garnet, these directly reflect a two-939 mineral assemblage (garnet host + inclusion phase), which 940 coexisted during the same metamorphic stage (Schönig et al., 941 2018).

942 By focusing on diagnostic two-mineral assemblages, i.e., single 943 mineral inclusions, the bulk of inclusion types detected in the 944 analyzed garnets can be grouped regarding their use in source 945 rock identification as (i) inclusions of little or no information, 946 (ii) inclusions indicating a wide range of metamorphic source 947 rocks. (iii) inclusions indicating a smaller range of 948 metamorphic source rocks, and (iv) inclusions which yield 949 specific source rock information.

(i) Quartz, feldspar, zircon, apatite, mica, carbonate minerals,
and sulfate minerals are assigned to the group of inclusions of
little or no information. Quartz and feldspars are abundant
rock-forming minerals, which cannot be linked to a specific
rock type. In general, quartz and alkali feldspar are more
abundant in felsic lithologies, and plagioclase is more abundant
in mafic lithologies, but all can also occur vice versa.

957 Especially eclogites often contain quartz. The presence of 958 quartz inclusions also does not exclude pressure conditions of 959 the coesite stability field during entrapment because quartz 960 inclusions can be present very close to coesite inclusions, 961 which – in some cases – is suggested to be a preservation effect 962 of coesite inclusions (Schönig et al., 2018). Similar to quartz 963 and alkali feldspar, apatite and zircon are also more frequent in 964 felsic lithologies, but they occur in mafic rocks as well, so that 965 they cannot be linked to a specific rock type. Likewise, also the 966 mica minerals, here muscovite-paragonite group and 967 phlogopite-biotite, carbonate and sulfate minerals, can occur in 968 a wide range of metamorphic rocks, and do not point to specific 969 P-T conditions during entrapment.

970 (ii) Amphibole group and epidote group inclusions are assigned 971 to the group indicating a wide range of metamorphic source 972 rocks. These hydrous minerals disappear at high temperatures 973 in the granulite facies (Bucher and Frey, 2002). Thus, these 974 inclusions cannot be captured by garnet growing during 975 granulite facies conditions, and either temperature conditions 976 have to be lower (e.g., amphibolite facies) or pressure 977 conditions have to be higher (eclogite facies) during 978 entrapment.

979 (iii) Kyanite, rutile, diopside, and enstatite are assigned to the
980 group of inclusions indicating a smaller range of
981 metamorphic source rocks. Kyanite belongs together with

982 sillimanite and andalusite to the trimorphic Al₂SiO₅ group, 983 which all together are only stable at the triple point at ~4.2 kbar 984 and ~530°C (Bohlen et al., 1991). Due to their different 985 stability fields, these aluminosilicates are often used as 986 metamorphic index minerals, whereby the presence of kyanite 987 is restricted to the HP parts of the greenschist, amphibolite, and 988 granulite facies or even higher P-T ratios (Fig. 10a). Rutile is 989 an abundant accessory mineral in medium- to high-grade 990 metamorphic rocks (e.g., Force, 1980). It is newly formed at 991 upper amphibolite facies conditions (or higher), and increasing 992 pressure favors rutile formation (Zack et al., 2004). When 993 subjected to lower-grade conditions at subsequent events, 994 matrix rutile breaks down to form other titanium-bearing 995 phases (Force, 1980; Zack et al., 2004). Therefore, it is very 996 unlikely that garnet is able to capture rutile outside the P-T997 conditions where it is newly formed. Thus, a rutile inclusion in 998 garnet reflects entrapment at upper amphibolite facies 999 conditions or higher. Similarly, diopside inclusions also 1000 indicate upper amphibolite facies conditions or higher because 1001 diopside typically occurs in mafic to ultramafic rocks, in which 1002 the appearance of clinopyroxene defines the lower-grade 1003 boundary of the upper amphibolite facies (Bucher and Frey, 1004 2002). The suggested omphacite component of the diopsides in 1005 furthermore, point to even higher-grade this study, 1006 metamorphic conditions. Enstatite is a typical orthopyroxene of

high-temperature (HT) metamorphic rocks like granulites, and
it can also be present in HT eclogites (Bucher and Frey, 2002;
Okrusch and Matthes, 2014). Thus, enstatite inclusions are
thought to reflect HT conditions during entrapment in garnet.

1011 (iv) Omphacite and coesite inclusions belong to the group of 1012 inclusions which yield specific source rock information. The 1013 mineral assemblage garnet plus omphacite is the diagnostic 1014 mineral assemblage of eclogite (e.g., Bucher and Frey, 2002), 1015 and therefore omphacite inclusions in garnet directly indicate 1016 an eclogitic source. Moreover, the presence of coesite 1017 inclusions specifically indicates UHP metamorphic conditions 1018 because it is not possible to form coesite inclusions in garnet at 1019 conditions lower than the coesite stability field (e.g., Schönig et 1020 al., 2018, and references therein).

1021 **6.2** Restriction of possible metamorphic source rocks

1022 By combining the information given by the obtained inclusion 1023 types in the detrital garnets with the geochemical garnet 1024 compositions, the suite of possible source rocks in both 1025 catchments can be strongly reduced.

1026 First of all, kyanite inclusions are present in significant 1027 amounts of the garnets with ~7% at Flatraket and ~17% at 1028 Runde. Due to their abundance, the occurrence in garnets of all 1029 obtained geochemical compositions (Figs. 8c, 9c), and the 1030 absence of any other aluminosilicate inclusions, all garnet

1031 source rocks in the catchments are assumed to be equilibrated 1032 in the kyanite stability field. Thus, all metamorphic source rock 1033 facies types of the andalusite and sillimanite stability field 1034 metamorphism, (contact lowermost greenschist, lower 1035 amphibolite, and lower to intermediate granulite facies) can be 1036 excluded (field A in Fig. 10b). Note that even in these proximal 1037 samples, kyanite made up <2% of the heavy mineral 1038 assemblage (Krippner et al., 2016), indicating that this 1039 important metamorphic facies index mineral preferably occurs 1040 in the form of inclusions in garnet rather than as single mineral 1041 in the typical grain-size range of 63-125 µm (or coarser) 1042 analyzed in heavy mineral studies. This trend will probably 1043 intensify with increasing transport distance because kyanite is 1044 mechanically less stable than garnet (e.g., Morton and 1045 Hallsworth, 1999).

1046 In the blueschist facies, garnet first appears in the HP part 1047 (Bucher and Frey, 2002). If garnet from the sampled 1048 catchments would have developed under HP blueschist facies 1049 conditions, some typical blueschist facies mineral inclusions 1050 like glaucophane or lawsonite would be expected. The absence 1051 of these inclusion types, and the high abundance of phlogopite-1052 biotite inclusions (>20% of garnet grains), which are atypical 1053 for blueschist facies rocks (Bucher and Frey, 2002), makes 1054 blueschist facies rocks unlikely as a garnet source (field B in 1055 Fig. 10b). Additionally, the absence of lawsonite inclusions makes a very-low-temperature eclogitic source unlikely (field
C in Fig. 10b) because lawsonite inclusions are well known in
garnets from lawsonite-eclogites (e.g., Tsujimori et al., 2006,
and references therein).

1060 As in the blueschist facies, greenschist facies garnet first 1061 appears in the HP part (Bucher and Frey, 2002). Therefore, 1062 metamorphic rocks of subgreenschist and lower greenschist 1063 facies can be excluded as a garnet source (field D in Fig. 10b). 1064 Furthermore, also the HP part of the greenschist facies and the 1065 lower amphibolite facies can be excluded (field E in Fig. 10b) 1066 due to the very high abundance of rutile inclusions (present in 1067 >40%) in garnets of all obtained compositions (Figs. 8c, 9c). 1068 Note that although rutile is ultrastable in sedimentary 1069 environments, it makes up only <2% of the heavy mineral 1070 assemblage (Krippner et al., 2016) but is, besides quartz, the 1071 dominant inclusion type in the detrital garnets.

1072 In summary, by considering the conditions where garnet first 1073 appears, and the presence/absence of a few inclusion types, the 1074 possible source rocks of garnets in both catchments can be 1075 reduced to rocks of upper amphibolite, HP granulite, and 1076 eclogite facies (excluding very-low-temperature eclogites). 1077 This well reflects the experienced conditions of the whole 1078 region and the range of outcropping metamorphic rocks in the 1079 sampled catchments. Beyond these general constraints, the 1080 occurrence of certain mineral inclusions in combination with their preferable host garnet composition reveals some specificcatchment characteristics.

1083 6.3 Specific catchment characteristics of Flatraket (AK1084 N13-1)

1085 The geochemical classification of the garnets from Flatraket 1086 implies that by far the most garnets are derived from granulite 1087 to eclogite facies source rocks (Fig. 8a). However, a clear 1088 separation into different sources by only considering the 1089 geochemical composition is not possible.

1090 The fact that eclogite facies rocks are involved as source rocks 1091 for parts of the garnets is clearly indicated by the presence of 1092 omphacite inclusions in ~8% of the garnets, and an additional 1093 indication is given by the presence of diopside inclusions in 1094 $\sim 2\%$ of the garnets, which probably have an omphacite 1095 (jadeite) component. Most of these garnets are correctly 1096 assigned to type A garnets by the multivariate discrimination 1097 (Fig. 8d), confirming that the discrimination of eclogite facies 1098 garnets works well as reported by Tolosana-Delgado et al. 1099 (2017). However, occasionally some of them have the highest 1100 probability for type C. This is also highlighted by garnet 1101 compositions from the local eclogites determined by Krippner 1102 et al. (2016), which mainly have the highest probability of 1103 belonging to type A, but garnets from one of the analyzed 1104 eclogites also show highest probabilities of belonging to type C 1105 (Fig. 8a). Thus, the occurrence of omphacite and diopside inclusions in type C garnets reveals that parts of the type C
classified garnets are also derived from eclogite facies rocks.
This part can be roughly estimated to be restricted to garnets
with a high probability of belonging to type A (>30%), and a
small probability of belonging to type B (<12%).

1111 Besides the mineral inclusions which are present in almost all 1112 detrital garnets (i.e., quartz, rutile, kyanite, apatite, zircon), the 1113 preferable occurrence of epidote and amphibole inclusions in this eclogite facies garnets (Fig. 8e) indicates that these 1114 1115 minerals are characteristic for the eclogite facies source rocks. 1116 The presence of these hydrous mineral inclusions also makes a 1117 granulite facies source improbable. Overall, the deduced 1118 mineral assemblage of garnet + omphacite/diopside + epidote + 1119 amphibole + rutile + quartz \pm kyanite for the eclogite facies 1120 garnet type match well with mineral assemblages from 1121 eclogites reported from the region of Flatraket (Table 1). An 1122 additional sign for a mafic source is the low abundance of 1123 alkali feldspar inclusions in these garnets (Fig. 8g).

In addition to the eclogite facies garnets, another large group of garnets are assigned to type C with probabilities $\geq 12\%$ of belonging to type B. Besides the mineral inclusions which are present in almost all detrital garnets, inclusions of mica group minerals (muscovite–paragonite and phlogopite–biotite) and alkali feldspar are frequent in these garnets, which make a felsic source more probable (Figs. 8f, 8g). Additionally, also

1131 quartz and apatite are slightly more frequent (Figs. 8b, 8h), 1132 supporting the presumption of a felsic source. This is also 1133 highlighted by garnet compositions from the local gneisses 1134 determined by Krippner et al. (2016). Garnet composition from 1135 the felsic gneiss with a medium garnet content (AK-N13-2d, 1136 plotting as type C in Fig. 8a) is very similar to the composition 1137 of detrital garnets which are suggested to be of felsic origin. In contrast, no garnets with a comparable composition to the other 1138 1139 felsic gneiss (AK-N13-2c, plotting as type B in Fig. 8a) are 1140 present in the detritus. This is not surprising, as this gneiss 1141 exhibits only a very low garnet content. Overall, it seems 1142 reliable that the group of garnets assigned to type C with 1143 probabilities $\geq 12\%$ of belonging to type B are mainly derived 1144 from the felsic gneisses. Geochemical characterization favors a granulite facies source, but a precise assignment to a 1145 1146 metamorphic facies type by inclusions cannot be specified, 1147 because no unique inclusion types for amphibolite-, granulite-1148 and/or eclogite facies exist or were detected.

1149 In summary, the main proportion of detrital garnets is estimated 1150 to be derived from eclogites (~55%), whereas the residual 1151 garnets (~45%) derived from the felsic gneisses, are probably 1152 mainly of granulite facies. Amphibolite facies garnet source 1153 rocks play a subordinate role due to their low garnet content.

1154 Considering the different grain-size fractions, the eclogitic 1155 garnets are more frequent in the 125–250 μ m (~63%) and 250–

1156	500 μ m grain-size fraction (~58%), and less frequent in the 63–
1157	125 μ m fraction (~45%). By contrast, felsic garnets are less
1158	frequent in the 125–250 μm (~38%) and 250–500 μm grain-
1159	size fraction (~42%), and more abundant in the 63–125 μm
1160	fraction (~55%). Although these numbers are only a rough
1161	approximation, it is obvious that detrital eclogitic garnets are
1162	much more abundant than eclogitic rocks in the catchment area
1163	(Fig. 1), and that these are more frequent in the coarser grain-
1164	size fractions. Similar observations were made in other regions
1165	comprising eclogites in their catchment (e.g., Krippner et al.,
1166	2015). Typical factors which quantitatively modify the
1167	composition of heavy minerals from source to the sampling
1168	location can be widely excluded in the sampled catchment. The
1169	high topography and the climatic conditions of the area cause a
1170	mainly mechanical weathering of the source rocks. Chemical
1171	weathering during transport should be of minor importance due
1172	to the high energy of the drainage system and the short
1173	transport distance (<4 km). The short transport distance also
1174	minimizes the elimination of heavy minerals due to hydraulic
1175	effects. Probably the mineralogical composition of the source
1176	rocks is the main reason why detrital eclogitic garnets are so
1177	prominent. Eclogites mainly consist of omphacite and garnet,
1178	and thus supply a much higher amount of heavy minerals to the
1179	sedimentary load than felsic rocks compared to their volumetric
1180	dimension. Additionally, the heavy mineral assemblage derived

1181 from eclogites (omphacite + garnet) is usually enriched in 1182 garnet compared to the assemblage of felsic rocks. The 1183 predominant occurrence in the coarser grain-size fractions is 1184 probably an effect of inherited grain size due to the short 1185 transport distance and the usually large garnet grain size in 1186 eclogites.

Specific catchment characteristics of Runde (AK-N37)

1187

6.4

1188 The geochemical classification of the garnets from Runde 1189 implies at least two different sources (Fig. 9a). One group 1190 shows the highest probabilities for belonging to type A source 1191 rocks, agreeing with the geochemical garnet composition from 1192 an eclogite located on the island. The other group of garnets 1193 gives probabilities of $\leq 25\%$ for type A and high probabilities 1194 for type C (\geq 35%) with variable type B component. 1195 Additionally, several garnets are in the transition zone between 1196 these two groups, showing high probabilities for type C and 1197 type A.

1198 The fact that eclogite facies source rocks are involved as a 1199 source for the garnets of the beach-sediment sample AK-N37 is 1200 given by the scarce presence of omphacite inclusions in $\sim 2\%$ of 1201 the grains. Presumably, also the garnets containing diopside 1202 inclusions (~11%) were derived from eclogite facies rocks, 1203 because the diopsides probably have an omphacite (jadeite) 1204 component, and eclogites with sodic diopside (13% jadeite) are 1205 known from the island of Runde. Garnets containing omphacite

1206 are geochemically well assigned to type A, but garnets with 1207 diopside inclusions are located in the transition zone between type A and type C (Fig. 9d). The garnets having higher 1208 1209 probabilities for type C often contain, besides diopside, also 1210 amphibole group mineral inclusions (Fig. 9e), which makes an affiliation to granulite facies source rocks (type C) less 1211 1212 probable. This also means that at least some of the enstatite-1213 bearing garnets (Fig. 9d), which additionally often contain 1214 diopside and/or amphibole group inclusions, probably do not 1215 originate from granulite facies rocks. Instead, the coexistence 1216 of garnet + omphacite/diopside + amphibole + enstatite, and the 1217 absence of epidote group minerals, favors a high-temperature 1218 eclogitic source for the garnets with highest probabilities of 1219 belonging to type C but which also have high probabilities for 1220 type A. However, caution should be taken when considering 1221 more than one inclusion, but in general, the higher temperatures 1222 supposed for the source rocks of the garnets from Runde 1223 compared to Flatraket are in agreement with the estimated 1224 higher peak metamorphic temperatures of the region (Fig. 1). 1225 Additionally, the low frequency of alkali feldspar and quartz 1226 inclusions in these garnets supports a mafic source. Thus, also 1227 the garnets in the transition zone between the two groups of 1228 garnets are assigned to the eclogitic source.

1229 The other group of garnets with probabilities of $\leq 25\%$ for type 1230 A and high probabilities for type C ($\geq 35\%$) are characterized by

1231 an increased occurrence of alkali feldspar, quartz, rutile, and 1232 phlogopite-biotite, favoring a felsic source. There is no 1233 coincidence with geochemical garnet compositions from the 1234 gneiss reported by Krippner et al. (2016) (Fig. 9a), which was 1235 to be expected because the sampled gneiss unit (AK-N39-1) is 1236 only located at the outermost part of the catchment (Fig. 1). 1237 Probably the garnets mainly derived from the micaceous 1238 quartzo-feldspathic gneiss, which is the main rock type of the 1239 catchment.

Moreover, from the coesite inclusions in two garnets assigned to the felsic source (Fig. 9b), and the coesite inclusions reported by Schönig et al. (2018), which can be assigned to the felsic and the eclogitic source (including garnets of the transition zone), it is indisputable that at least parts of some felsic source rocks experienced UHP metamorphic conditions.

1246 In summary, the main portion of detrital garnets is estimated to 1247 be derived from eclogites (~57%), which probably equilibrated 1248 under higher temperatures than that located in the catchment of 1249 Flatraket. Another large portion of garnets (~36%) is assigned 1250 to felsic source rocks, which are mainly assigned to granulite 1251 facies and to a lesser extent to upper amphibolite facies. Source 1252 rocks of the residual ~7% cannot be specified, and are thus 1253 assigned to source rocks of upper amphibolite facies or higher.

1254 Considering the different grain-size fractions, the supposed 1255 eclogitic garnets are more frequent in the 125-250 µm grain-1256 size fraction (~65%), and less frequent in the 63-125 µm 1257 (~57%) and 250–500 µm fraction (~54%). Contrary, felsic 1258 garnets are less frequent in the 125–250 µm grain-size fraction 1259 (~27%), and more abundant in the 63–125 μ m (~36%) and 1260 250-500 µm fraction (~42%). Overall, at Runde eclogitic 1261 garnets are much more frequent than eclogitic rocks in the 1262 catchments due to the mineralogical composition of eclogites, 1263 but the grain-size effect is less pronounced than at Flatraket.

1264 **7. Conclusions and Outlook**

1265 The results of this detailed study using mineral inclusions in 1266 garnet as provenance indicator show that by far the most garnets from HP/UHP metamorphic source rocks contain 1267 1268 mineral inclusions $\geq 2 \mu m$. To identify these tiny mineral 1269 inclusions in the entire volume of a garnet grain, Raman 1270 spectroscopy is a very powerful tool. Some minerals, however, 1271 cannot be easily specified precisely, and therefore have to be 1272 compiled into mineral groups.

Potential metamorphic source rocks for the analyzed detrital garnets can be significantly reduced by considering the conditions where garnet first appears (upper blueschist and upper greenschist facies), and the presence/absence of a few inclusion types. Especially the presence of kyanite and rutile inclusions in garnets of all chemical compositions, the absence
of other aluminosilicate inclusions, and the absence of
blueschist facies mineral inclusions (glaucophane, lawsonite)
lead to the restriction to rocks of upper amphibolite, HP
granulite, and eclogite facies as a possible source.

Moreover, the presence of eclogite facies rocks in the catchments could be verified by the presence of omphacite inclusions, and also by the presence of diopside inclusions which probably have an omphacite (jadeite) component. In the catchment of Runde, the presence of coesite inclusions in the detrital garnets further verifies an involvement of UHP metamorphic rocks, including mafic and felsic compositions.

1290 The combination of single inclusion data, coexisting mineral 1291 inclusions, and geochemical data allows a portioning of the 1292 detrital garnets into source rock groups, which goes beyond the 1293 classical geochemical classification. As a result, garnets with 1294 high probabilities for more than one host-rock group can be 1295 more confidently assigned to one of the groups, and 1296 additionally, some information can be gained about protolith 1297 composition (mafic vs. felsic) and coexisting mineral phases.

Overall, the mineral inclusions, especially in combination with geochemical host garnet composition, well reflect the geological characteristics of the sampled catchments, implying that they are useful indicators for HP/UHP provenance. Several

1302 of the most meaningful inclusion types in this study like 1303 kyanite, omphacite, diopside, enstatite, coesite, amphibole 1304 group, and epidote group minerals are either chemically and/or 1305 mechanically less stable than garnet. Thus, garnet can shield 1306 these inclusions from processes of the sedimentary cycle and 1307 preserve the mineralogical information as long as garnet is 1308 stable. This becomes more and more important when looking 1309 for HP/UHP provenance in less proximal, buried, and/or 1310 recycled sediments and sedimentary rocks. Furthermore, some 1311 provenance indicator minerals like kyanite and rutile seem to 1312 occur much more frequently as inclusions in garnet than as 1313 single grains in the heavy mineral assemblage, making 1314 inclusion analysis to a preferred tool when looking for these minerals. 1315

1316 The use of mineral inclusions in detrital garnet as a provenance 1317 tool has wide implications for prospective investigations. In 1318 particular, the contribution of material sourced from HP/UHP 1319 metamorphic rocks, which has significant geologic and 1320 geodynamic implications, can be determined more confidently 1321 than by using existing techniques. In contrast to conventional 1322 heavy mineral studies, where individual mineral grains are not 1323 in contact to each other, mineral assemblages deduced from 1324 inclusions can be directly transmitted into metamorphic mineral 1325 assemblages (minerals in mutual grain contact). Thereby, the 1326 preservation of HP/UHP index minerals from retrograde

1327 metamorphic stages and processes of the sedimentary cycle by 1328 the garnet host constitutes a major advantage. Compared to 1329 geochemical garnet single-grain analysis, where an assignment 1330 to a specific source rock is often limited to statistical 1331 probabilities, the introduced approach can provide direct 1332 mineralogical evidence for HP/UHP sources unbiased from the 1333 geochemical garnet composition. Moreover, the possibility of 1334 discriminating UHP garnets is not yet given by any other 1335 discrimination scheme.

1336 Due to the mentioned advantages and the abundance of garnet 1337 in sediments sourced from metamorphic rocks, the introduced 1338 approach is thought to become a frequently used and valuable 1339 tool in sedimentary provenance analysis. The limiting factor at 1340 present is the analytical time needed. However, in combination 1341 with geochemical discrimination schemes, analyzing the 1342 inclusions in a much smaller quantity of pre-selected grains, 1343 with compositions likely reflecting a specific source, would 1344 reduce the analytical time significantly. Further acceleration of 1345 analysis is expected by the rapid technical development of 1346 Raman spectrometry, allowing shorter acquisition times and 1347 automated measurement routines.

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1364 Appendices

1365 Supplementary Tables 1–6 can be found online as
1366 Supplementary Data at http://xxxx

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1627 **Tables**

1628	Table 1. Compilation of mineral assemblages from
1629	metamorphic rocks occurring in the Flatraket catchment
1630	(sample AK-N13-1) and its close vicinity. Mineralogical data
1631	according to ¹ Krabbendam et al. (2000), ² Wain (1997), ³ Root et
1632	al. (2004), and ⁴ Smith (1995). Abbreviations: Grt – garnet; Qz
1633	– quartz; Coe – coesite; Rt – rutile; Afs – alkali feldspar; Pl –
1634	plagioclase; Wmca – white mica; Phl-Bt – phlogopite-biotite;
1635	Omp – omphacite; Aug – augite; Opx – orthopyroxene group;
1636	Amp – amphibole group; Ky – kyanite; Ep – epidote group; Cb
1637	– carbonate minerals.

1638 Figure captions

Fig. 1. Location map and geological map of the sampling areas (modified from Krippner et al., 2016). The left side shows an overview map of the region around Faltraket and Runde (indicated by the red boxes), including an inset marking the map section (black box), location of the UHP domains according to Root et al. (2005), and peak metamorphic temperature isolines according to Kylander-Clark et al. (2008). 1646 Right side shows the geology of the two sampling areas and the1647 location of the sediment, crystalline rock, and pebble samples.

Fig. 2. Example of isolating an inclusion spectrum from the inclusion/host mixed spectrum by means of a zircon inclusion in one of the host garnets. For isolation, the garnet host spectrum captured directly next to the inclusion (blue) gets subtracted from the inclusion/host mixed spectrum (red). This results in an isolated spectrum of the inclusion (green).

Fig. 3. Examples of different inclusion spectra from the samples AK-N13-1 and AK-N37. Oxides $(a - SiO_2 \text{ polymorphs})$ quartz and coesite; b – rutile), nesosilicates (c – kyanite; d – zircon), the phosphate apatite (e), and feldspars (f – blueish spectra plagioclase and reddish spectra alkali feldspar). Bands of the host garnets are labeled with 'Grt'.

Fig. 4. Spectra of phyllosilicate inclusions giving the highest
probabilities for phlogopite (blueish spectra) and muscovite
(reddish spectra) by comparison with the RRUFF database.

Fig. 5. Spectra of clinopyroxene (a), orthopyroxene (b), and amphibole inclusions (c). Clinopyroxene spectra (a) of 9 inclusions in the analyzed garnets are shown, compared with spectra from the RUFF database (diopside: ID 040009, ID 060171; omphacite: ID 061129.2) and an omphacite in the UHP eclogite at Flatraket harbor (AK-N12).

Fig. 6. Proportion of host garnets from the stream-sediment sample of Flatraket (AK-N13-1) containing specific inclusion types (see 5.1.1) as a function of grain-size fraction (also listed in Supplementary Table 2, and more detailed in Supplementary Table 1).

1674 Fig. 7. Proportion of host garnets from the beach-sediment
1675 sample of Runde (AK-37) containing specific inclusion types
1676 (see 5.1.1) as a function of grain-size fraction (also listed in
1677 Supplementary Table 4, and more detailed in Supplementary
1678 Table 3).

1679 Fig. 8. Probabilities for the detrital host garnets from Flatraket 1680 (AK-N13-1) of belonging to the major metamorphic host-rock 1681 groups (A – eclogite facies rocks; B – amphibolite facies rocks; 1682 C – granulite facies rocks), based on geochemical composition 1683 and the calculation according to Tolosana-Delgado et al. (2017) 1684 (Supplementary Table 6). (a) - probabilities for all analyzed 1685 garnets divided in grain-size fractions (same symbols are used 1686 in (b-i)). Additionally, the probabilities for some local 1687 crystalline rocks are given, based on Krippner et al. (2016). (b-1688 i) - probabilities for garnets containing specific mineral 1689 inclusion types. Pie charts are showing the proportion of grains 1690 with the highest probability of belonging to one of the major 1691 metamorphic host-rock groups (A, B, or C) for the entire grain-1692 size fraction (63–500 µm).

1693	Fig. 9. Probabilities for the detrital host garnets from Runde
1694	(AK-N37) of belonging to the major metamorphic host-rock
1695	groups (A – eclogite facies rocks; B – amphibolite facies rocks;
1696	C – granulite facies rocks), based on geochemical composition
1697	and the calculation according to Tolosana-Delgado et al. (2017)
1698	(Supplementary Table 6). (a) – probabilities for all analyzed
1699	garnets divided in grain-size fractions (same symbols are used
1700	in (b-i)). Additionally, the probabilities for some local
1701	crystalline rocks are given, based on Krippner et al. (2016). (b-
1702	i) - probabilities for garnets containing specific mineral
1703	inclusion types. Pie charts are showing the proportion of grains
1704	with the highest probability of belonging to one of the major
1705	metamorphic host-rock groups (A, B, or C) for the entire grain-
1706	size fraction (63–500 μm).

1707 Fig. 10. Metamorphic facies diagram. (a) P-T regions of 1708 metamorphic facies types adopted from Bucher and Frey (2002), which were extended to ultrahigh-pressure (UHP) 1709 1710 conditions. Stability fields for quartz/coesite adopted from 1711 Guiraud and Powell (2006), and stability fields for 1712 aluminosilicates kyanite/sillimanite/andalusite adopted from 1713 Bohlen et al. (1991) and linearly extended (dashed line). (b) 1714 Metamorphic facies types which can be excluded as a source 1715 for the analyzed detrital garnets (grey fields: A-D). A: Facies 1716 types of the andalusite and sillimanite stability field (contact metamorphism, lowermost greenschist, lower amphibolite, and 1717

- 1718 lower to intermediate granulite facies); B: Blueschist facies
- 1719 rocks; C: Very-low-temperature eclogites (lawsonite-eclogites);
- 1720 D: Subgreenschist and lower greenschist facies rocks; E: Upper
- 1721 greenschist and lower amphibolite facies rocks.














Fig. 4



Fig. 5



Fig. 6







Fig. 8



Fig. 9





Rock types		Grt	Qz	Coe	Rt	Afs	PI	Wmca	Phl-Bt	Omp	Aug	Opx	Amp	Ky	Ep	СЬ
amphibolite facies	Micaceous Qz-Fsp-gneiss ¹	±	+	_	_	+	+	±	+	_	_	-	±	_	+	-
	Megacrystic felsic gneiss ¹	+	+	-	-	+	+	-	+	-	-	-	±	-	±	-
	Layered garnetiferous felsic gneiss ¹	+	+	-	-	+	+	±	+	-	-	-	+	-	+	-
	Layered garnetiferous felsic gneiss (mafic interlayers) ¹	+	_	_	_	+	+	_	+	_	_	_	+	_	Ι	_
granulite facies	Megacrystic felsic gneiss ¹	+	+	-	-	+	+	-	+	-	±	-	±	-	-	-
	Layered garnetiferous felsic gneiss ¹	+	+	-	-	±	+	-	+	-	±	-	-	±	-	-
	Layered garnetiferous felsic gneiss (mafic interlayers) ¹	+	_	_	_	_	+	_	_	_	+	_	_	_	Ι	_
	Mafic to intermediate dykes and pods ¹	+	±	_	-	±	+	—	±	-	+	-	-	-		-
	Dioritic to gabbroitic pods ¹	+	-	-	-	-	+	-	±	-	+	-	-	-	Ι	-
	Anorthositic pods ¹	+	±	_	-	±	+	_	±	_	+	±	-	_		-
eclogite facies	Layered garnetiferous felsic gneiss ¹	+	+	_	_	_	_	+	-	+	_	-	-	+	+	-
	Layered garnetiferous felsic gneiss (mafic interlayers) ¹	+	+	-	±	-	-	-	-	+	-	-	-	-	Ι	_
	Mafic to intermediate dykes and pods ¹	+	+	_	±	_	_	±	-	+	_	-	-	_	±	-
	Dioritic to gabbroitic pods ¹	+	+	-	-	-	-	-	+	+	-	-	-	-	+	-
	UHP eclogite AK-N12 ^{2,3}	+	+	±	+	_	_	+	-	+	_	-	+	+	+	+
	UHP eclogite Straumen ⁴	+	+	±	+	-	-	+	-	+	-	-	+	+	+	+
	•		+	alway	s prese	ent	///	/	± r	are	///	/	– n	ot pres	ent	

Table 1