#### Point-by-point response to reviews (replies marked in blue)

#### Referee #1: Carita Augustsson

The manuscript by Stutenbecker et al. with the title "Miocene basement exhumation in the Central Alps recorded by detrital garnet geochemistry in foreland basin deposits" is an Alpine provenance study based on garnet. The authors use the produced data to reveal source areas that have not been considered before, with implications for the exhumation history in the area. I have no scientific objections to the methodology, results quality or interpretations. My comments rather consider the text structure and how to improve the figures. Therefore, I estimate that only minor modifications are needed before publication in Solid Earth can be considered.

Reply: Thank you for the thorough review. We have addressed all changes to the figures as requested. Several suggestions concerned the discussion section, because it contained some repetitions from the results section. Accordingly, we restructured the discussion and eliminated the repetitions (see further below).

Below are my main comments. More detailed comments are in the manuscript file itself. I just realised that I somehow have managed to delete all my comments (not text modifications) up til Figure 3, so I have tried to reconstruct the most important ones. . .

1. Direct references to figures in the text

The authors refer directly to figures in the normal text flow. This may cause a break in the reading. Therefore, I recommend rephrasing the text such that it rather focusses on the chemical composition of the analysed garnet than on discrimination fields in specific plots, see my example directly in the text.

Reply: We have removed these direct references as suggested.

#### 2. Results in the Discussion chapter

Some results are repeated in the Discussion chapter. This is unnecessary. Here, only the data should be interpreted, not presented again. I have marked such result entries in the Discussion chapter with green directly in the manuscript.

Reply: We agree. We have restructured the discussion extensively based on suggestions by both reviewers. Instead of using the old structure "origin of amphibolite-facies garnets", "origin of eclogite-facies garnets" etc. we have created the new paragraphs "Late Oligocene (~25 My)", "Early Miocene (~19 My)" and "Middle Miocene (~14 My)". Like this, we focus the important information in a better way and avoid the repetition and the interference of results and discussion. This new discussion is structured around the new Fig. 6, which is a summary of the paleogeography of the Central Alpine hinterland and the catchment of the Napf fan.

All other comments (n = 30) are written directly in the .doc manuscript file. This includes comments on the figures and the table. If you cannot read the file, please contact me (carita.augustsson@uis.no).

Although I am not a native English speaker, I have made some linguistic suggestions. I apologise if I have introduced any grammatical errors.

Reply: All spelling mistakes, linguistic improvements and "minor" corrections (e.g. the use of "sedimentary rock" instead of "sediment", "Alpine" instead of "alpine", the use of the singular mineral/rock instead of the plural, etc.) were accepted using the track changes function directly in the revised document. Please see the revised version of the manuscript for all implemented changes (including the ones from reviewer #2 and our own corrections). For the answers to the detailed comments/suggestions, see the additional supplementary file ("Answer to Comments").

#### General reviewer comments:

In the Paper titled "Miocene basement exhumation in the central Alps recorded by detrital garnet geochemistry in foreland basin deposits" Stutenbecker et al. use a relatively new provenance tool to infer a minimum peak age of the exhumation of the External Alpine Massifs and their consequent exposure as a surface lithologies. Their major outcomes highlight the possibility that portions of the external massifs have been exhumed and eroded since ~14 Ma. This could be regarded as a potential novel find and I think that is a good starting point to speculate on the models of exhumation of the External Basement Massifs in the Alps. However, in my opinion, their work has a few new data to convince the audience that the onset of External Massifs Rocks has been driven during the mid-Miocene by high denudation coupled with crustal delamination and buoyancy-driven vertical uplift. They use this model as a key to interpreting their detrital data. This is, due to the lack of data is a bit redundantly stressed and needs to be reformulated. I, therefore, suggest the authors reworking the structure of their paper focussing in describing the previously proposed model with more objectivity with respect to their new data.

Reply: Thank you for your review. Please see the reply to the comments below.

I have tried to highlight two major points of weakness of this manuscript which I think the author might want to improve:

First the paucity of new data, the authors present results from only three samples (and additional previously published data) comparing the chemistry of the garnet with the source rocks information (3 additional samples). This is a good pilot approach but needs more constraints, possibly expanding the area of investigation to different fan deposits in the foreland to gain confidence in drawing interpretation for the onset of exhumation and erosion of the External Massifs Units. Furthermore, I find that the authors lack while interpreting/presenting their detrital datasets of a correct acknowledgment and discussion of works that focussed on the present-day evolution of detrital thermochron/petrographic proxies in the Alps. I think that would be useful to compare other proxies available in the literature with garnet chemical composition. What other analytical detrital/in-situ methods describe?

Reply: We are aware that further data from additional samples could affirm our interpretation. However, the presence of the described "exotic" garnet, even if only in one sample, proves that a source supplying those garnets must have been exposed in the hinterland. We thoroughly reviewed other provenance proxies from the study area (von Eynatten, 2003; Spiegel et al., 2000; 2003, amongst others, see Fig. 3). The problem with other proxies is that they are not unambiguous and could be interpreted in different ways, as explained in the text. I understand that the reviewer has a thermochronology background and therefore misses a more detailed discussion of the thermochronological data available. The problem with thermochronology in this setting is that we do not have bedrock data available to compare the detrital ZFT distributions (Spiegel et al. 2002) to. Because the top of the external massif has been eroded, bedrock thermochronological data are obviously not available anymore, and the oldest grains in the external massifs are around 21My (ZFT) and 10My (AFT) old. We cannot know what happened before and what kind of FT ages the (now eroded) part of the external massifs would have supplied and what that would imply in terms of exhumation rates. In contrast, garnet that is only found in specific lithologies of specific metamorphic grade provides direct evidence and

its presence is largely independent from assumptions on factors such as geothermal gradients and closing temperatures. We do not see the benefit of including modern-day thermochronological data in this context.

Second, the authors seem supporting "a priori" the model of "buoyancy-driven vertical displacement" associated with slab dynamics and erosional unloading, as a prerequisite to interpret their dataset (e.g. Herwegh et al., 2017; Nibourel et al., 2018). Those models and other proposed interpretations could, in my opinion, be described in more detail in the introduction, whereas in the discussion the authors reconcile their data with the geometric interpretation of Nibourel et al. (2018). This is an interesting ongoing discussion and might be expanded (e.g. Herman et al., 2013, Herwegh et al., 2017, Schildgen et al., 2018).

Reply: As suggested we have included a brief description of alternative models into the introduction (lines 38-42 as well as 53-57 in the revised version). The discussion on climatic effects and glacial erosion led by authors such as Herman and Schildgen concerns the late Neogene (essentially <5 My). Whether or not the late Neogene cooling had an effect on exhumation rates (and consequently erosion and sediment accumulation) is interesting, but is not directly linked to our study, because Molasse deposition ceases around 14 My ago. We do not claim that the Herwegh model is the only explanation for exhumation and erosion in the Alps and we do not exclude that climatic changes are important as well. Our study improves our understanding of the timing (onset) of exhumation rather than the process that is causing it.

I would suggest redrawing your discussion by inserting yours and available literature data in a more precise metamorphic, tectonic and erosional patterns context. The latest, in my opinion, would require a bit of discussion on how the foreland deposits might have been biased by e.g. river patterns reorganization during Miocene to present-day time, heterogeneous erosional patterns along strike, glacial processes, etc. Those processes are important for the evolution of the detrital record and need to be accounted while interpreting provenance data.

Reply: The aim of this study was to test whether or not detritus from the external massifs is present in the Molasse and whether or not this tectonic unit should be considered as a sediment source already in the Miocene. The aim of this study was not the review of the paleogeography or the drainage development during the Miocene. This would, as the reviewer points out, need further data from other locations and also other provenance proxies. We have, however, included a figure showing the paleogeography of the study area at the different time slices that show the interpreted drainage divide (new Fig. 6). We have essentially used published models for this, and have applied changes according to our new findings.

Glacial processes are not relevant in the Oligo- and Miocene.

It would be really helpful to show a compilation of different available datasets as a map view tracking External Massifs source units and their contribution in the Molasse sedimentary deposits. How does the hinterland info's are correlated with the detrital ones? A Map would greatly help the reader to track source hinterland and detrital provenance, the author cuould benefit by using their previous work e.g. Stutenbecker et al. (2017). An effort has been done in Figure. 2. However, there is not a correspondence between the legend and metamorphic grade indicated in the map. This map might be redrawn as a simplified map highlighting the information that is essential to understand the authors' discussion.

Reply: As suggested we simplified Fig. 1 and 2 to make it easier for the reader to follow the discussion. We added some important names to Fig. 1 (Lepontine dome, Prealps Romandes,

...) to guide the reader. We also added the fission track data from Bernet et al. (2009) into a new map (Figure 2b)

If I understood the reviewer's comment correctly, he asks for a map or several maps showing the evolution of the hinterland through time, so basically a paleogeographic reconstruction. We have added an interpretation of the paleogeography (new figure 6). This figure is based on previous interpretations and we have added some changes according to our new findings.

Overall, the paper reads well but there are a few changes required. I have noticed a few interferences between results description and discussion, this might be changed. The English language is good, although I might not be the best example of scrutinizer on this topic, I, therefore, suggest a native English colleague reading the manuscript once.

Reply: We have revised the discussion and restructured it to eliminate the described interferences between results and discussion. Co-author Peter Tollan is a native speaker and he has carefully reviewed this manuscript.

#### Comments by line:

25. "Tectonic processes influence" I find "influence" a bit week, maybe change with "regulate" or "drive" the evolution of mountain chains.

Reply: We replaced "influence" by "drive"

34. Please be more specific, what you mean for highest erosion rates in the Alps in (mm/yr) or as you mention in line 43 km/Myr.

Reply: Erosion rates in the Aar massif are >1mm/y as presented in Wittmann et al. (2007) and Stutenbecker et al. (2018). We modified accordingly: "...the highest denudation rates measured in the Alps (up to 1.4 mm/y), which all contribute..." (line 36 in the revised version)

61. New provenance studies that used detrital thermochronology multi-proxy approach to constrain exhumation rates and its spatial variability has been recently used in the Alps (e.g. Carrapa et al., 2016; Tectonics; Gemignani et al., 2017. Tectonics) and need to be acknowledged.

Reply: These do not concern the Molasse deposits in the Central Alps (Carrapra et al. worked on the Western Alps, and Gemignani et al. studied modern rivers). Perhaps the reviewer could be more specific on the value of these data for our interpretation.

# 72-75. Additional information to what. Does the author mean to previously published papers? Such as for instance Stutenbecker et al. (2017). Tectonic forcing of the Molasse basin or in the hinterland? Please be more specific.

Reply: No, we do not mean Stutenbecker et al. (2017), which is not a Molasse-related study. We have rephrased this sentence: "We aim (1) to explore if detrital garnet geochemistry can help identifying additional provenance changes in the Miocene Molasse deposits that have gone unnoticed so far and (2) to test whether detritus from the external massifs is present in the younger Molasse deposits in order to give independent constraints on the timing of crystalline basement exhumation." (lines85-88 in the revised version)

#### 82-84. Reference is needed

Reply: We added Allen et al., 1991 and Sinclair, 1997 (line 100 in the revised version)

105. architectural elements are capital, column, architrave, etc. Do the authors mean tectonic units or litho-tectonic units?

Reply: We replaced "architectural elements" by "tectonic units". Please note, however, that "architectural elements" is frequently used also in geological contexts (e.g. in sedimentology, Miall (1985)).

119-120: It would be useful if the author could refer to a temporal frame when invoking for timing and rates comparing it with other's colleague works. This will help the reader to follow the argumentation in chronologic order.

Reply: I do not understand this. The history of the burial and exhumation of the external massifs is reported here in chronologic order. We mention the exhumation rates already before (line 52 in the revised version).

106-142. What is the relationship of this description of the potential source rocks with the garnet composition? This is important for a clear understanding of the relationship between hinterland source units and syn-sedimentary sequences in the foreland. I think would be worth to expand this description with a map or figure showing potential source in the hinterland and their present-day distribution in the foreland units.

Reply: In this section we introduce the tectonic units to readers not familiar with the Alps. The most relevant information here for the later interpretation of the garnet chemistry is the metamorphic grade of the units, which the later interpretation relies on.

We simplified the maps in Fig. 1 and 2 to show only the primary tectonic units mentioned in this paragraph and eliminated/simplified some unnecessary details. However, I do not understand what the reviewer means by "their present-day distribution in the foreland units". The tectonic units are in the hinterland, not in the foreland... The distribution of what?

### 143. The Napf fan It is the first time that this fan is mentioned in the text. This information is missing in section 1 and should be introduced before in the text.

Reply: I am not sure I agree with this. In this paragraph we introduce the fan system properly and justify why we chose this one in particular. This is a subsection to section 1 and I do not see the need to introduce it before.

### 208. Fertility is a specific definition applied to detrital sediments. Please make sure you properly introduce this concept and acknowledge the promotors of this new definition.

Reply: This is false. Fertility is not used in <u>sediments</u> (this would be the heavy mineral concentration, see Garzanti & Andò, 2007), but it is a measure of the abundance of a specific mineral in a <u>source rock</u> (e.g. Malusà et al. 2016). We do not see the necessity of introducing this concept, as it is not relevant for the study. Instead, we refer to the review of Malusà et al., (2016) for more information.

## 213. What is the effect that you might obtain by using pestle and mortar on the round-shaped grains of garnets? There is not a less invasive mineral separation technique?

Reply: The sandstones we collected were not well cemented and disaggregated easily without applying force. We have not noticed much difference to other separation techniques. If any, the pestle + mortar technique produces less dust than a jaw breaker/ mill, indicating that it is destroying the particles less, at least in this kind of sandstone.

228-229. This might be related to an incorrect mineral separation approach and mislead to biased interpretation of the data. How could you check for consistency of the data? In other words, how fractures might bias your chemical analysis? Please explain.

Reply: Cracked grains were an exception and in >95% of grains we were able to measure the center of the grain. Fractures in garnets do not influence the chemical composition!

229. Could the authors specify the amount of "randomly selected grains"? Reply: These were 22 core-and-rim-pairs. See supplementary material for details.

246. figure 4 is confusing because the authors use black and white tones to indicate a different aspect of the different ternary plots. This could be improved by using a colored version of the figure with a color-coded legend.

Reply: We modified the figure accordingly.

272-275. Here, you are discussing the data. Please objectively describe the data. Reply: We removed this sentence.

295-297. Here, you are presenting results. Please reformulate this sentence. Reply: We removed this sentence as we restructured the discussion section.

348-354. The authors describe their data but what is lacking, in my opinion, is a clear discussion of what is the importance of those data for interpreting the evolution of the External Basement Massifs. In particular, I think that would be really interesting to insert this new preliminary finding i.d. the External Massifs Units reached the surface at ~14 Ma as constrained by Grn chemical composition, in relation with the thermokinematic model of low-temperature chronometers arguing for a sustained increase of denudation during the Pliocene. This has been the focus of a recent debate in literature see e.g. Schildgen et al. 2018 vs. Fox et al. 2015, 2016, Herman et al., 2013, etc., and I think it is important to discuss it.

Reply: We have expanded the introduction and added the suggested references to the introduction (lines 38-42 in the revised version). However, the Pliocene exhumation history is not related to our study, which is why we do not mention this in the discussion.

363-364. What is the present-day evolution of the detrital provenance/thermochronological signal? Which units constitute the present-day major erosional contributions in the Alpine river patterns? I think that might be useful for the authors to acknowledge recent studies that worked on tracking source rocks information with detrital thermochronologic evolution of modern river sands in the Alpine river patterns. There are several works that investigate these processes in a different portion of the Alps and should be, in my opinion, acknowledged (Bernet et al., 2009, Carrapa et al., 2004, Gemignani et al., 2017; Resentini et al., 2012).

Reply: We have included the modern-day bedrock ages from Bernet et al. 2009 to Figure 2 (Figure 2b) and now refer to this publication when comparing the detrital fission track ages from Spiegel et al. (2000) to potential sources. All of the other suggested references deal with thermochronology in different areas of the orogen (Western Alps, retro-foreland, axial drainage of the northern foreland basin). I do not see how this would help our interpretation. Perhaps the reviewer could be more specific on the value of these data for our interpretation.

365. "Very young", how young <2Ma, <5 Ma, <10 Ma, <30 Ma?

Reply: According to Spiegel et al. (2000) the youngest age peak in Molasse of this age is  $19.5\pm0.9$  My. We added this information to lines 195 and 415 in the revised version.

370-393. At this point, it is clear that the compositional change of the garnets in the youngest ~14 Ma foreland deposits with respect to the older ~19 Ma interval (where Grn yield a different composition = different provenance) has been interpreted by the authors as the lower temporal limit for the surficial exposure of the External Basement Massifs units. Using this new observation they argue for "important implication for the tectonic evolution of the orogen" (Lines 375-376). Furthermore, the authors support the geometric restoration of the central Alps (Aar Massif-Helvetic nappes) as proposed by Nibourel et al., 2018, where ~7-8 km of basement rocks have been exhumed and eroded since ~14 Ma lead by "lithospheric mantle roll back" associated with "crustal delamination" and "buoyancy-driven vertical exhumation coupled with surface erosion" of the External Basement Massifs (e.g. Herwegh et al., 2017). This point in the discussion is clear and well expressed, however, I think that you should describe also the other proposed model in the introduction, and, lately, data on hands, describe why your data support this proposed hypothesis. This is, in my opinion, a bit lacking in the text and would require some improvements

Reply: As already pointed out in the second comment, we have added the alternative exhumation models in the introduction (lines 38-42 in the revised version). Our results do not improve our understanding of the <u>process</u> of exhumation, but that of its <u>timing</u>. As pointed out before, we cannot reconstruct the exhumation rates, because we lack bedrock data of what has already been eroded, and we can therefore not say if exhumation was faster or slower in the Miocene.

### List of all relevant changes made to the manuscript

- Text:
  - Introduction: We have added some relevant references that concern the ongoing discussion on the exhumation mechanism in the study area (mantle processes vs. erosional unloading and related climatic triggers) as requested by by referee #2.
  - Discussion: We have reorganized the discussion section extensively. Both referees pointed out that we had some interference between the results and discussion sections and that some points were repeated. Before, the subsections were describing the origin of individual garnet groups ("origin of eclogite facies garnets", "origin of amphibolite facies garnets" etc.) and then there was a final sub-section showing the implications to the timing of exhumation. Now, we have reduced the subsections to three that are organized around the new Fig. 6 (showing the paleogeography, see below) and discuss the implications of our results to the respective time slices at 25 My, 19 My and 14 My ago.
- Tables:
  - We have extended Table 3 to show information that was previously contained in Fig. 6 (now deleted, see below)
- Figures:
  - We have simplified Fig. 1 by combining some tectonic units, adding black lines to their outlines and we added the names of some important tectonic units that we mention in the text
  - We have simplified Fig. 2 (now Fig. 2a) by removing some unnecessary details. We used an easier color scheme and added the outlines of tectonic units (in Fig. 1) to facilitate the comparison with Fig. 1.
  - We have added a new Fig (Fig. 2b) showing the zircon fission track ages of bedrock exposed in the Central Alps (as suggested by referee #2)
  - We have changed Fig. 4 from greyscales to a colored version (as suggested by referee #2)
  - We have deleted the former Fig. 6 and instead combined the information contained in that figure into Table 3 (as suggested by referee #1)
  - We have prepared a new Fig. 6 showing a reconstruction of the paleogeography and the source areas present at each time slice (as suggested by referee #2)

1	Marked-up manuscript version
2	
3	Miocene basement exhumation in the Central Alps recorded
4	by detrital garnet geochemistry in foreland basin deposits
5	Laura Stutenbecker <sup>1</sup> *, Peter M.E. Tollan <sup>2</sup> , Andrea Madella <sup>3</sup> , Pierre Lanari <sup>2</sup>
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#### 14 <u>Abstract</u>

15 The Neogene evolution of the European Alps was characterized by the exhumation of crystalline 16 basement, the so-called external crystalline massifs. Their exhumation presumably controlled the 17 evolution of relief, distribution of drainage networks and generation of sediment in the Central Alps. 18 However, due to the absence of suitable proxies, the timing of their surficial exposure, and thus the

19 initiation of sediment supply from these areas, are poorly constrained.

20 The northern <u>Aalpine foreland basin preserves the Oligocene to Miocene sedimentary record of</u> 21 tectonic and climatic adjustments in the hinterland. This contribution analyses the provenance of 25 to

22 14 My-old alluvial fan deposits by means of detrital garnet chemistry. Unusually grossular- and

spessartine-rich garnets is are found to be(1) to be a unique proxyies for identifying detritus from the external crystalline massifs and (2) to occur abundantly in ca. 14 My-old deposits of the. In the

external crystalline massifs and (2) to occur abundantly in ca. 14 My-old deposits of the. In the foreland basin, these this garnets are is abundant in 14 My-old deposits. In contrast to previous

assumptions, we therefore propose that the external massifs were already exposed to the surface ca. 14

My ago., thus providing a minimum age for the surficial exposure of the crystalline basement.

#### 28 1. <u>Introduction</u>

29 Tectonic processes influence drive the evolution of relief in mountain chains and consequently control 30 the development of the drainage network, sediment supply and deposition in the foreland basin. The 31 Central European Alps and their northern foreland basin, formed through the collision of the European 32 and the Adriatic continents since the Eocene (Schmid et al. 1996, Handy et al. 2010), are a classic 33 example of such interactions (e.g. Schlunegger et al., 1998; Pfiffner et al., 2002; Vernon et al., 2008, 34 2009; Baran et al., 2014; Fox et al., 2015). The exhumation of large slices of mid-crustal rocks from 35 the European plate, the so-called external crystalline massifs, occurred during a late stage orogenie 36 eventrelatively late in the Alpine evolution, probably during the late Miocene, although the exact 37 timing is not well constrained. The external crystalline massifs are today characterized by high relief, 38 intense glaciation and some of the highest denudation rates measured-in the Alps (up to 1.4 mm/y), 39 which all contribute to their importance relevance as a sediment source (Kühni and Pfiffner, 2001; Wittmann et al., 2007; Stutenbecker et al., 2018). The exhumation is discussed to be related to possibly 40 41 controlled by crustal delamination in response to lithospheric mantle rollback (Herwegh et al., 2017), slab detachment (Fox et al., 2015) or erosional unloading (Champagnac et al., 2009), possibly due to 42 increased precipitation rates in the Pliocene (Cederborn et al., 2004) or enhanced glacial erosion in the 43 44 Pleistocene (Fox et al., 2015; Herman et al., 2013). The areas exhumed during this event are today 45 characterized by high relief, intense glaciation and some of the highest denudation rates measured in 46 the Alps (up to 1.4 mm/y), which all contribute to their importance as a sediment source (Kühni and

47 Pfiffner, 2001; Wittmann et al., 2007; Stutenbecker et al., 2018).

48 Peak metamorphism of lower to upper greenschist -facies conditions occurred between 17 and 22 Ma 49 in all northern external crystalline massifs (Mont Blanc, Aar massifs and the Gotthard nappe, 50 Challandes et al., 2008; Rolland et al., 2008; Cenki-Tok et al., 2014; Nibourel et al., 2018). Their 51 subsequent exhumation has been investigated using thermochronology by a number of studies (e.g. 52 Schaer et al., 1975, Wagner et al., 1977; Michalski and Soom, 1990; Vernon et al., 2009; Glotzbach et 53 al., 2010). While-Whereas some studies concluded that exhumation was episodic (e.g. Vernon et al., 54 2009), others suggest relatively constant exhumation rates of 0.5-0.7 km/My since 14 My (Michalski and Soom, 1990; Glotzbach et al., 2010). A focus in this debate concerns the late Neogene cooling and 55 the onset of glaciation in the Pleistocene and their possible effect on the exhumation, erosion and 56 sediment accumulation rates (e.g. Kuhlemann et al., 2002; Herman et al., 2013; Schildgen et al., 57 58 2017). In contrast, the Paleogene and early Neogene exhumation history isreceived comparably little 59 attention-less well studied. In particular, Tthe timing of the first surficial exposure of the external massifs has, however, for example, has never been constrained, because estimates of their total 60 thickness have not yet been established yet. In most geometric reconstructions (e.g. Pfiffner, 1986; 61 62 Pfiffner, 2017; Schmid et al., 2004), the contact between the crystalline basement and the overlying 63 Mesozoic cover is assumed to be relatively flat, and the top of the crystalline basement is hypothesized 64 to have been less than one kilometer above the modern topography. Conversely, a new reconstruction of this tectonic contact allows for a substantially greater amount (~8 km) of (now eroded) crystalline 65 rock on top of the present-day topography (Nibourel et al., 2018). 66

This study aims to constrain the timing of exposure, and thus the beginning of sediment supply from 67 the external crystalline massifs, by determining the provenance of the foreland basin deposits. 68 69 Sedimentary rockss preserved in the northern peripheral foreland basin of the Central Alps, the Swiss 70 part of the Molasse basin, are a well-studied archive recording tectonic and climatic adjustments in the 71 central orogen between ca. 32 and 14 My ago (Schlunegger et al., 1993, 1996; Kempf et al., 1999; 72 Spiegel et al., 2000; Kuhlemann and Kempf, 2002; von Eynatten, 2003; Schlunegger and Kissling, 73 2015). So far, the provenance of the Molasse deposits has been investigated using optical heavy 74 mineral analysis, framework petrography and both bulk and single-grain geochemical techniques, including epidote geochemistry and cooling ages derived from zircon fission track analysis and Ar\_/Ar
dating of white mica (Spiegel et al., 2000, 2002; von Eynatten, 2003; von Eynatten and Wijbrans,
2003). No cConclusive evidence for a contribution from the external crystalline massifs, however, has
remained-been found thus farelusive, leading to the assumption that their exposure must post-date the
youngest preserved (ca. 14 My-old) Molasse sediments-deposits (von Eynatten, 2003).

In this study, we use detrital garnet major element geochemistry of detrital garnet in Miocene deposits 80 81 preserved infrom the central part of the Swiss foreland basin. The great compositional variability displayed by garnet from different source rocks means that it is a useful provenance tracer in a variety 82 of settings (Spear, 1994; Mange and Morton, 2007). Furthermore, it is a common heavy mineral in 83 84 (orogenic) sediments and sedimentary rocks (Garzanti and Andò, 2007) and is relatively stable during 85 transport and diagenesis (Morton and Hallsworth, 2007). In the Central Alps, detrital garnet has 86 recently been shown to be a valuable provenance indicator, especially for distinguishing detritus supplied from the external crystalline massifs (Stutenbecker et al., 2017). We aim (1) to explore if 87 detrital garnet geochemistry can help identifying additional provenance changes in the Miocene 88 Molasse deposits that have gone unnoticed so far provide additional provenance information to 89 unravel the Miocene history of the Molasse deposits and its tectonic forcing and (2) to test whether 90 detritus from the external massifs is present in the younger Molasse deposits in order to give 91

92 independent constraints on the timing of crystalline basement exhumation.

#### 93 **1.1 Geological Setting**

94 The Central Alps evolved through convergence between the European continental margin in the north 95 and the Adriatic plate in the south (Schmid et al., 1996). The cConvergence started in-during the late 96 Cretaceous with the subduction of the Aalpine Tethys Oocean below the Adriatic microplate (Froitzheim et al., 1996), and ceased in-during the Paleogene after the European continental 97 98 lithosphere entered the subduction zone. These Cretaceous to early Neogene orogenic processes are 99 reflected by the syn-orogenic deposition of deep-marine flysch units preserved throughout the Alps 100 (see e.g. Wildi, 1985; Winkler, 1996). Around 32 Ma ago, the sedimentation style in the northern 101 foreland basin changed from marine, flysch-like deposition to shallow marine and terrestrial 102 sedimentation (Allen et al., 1991; Sinclair, 1997). This is thought to represent the transition to 103 Molasse-type sedimentation in an overfilled basin and is discussed to be potentiasly related to a 104 breakoff of the European slab around the time of the Eocene-Oligocene boundary (e.g. Sinclair et al., 105 1991; Sinclair, 1997; Schlunegger and Kissling, 2015). Since this time, the northern foreland basin has 106 become a major sink of orogenic detritus and an important sedimentary archive.

107 The sedimentary rockss in the Swiss part of the northern foreland basin are divided into four litho-108 stratigraphic units that represent two shallowing- and coarsening-up megacycles (Schlunegger et al., 109 1998). The first cycle consists of the Rupelian Lower Marine Molasse (LMM) and the Chattian and 110 Aquitanian Lower Freshwater Molasse (LFM). The second megacycle comprises a transgressive facies 111 of Burdigalian age (the Upper Marine Molasse, UMM) overlain by Langhian to Serravalian deposits 112 of the Upper Freshwater Molasse (UFM). The depositional ages of these units were constrained using 113 mammal biostratigraphy and magnetostratigraphy (Engesser, 1990; Schlunegger et al., 1996). 114 Throughout the Oligocene and the Miocene, the proximal Molasse deposits are thought to have been 115 formed through a series of large alluvial fans (Fig. 1) aligned along the Aalpine thrust front 116 (Schlunegger et al., 1993; Kuhlemann and Kempf, 2002). The more distal parts of the basin were 117 instead characterized by axial drainage directed towards the Paratethys in the Easteast/Northeast 118 northeast (31-20 My) and the Western Western Mediterranean Sea in the Southwest southwest (after 119 20 My), respectively (Kuhlemann and Kempf, 2002). Whereas the more distal deposits could be

120 significantly influenced by long-distance transport from the northeast or southwest, the alluvial fans

- are thought to carry a local provenance signal from the rocks exposed immediately south of each fansystem due to their proximal nature.
- 123 The hinterland of the central Swiss foreland basin comprises, from north to south, potential source124 rocks derived from the following architectural elements tectonic units (Figs. 1, 2):
- (1) The Prealps Romandes; a stack of non-metamorphic and weakly metamorphosed sedimentary cover nappes (Mesozoic carbonates and Cretaceous-Eocene flysch), interpreted as the accretionary wedge of the <u>A</u>alpine Tethys, detached from its basement and thrusted northwards onto the European units.
  - (2) The Helvetic nappes; the non- or very low-grade metamorphic sedimentary cover sequence of the European continental margin (mostly Mesozoic carbonates).
- 131 (3) The external crystalline massifs; lentoid-shaped autochthonous bodies of European continental crust that consist of a-pre-Variscan polycyclic gneiss basement intruded by Upper 132 Carboniferous to Permian granitoid rocks and an overlying metasedimentary cover. They were 133 buried within the Alpine nappe stack in-during the Oligocene (Cenki-Tok et al., 2014), 134 135 reaching greenschist facies peak-metamorphic conditions between 17 and 22 My ago (Fig. 2a) and were exhumed during the Miocene. The Gotthard nappe, although not a "massif" sensu 136 137 stricto because of its allochthonous nature, will be included into the term "external crystalline massifs" from here on, because the timing and the rates of exhumation are comparable (Fig. 138 **2b.** Glotzbach et al., 2010). 139
- (4) The Lepontine dome; an allochthonous nappe stack of European Paleozoic gneiss basement and its Mesozoic metasedimentary cover (Berger et al., 2005). Amphibolite\_-facies peak metamorphism (Frey and Ferreiro Mählmann 1999;; Fig. 2a) in the Lepontine occurred diachronously at around 30-27 My ago in the south (Gebauer, 1999) and possibly as late as 19 My ago in the north (Janots et al., 2009). Although the onset of exhumation of the Lepontine dome might have been equally diachronous, it is generally assumed to have occurred before 23 My ago (Hurford, 1986).
  - (5) The Penninic nappes, containing ophiolites of the <u>Aa</u>lpine Tethys as well as the continental crust of Briançonnais, a microcontinent located within the <u>aA</u>lpine Tethys between the southern Piedmont-Ligurian ocean and the northern Valais trough (Schmid et al., 2004).
- (6) The Austroalpine nappes, containing the basement and sedimentary cover of the Adriatic plate
  with a Cretaceous ("Eoalpine", ca. 90-110 My) metamorphic peak of greenschist facies
  conditions (Schmid et al., 2004). Although the The Austroalpine nappes were probably part of
  the nappe stack in the Central Alps prior to their erosion during the Oligocene and Miocene
  although they are found exclusively in the Eastern Alps to the east of the Lepontine dome
  today., we mention them here as well, because they were probably part of the nappe stack in
  the Central Alps prior to their erosion during the Oligocene and Miocene.
- (7) The Sesia/Dent Blanche nappe, probably representing rifted segments of the basement and sedimentary cover of a distal part of the Adriatic plate (Froitzheim et al., 1996). In contrast to the Austroalpine nappes, the Sesia/Dent Blanche nappe was subducted and exposed to blueschist\_facies (Fig. 2a; Bousquet et al., 2012, Fig. 2) to eclogite\_facies metamorphism (e.g. Oberhänsli et al., 2004).
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#### 163 **1.2 Compositional trends in the <u>Honegg-</u>Napf fan**

Rocks from the <u>The</u> Central Alps are generally considered as the major sediment source of all proximal
 Molasse basin deposits, <u>while and</u> compositional changes in the foreland are thought to directly reflect
 tectonic and erosional processes in the immediate <u>A</u>alpine hinterland (Matter, 1964; Schlunegger et

al., 1993; 1998). The compositional evolution in the basin is diachronous and nont-uniform between
the different fan systems (e.g. Schlunegger et al., 1998; Spiegel et al., 2000; von Eynatten, 2003). In
this study, we will focus on the <u>Honegg-Napf</u> fan, located in the central part of the basin. <u>, It which is</u>
the most likely to archivepreserves a provenance signal related to external massif exhumation due to
its proximity to the large crystalline basement slices of the Aar massif and the Gotthard nappe (Fig. 1).
In the Honegg-Napf fan, three major compositional trends have been previously identified (Fig. 3):

(Phase 1) Between ~31 and ~25 My ago, the heavy minerals are dominated by the zircon-tourmalinerutile (ZTR)-assemblage and garnet (von Eynatten, 2003). Rock fragments are dominantly of
sedimentary origin and zircon fission track ages are Paleozoic to late Mesozoic (Spiegel et al., 2000).
This phase is consistently interpreted by different authors to reflect the erosion of (Austroalpine)
flysch-like sedimentary cover nappes, which are structurally the highest-top in of the central aAlpine
nappe stack and probably extended further west during this time (Schlunegger et al., 1998; Spiegel et al., 2000).

(Phase 2) 25-21 My ago: Around 25 My ago, the occurrence of epidote as well as an increase in 180 181 granitic granitoid rock fragments mark a major compositional change in the foreland. The presence of 182 characteristic colorful granite pebbles suggests an origin from the Austroalpine Bernina nappe (Matter, 183 1964). Sediments of this phase clearly reflect the down-cutting down into crystalline basement and are 184 consistent with a continuation of a normal unroofing sequence. Additionally, (Schlunegger et al., (1998) report the occurrence of quartzite pebbles, possibly sourced from the middle Penninic Siviez-185 186 Mischabel nappe and argue that parts of the epidote could originate from Penninic ophiolites as well, 187 thus suggesting that erosion might have already reached down into the Penninic nappes already by 188 then. Spiegel et al., (2002) argued against this Penninic contribution based on the  ${}^{87}$ Sr/ ${}^{86}$ Sr and <sup>143</sup>Nd/<sup>144</sup>Nd isotopic signatures of the epidote. 189

190 (Phase 3) 21-14 My ago: At ~21 My, metamorphic rock fragments occur in the sediments, while 191 whereas the heavy mineral assemblages remain epidote-dominated and overall similar to the second 192 phase. Zircon fission track ages are exclusively Cenozoic (ages peaks between ~32 and ~19 Ma). In 193 contrast to the first two phases, the sediment composition allows several, partially contradicting 194 interpretations. Whilst petrographical and mineralogical data might suggest recycling and sediment 195 mixing (von Eynatten, 2003), young <sup>40</sup>Ar/<sup>39</sup>Ar cooling ages in white mica (von Eynatten, 2003; von 196 Eynatten and Wijbrans, 2003) and exclusively young a population of zircons with a fission track 197 central ages of 19.5±0.9 My -(Spiegel et al., 2000) point to an additional, newly exhumed source that 198 these authors-identified as the Lepontine dome (Fig. 2b; von Eynatten, 2003; Spiegel et al., 2000). Based on the abundance of flysch pebbles after ~21 My, Schlunegger et al. (1998) favor an alternative 199 200 scenario, in which the erosional front shifted northwards into the flysch nappes of the Prealps 201 Romandes. A mixture of both sources seems possible. Furthermore, the isotopic signature of detrital 202 epidotes suggests a contribution of mantle source rocks between ca. 21 and 19 My ago, which could 203 point to a contribution by Penninic ophiolites (Spiegel et al., 2002). However, this is not reflected in 204 the heavy mineral spectra (von Eynatten, 2003) that, which do not contain typical ophiolite minerals 205 such as Cr-spinel.

In none of these scenarios were tThe external crystalline massifs <u>have not been</u> considered as a possible sediment source. The exact time of their surficial exposure is unknown, but it is believed to post-date the youngest preserved Molasse <u>sedimentsdeposits</u>. This interpretation is based on the lack of granitic pebbles attributable to the external massifs in the Molasse (Trümpy, 1980) and on structural reconstructions (e.g. Pfiffner, 1986) in combination with thermochronological data (e.g. Michalski and Soom, 1990).

#### 212 2. <u>Sampling strategy and methodology</u>

- 213 In order to characterize the detrital garnets in the foreland, three samples were taken from 25 My-, 19
- My- and 14 My-old fine- to medium-grained fluvial sandstones within the <u>Honegg-Napf</u> fan deposits
- located ca. 40 kilometers to the East-east and Southeast southeast of Berne in the central part of the
- 216 Swiss Molasse basin. The exact sampling sites were chosen based on the availability of published
- 217 petrographical, chemical and mineralogical data (von Eynatten, 2003) as well as magnetostratigraphic
- 218 calibration (Schlunegger et al., 1996).
- 219 <u>It is possible to compare potential source compositions to the detrital ones, b</u>Because the potential
- source rocks were already narrowed down to particular regions based on other provenance proxies,
- and because many of these rocks are still preserved in the <u>A</u>alpine chain today, it is possible to
- 222 compare potential source compositions to the detrital ones. For comparison we used detrital data from
- Stutenbecker et al. (2017) as well as published source rock data from different units across the Central
  Alps (Steck and Burri, 1971; Chinner and Dixon, 1973; Ernst and Dal Piaz, 1978; Hunziker and
  Zingg, 1980; Oberhänsli, 1980; Sartori, 1990; Thélin et al., 1990; Reinecke, 1998; von Raumer et al.,
- 226 1999; Cartwright and Barnicoat, 2002; Bucher and Bousquet, 2007; Angiboust et al., 2009; Bucher
- and Grapes, 2009; Weber and Bucher, 2015).
- 228 In addition, three river sand samples were collected from small monolithological catchments (3-30 229 km<sup>2</sup>) draining potentially garnet-bearing potential source rocks that were previously not, or only 230 partially, considered in the literature. We prefer this "tributary sampling approach" (first-order 231 sampling scale according to see e.g. Stutenbecker et al., 2017Ingersoll, 1990) over in-situ sampling of 232 specific source rocks, because small monolithological catchments are more likely to comprise all 233 garnet varieties of the targeted source rock and to average out spatial variations of the source rock 234 properties, e.g. mineral size or fertility (Malusà et al., 2016). differences in garnet fertility. The 235 targeted plausible source areas are located within in the Gurnigel flysch (Prealpes Romandes), the Antigorio nappe orthogneisses of the Lepontine dome, and the Lebendun nappe paragneisses of the 236 237 Lepontine dome (Fig.1). Sample characteristics are summarized in Table 1 and Table 2. For detailed 238 lithological descriptions of the sampled sampling sites in the Honegg-Napf area, see Schlunegger et al.
- **239** (1993) and von Eynatten (2003).
- The sandstone samples were carefully disintegrated using a jaw breaker and a pestle and mortar. The disintegrated sandstones as well as and the source rock tributary sands were sieved into four grain size classes of <63  $\mu$ m, 63-125  $\mu$ m, 125-250  $\mu$ m and >250  $\mu$ m. The fractions of 63-125  $\mu$ m and 125-250  $\mu$ m were further processed in sodium polytungstate heavy liquid at 2.85 g/cm<sup>3</sup> to concentrate heavy minerals. The heavy mineral concentrates were dried and, depending on the obtained amounts, split into 2-4 parts using a microsplitter. All measured-analysed garnet grains were hand-picked from the concentrate of one split part per fraction under a binocular microscope.
- 247 The grains were subsequently arranged in lines on sticky tape, embedded into in epoxy resin, ground with SiC abrasive paper (grits 400, 800, 1200, 2500, 4000), polished using 3, 1 and 1/4 µm diamond 248 249 suspensions and graphite-coated. Major element oxides were analyzed using a JEOL JXA-8200 250 electron probe micro-analyzer at the Institute of Geological Science at University of Bern, Switzerland, under standard operating conditions for garnet (see Giuntoli et al., 2018): accelerating 251 252 voltage of 15 kKeV, electron beam current of 15 nA, beam diameter of 1 $\mu$ m, 20 s peak acquisition 253 time for Si, Ti, Al, Fe, Mn, Mg, Ca and 10 s for both backgrounds. Natural and synthetic standard olivine (SiO<sub>2</sub>, MgO, FeO), anorthite (Al<sub>2</sub>O<sub>3</sub>, CaO) ilmenite (TiO<sub>2</sub>) and tephroite (MnO) were used for 254 calibration by applying a CITIZAF correction (Armstrong, 1984). Garnet compositions were measured 255 256 as close as possible to the geometric centers of the grains, unless the area was heavily fractured or 257 showed inclusions of other minerals. In some randomly selected grains core and rim compositions

- were measured to identify intra-grain chemical variability; these core/rim pairs are reported separately in Stutenbecker (2019).
- 260 Molecular proportions were calculated from the measured main oxide compositions on the base of 12 anhydrous oxygen <u>atoms</u>. Because ferric and ferrous iron were not measured separately (FeO = Fe<sub>total</sub>), 261 Tthe Fe<sup>2+/</sup>Fe<sup>3+</sup> ratio was determined based on charge balance (Locock, 2008), because ferric and 262 ferrous iron were not measured separately (FeO =  $Fe_{total}$ ). Garnet endmember compositions were 263 264 subsequently calculated using the Excel spreadsheet by Locock (2008). Garnet is a solid solution 265 between different endmembers, the most common ones being almandine (Fe<sub>3</sub>Al<sub>2</sub>Si<sub>3</sub>O<sub>12</sub>), grossular (Ca<sub>3</sub>Al<sub>2</sub>Si<sub>3</sub>O<sub>12</sub>), pyrope (Mg<sub>3</sub>Al<sub>2</sub>Si<sub>3</sub>O<sub>12</sub>), spessartine (Mn<sub>3</sub>Al<sub>2</sub>Si<sub>3</sub>O<sub>12</sub>) and andradite (Ca<sub>3</sub>Fe<sub>2</sub>Si<sub>3</sub>O<sub>12</sub>). The 266 267 relative proportions of these endmember components almandine (Fe<sub>3</sub>Al<sub>2</sub>Si<sub>3</sub>O<sub>12</sub>), grossular 268  $(Ca_3Al_2Si_3O_{12})$ , pyrope  $(Mg_3Al_2Si_3O_{12})$ , spessartine  $(Mn_3Al_2Si_3O_{12})$  and andradite  $(Ca_3Fe_2Si_3O_{12})$ depend on bulk rock composition and intensive parameters (such as temperature and pressure), which 269 can vary substantially depending on the metamorphic or magmatic history of the protolith (Deer et al., 270 271 1992; Spear, 1994). The data were plotted and classified using the ternary diagram of Mange and 272 Morton (2007) as well as the linear discriminant function method of Tolosana-Delgado et al. (2018) based on a global data compilation on garnet compositions from different source rocks (Krippner et 273 274 al., 2014).

#### 275 3. <u>Results</u>

276 Most of the detrital garnets are dominated by the Fe-rich almandine endmember with varying amounts 277 of grossular, pyrope, spessartine and andradite (Fig. 4). Other endmembers (e.g. uvarovite) are 278 negligible. Minimum, maximum and average Average endmember contents are summarized in Table 3; 279 for the full dataset we refer to Stutenbecker (2019). Garnet compositions do not differ significantly 280 between the two analyzed grain size fractions of the same sample, although some-slight variations are 281 visible in the ternary plot (Fig. 4): iIn sample LS2016-18 (25 My, Fig. 4a) garnets of the 125-250 µm fraction tend-isto be more enriched in pyrope with respect to than garnets of the 63-125  $\mu$ m fraction. In 282 sample LS2018-5 (19 My, Fig. 4b) 4 "outliers" that are very pyrope- and grossular-rich (n=2) or 283 284 grossular- and andradite-rich (n=2) occur only in the 63-125 µm grain size fraction. Furthermore, 285 garnet grains of the 63-125 µm fraction are more frequently grossular-rich compared to the 125-250 µm fraction. In sample LS2017-3 (14 My, Fig. 4c), the 63-125 µm fraction contains some garnet 286 287 grains (n=8) of high almandine and low grossular content that are absent in the  $125-250 \mu m$  fraction. 288

Although some individual garnet grainss show distinct internal compositional zoning from core to rim,
 the intra-grain chemical variability is generally negligible (see Stutenbecker, 2019).

291 According to the ternary classification plot of Mange and Morton (2007), tThe major part of garnet in all three samples (>80 %) belong to the B-type garnet of Mange and Morton (2007) and thus point to a 292 293 dominant contribution by amphibolite -facies source rocks (Table 4). Minor portions amounts are 294 derived from classified as C-type (high-grade metabasic), A-type (granulite facies) and D-type 295 (metasomatic) sourcesgarnet. The 25 My-old sandstone contains almost exclusively B-type garnet 296 (92%, Table 4). The 19 My-old sandstone shows a larger spread with some A-, C- and D-type garnet 297 (Fig. 4b, Table 4). The 14 My-old sandstone contains B-, C- and D-type garnet (Fig. 4c, Table 4). 298 Classification through linear discriminant analysis (Tolosana-Delgado et al., 2018) yields a similar 299 trend with generally high proportions of amphibolite -facies source rocks (class B-garnets, >70 %, 300 Table 4). Some grains (5 %, 3 % and 12 % in the 25 My-, 19 My- and 14 My-old samplesdeposits,

301 respectively) were classified as igneous garnet (Table 4).

302 Distinct compositional changes between the 25 My-, 19 My- and 14 My-old Molasse sediments 303 sandstones are mostly related to the ratio of almandine and grossular contents (Table 3, Fig. 5). At 25 304 My, the garnets are dominantly almandine-rich (average 70 %) and grossular-poor (average 9 %). At 305 19 My, both grossular-poor and grossular-richer garnets occur (average 16 %). Garnets in the 14 My-306 old sample-sandstone are generally almandine-poorer (average 50 %) and grossular-rich (average 32 307 %). This implies (1) that garnets contained in the younger sediment (14 and, to some extent, 19 My) 308 were not recycled in significant amounts from the older Molasse strata and (2) that at least two sources 309 supplied B-type garnets during Molasse deposition.

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Garnet compositions from the three potential source rock samples analyzed in this study are shown in

Fig. 4d (Lepontine paragneiss and Lepontine orthogneiss) and Fig. 4e (Gurnigel flysch). The average
 compositions are displayed in Fig. 6; for the full dataset we refer to Stutenbecker (2019). Likewise,
 average compositions of garnet from the literature (external massif granite garnets, eclogite facies
 garnets and granulite facies garnets) are displayed in Fig. 6.

316

All source rocks, except for the external crystalline massif granites, supply almandine dominated (i.e.

318 >50 % almandine component) garnet. The andradite content in all source rock garnets is very low, but 319 they contain varying amounts of grossular, spessartine and pyrope. Garnets from the Lepontine 320 gneisses (Table 3, Fig. 4d) are generally almandine-rich, but those in the paragneiss tend to be

grossular-richer (22 %) compared to the ones in the orthogneiss (11 %). The Gurnigel flysch garnets
(Fig. 4e) are almandine-rich with elevated pyrope contents (14 %). Garnets from the external

323 crystalline massifs (Fig. 4f) are unusually rich in grossular (35 %) and spessartine (21 %), and the
 324 almandine content is much lower than in the other source rock garnets (34 %). Eclogite facies garnets

325 have high grossular (23 %) and pyrope (16 %) contents (Fig. 4g). Granulite facies garnets (Fig. 4g)

326 have on average the highest pyrope content of all source rock garnets (25 %).

#### 327 4. <u>Discussion</u>

328 4.1 Origin of amphibolite-facies garnetsLate Oligocene (~25 My ago)

Although detrital garnet chemistry suggests the presence of only one relatively uniform, amphibolite – facies source rock in the hinterland of the Honegg-Napf fan during the late Oligocene, the identification of the exact nature of this source is difficult. This is mostly due to the large compositional overlap of garnet sourced by diverse amphibolite -facies metamorphic rocks (e.g. metasedimentary versus meta-igneous; Krippner et al., 2014; Tolosana-Delgado et al., 2018).

Amphibolite -facies conditions of Alpine age were only reached in the Lepontine dome (Fig. 2a; 334 335 Bousquet et al., 2012). However, many gneisses in the Central Alps preserve a pre-Alpine amphibolite -facies metamorphic signature as well (Frey et al., 1999), for example in the Austroalpine Bernina 336 337 nappe (Spillmann, 1993; Spillmann and Büchi, 1993), the middle Penninic Briançonnais basement 338 (Sartori et al., 2006) or the polycyclic basement of the external massifs (von Raumer et al., 1999). In 339 fact, the Gurnigel flysch, a Late Cretaceous to Eocene flysch nappe in the Prealps Romandes that did 340 not undergo Alpine metamorphism (Fig. 2a), contains abundant almandine-rich B-type garnets (Fig. 341 4e). 342 Zircon fission track ages from sandstones of the same age are mostly >100 My old with a smaller and 343 younger age peak of 41±9 My (Fig. 3; Spiegel et al., 2000). This would favor an input from the

Austroalpine nappes and/or the Prealps Romandes (Fig. 67a), which yield related cooling ages >50 My

345 (Fig. 2b; e.g. Bernet et al., 2009), rather than from the Lepontine dome, which is characterized by
 346 zircon fission track ages <30 My (Fig. 2b; e.g. Hurford, 1986). The presence of granite pebbles</li>

- attributable to the Austroalpine Bernina nappe (Matter, 1964; Schlunegger et al., 1998) would further
   support an Austroalpine rather than a Lepontine provenance.
- 349 The drainage divide was probably located close to the Insubric line (e.g. Schlunegger et al., 1998), but
- north of the Bergell pluton (Fig. 67a), whose detritus is exclusively found in the retroforeland to the
- south (Gonfolite Lombarda;, Giger and Hurford, 1989; Carrapa and Di Giulio, 2001).
- According to the compositional classification of Mange and Morton (2007) and Tolosana Delgado et
   al. (2018), the majority of detrital garnet grains in the Molasse were derived from amphibolite facies
- 354 source rocks ("B type"). Garnets derived from amphibolite facies rocks ("B type") seem to be the
- 355 <u>most frequent ones in all three considered samples.</u> In the Central Alps, amphibolite facies conditions
- 356 of alpine age were only reached in the Lepontine nappes (Fig. 2). However, many gneisses in the area
- 357 preserve a pre Mesozoic amphibolite facies metamorphic signature as well (Frey et al., 1999), for
- example in the Austroalpine Bernina nappe (Spillmann, 1993; Spillmann and Büchi, 1993), the middle
   Penninic Briançonnais basement (Sartori et al., 2006) or the polycyclic basement of the external
- 360 massifs (von Raumer et al., 1999). In fact, the Gurnigel flysch, a Late Cretaceous to Eocene flysch
- 361 nappe in the Prealps Romandes that did not undergo alpine metamorphism (Fig. 2), contains almost
- 362 exclusively almandine-rich B-type garnets (Fig. 4e).
- 363 These considerations indicate that, following the classification scheme of Mange and Morton (2007)
- alone, the provenance of Alpine B type garnets remains ambiguous. However, petrographic findings
- as well as zircon fission\_-track analysis and Ar/Ar dating in white mica (Spiegel et al., 2000; von
   Eynatten, 2003; von Eynatten and Wijbrans, 2003) strongly suggest a compositional change ca. 21 My
- 367 ago towards a metasedimentary source with a young cooling history. These authors relate this shift to
- 368 the erosion of the sedimentary cover of the Lepontine dome. Source rock samples taken within the
- 369 Lepontine dome from the crystalline basement (Antigorio nappe orthogneiss) and the meta-
- 370 sedimentary cover (Lebendun nappe paragneiss) contain generally almandine rich garnets, but those
- 371 from the paragneiss tend to be richer in grossular than those from the orthogneisses (Fig. 4). Because
- 372 the amount of grossular rich garnet is higher in the 19 My old sample compared to the 25 My old
- 373 sample, the data could support an origin from the Lepontine meta-sedimentary cover.

#### 374 4.2 Origin of granulite-facies garnetsEarly Miocene (~19 My ago)

Granulite facies garnet grains with relatively high pyrope and low grossular contents ("A type" and
"Class C" garnets according to Mange and Morton (2007) and Tolosana Delgado et al. (2018),
respectively) are only frequent in the 19 My-old Molasse sample (ca. 8-9 %, Table 3).

- The larger spread of garnet compositions in the early Miocene (~19 My) sample indicates the presence
   of several or mixed sources with different metamorphic grades, including amphibolite-, eclogite-, and
   granulite -facies rocks.
- The B-type garnet compositions match the range of garnets found in the Lepontine nappes (Fig. 4b, d), which is supported by the occurrence of predominantly young (<30 My) zircon fission track ages (Fig. 3) that in agreement withmatch the the young cooling ages of the Lepontine dome (Fig. 2eb; Bernet et al., 2009). Due to the overlap of amphibolite- facies garnets, it cannot be excluded <u>Alpine</u> that at least some of the garnets were contributed by Austroalpine sources or were recycled from older strata. The Lepontine dome was probably drained both towards the north and the south (Fig. 67b), because old basement detritus with young cooling ages (~30 My, derived from K-Ar on white mica)
- 388 was found in the Gonfolite Lombarda group in the southern retroforeland (Giger and Hurford, 1989).
- 389

Granulite\_-facies metamorphic conditions in the Central Alps were only reached in the Gruf complex
located close to the Insubric line between the Lepontine dome and the Bergell intrusion (Fig. 2<u>a</u>).
Furthermore, there is evidence for pre-Mesozoic granulite\_-facies metamorphism in some rocks in the
<u>Southern Alpine</u> Ivrea zone south of the Insubric line (Hunziker and Zingg, 1980), in the Sesia Zone

394 (Fig. 1; Engi et al., 2018; Giuntoli et al., 2018) and in the Dent Blanche nappe (Fig. 1; Angiboust et

395 al., 2009). It is unlikely that erosion reached so-that far to the South during the Miocene, because the 396 Penninic and probably also the exhuming Lepontine nappe stack would have acted as a topographic barrier to the fluvial drainage network (Fig. 67b). However, it was proposed that the flysch sediments 397 398 deposits preserved in the Prealps Romandes were partially fed by these units during the Late 399 Cretaceous and the Eocene (Wildi, 1985; Ragusa et al., 2017). This interpretation is supported by the 400 Gurnigel flysch sample (Fig. 4e), which contains garnet of granulite\_-facies type\_that are similar to 401 those found in the Ivrea zone (Table 3, Fig. 4h). A recycled flysch origin is supported further by the 402 abundance of flysch sandstone pebbles in Molasse strata of the same age (Schlunegger et al., 1998).

403 A potential, but minor contribution from ophiolites, as suggested by Spiegel et al. (2002), could be 404 supported by the two eclogite -facies garnet grains found in the 19 My-old sample (Fig. 4b) that match 405 eclogite -facies garnets from Alpine ophiolites (Table 3, Fig. 4g). Eclogite -facies garnets are known fromoccur both metamorphic rocks of the Penninic Alpine ophiolites (e.g. Bucher and Grapes, 2009; 406 407 Weber and Bucher, 2015, Fig. 2a), but also from Paleozoic (?) gneisses of the middle Penninic 408 Briançonnais basement (Sartori, 1990; Thélin et al., 1990). Both sources are not distinguishable (Fig. 409 4g), but would have probably been located in relative close geographic proximity, either in the 410 Penninic hanging wall south of the Simplon fault (Zermatt area) or in the Penninic nappes located 411 between the eastern rim of the Lepontine and the adjacent Austroalpine nappes (Arosa zone; Fig. 76b). 412

413 4.3 Origin of eclogite-facies garnetsMiddle Miocene (~14 My ago)

Previous provenance studies have identified meta-sedimentary detritus in the Middle Miocene
Molasse and located its source in the unroofing sedimentary cover of the Lepontine dome (e.g. von
Eynatten, 2003). This was strongly supported by the very-young detrital zircon fission track ages
(youngest peak at 19.5±0.9 My, Fig. 3; Spiegel et al., 2000) that match the zircon fission track ages of
the Lepontine dome (Fig. 2b, e.g. Hurford, 1986; Bernet et al., 2009).

419 However, garnet compositions in the youngest Molasse sandstones are not comparable to Lepontine garnets sampled in this study nor to any detrital garnet found in the main rivers draining the Lepontine 420 421 dome today (Andò et al., 2014). Instead, the detrital garnet signature of the 14 My-old sample mirrors 422 almost exactly the compositional range of garnets from the external crystalline massifs (Table 3, Fig. 423 4c, 4f). In the external crystalline massifs, these garnets grew in Permo-Carboniferous plutons under 424 Alpine greenschist -facies metamorphic conditions (Steck and Burri, 1971, Fig. 2a). They are 425 restricted to the granitoid basement of the external massifs and do not occur anywhere else in the 426 Central Alps, which makes them an excellent provenance proxy (Stutenbecker et al., 2017). A further 427 distinction among garnets supplied by the different plutons (e.g. the Central Aar granite from the Aar 428 massif, the Rotondo granite from the Gotthard nappe and the Mont Blanc granite from the Mont Blanc 429 massif) is not possible based on major element garnet geochemistry alone (Stutenbecker et al., 2017). 430 Until now, the surficial exposure of the external massifs in the Central Alps was thought to post-date 431 Molasse deposition. This interpretation relies principally on the absence of pebbles of external massif 432 origin (e.g. Aare granite) in the foreland basin (Trümpy, 1980). However, many Alpine granite bodies 433 closely resemble each other mineralogically and texturally, especially if present as altered pebbles in 434 the Molasse deposits, and hence it is difficult to discount a specific source only on this basis. Further 435 support of late surficial exposure of the external massifs comes from structural reconstructions (e.g. 436 Pfiffner, 1986; Pfiffner, 2017), that have located the top of the crystalline basement at an elevation that 437 is similar to the modern topography, based on a relatively flat-lying contact between the crystalline basement and the overlying Mesozoic sedimentary cover (Fig. 78a). According to this model and the 438 439 published exhumation rates of 0.5-0.7 km/My (Michalski and Soom, 1990; Glotzbach et al., 2010), the 440 top of the basement was buried 7-10 km below the surface 14 Ma ago. 441 However, Nibourel et al. (2018) recently proposed a revised geometry of the contact between

442 <u>crystalline basement and overlying cover, which allows ca. 8 km of additional crystalline basement on</u>

top of the present-day topography (Fig. 78b). The presence of external massif-sourced garnets in the 443

- youngest Molasse deposits provides independent evidence that parts of the crystalline crust comprised 444
- 445 in the external massifs were already at the surface at ca. 14 Ma (Fig. 67c). Assuming the
- 446 aforementioned average exhumation rates, 7-10 km of crystalline basement would have already been
- 447 exhumed and subsequently eroded during the past 14 My, which is in good agreement with the
- geometric reconstructions by Nibourel et al. (2018). 448
- We suggest that the drainage divide was shifted northwards due to the exhumation of the Gotthard 449 450 nappe and/or the Aar massif and that is was essentially located at its current position (Fig. 67c, d), but
- this warrants corroboration from other deposits in the foreland and the retroforeland. 451

#### 452 4.4 Origin of "igneous" garnets

Of the garnets from the youngest, 14 My old Molasse sample, 12 % can be classified as igneous 453 ("Class E", Table 4) according to Tolosana Delgado et al. (2018). Their high grossular and very low 454 455 pyrope content distinguishes them clearly from all the other, generally more almandine rich, garnets. 456 In the classification scheme after Mange and Morton (2007), however, this type of garnet plots in the D-type or in the rightmost part of the B-type or field (Fig. 4, Table 4). The detrital garnet signature of 457 458 the 14 My-old sample mirrors almost exactly the compositional range of garnets from the external crystalline massifs (Fig. 4c, 4f). In the external crystalline massifs, these garnets grew in Permo-459 460 Carboniferous plutons under alpine greenschist facies metamorphic conditions (Steck and Burri, 1971, 461 Fig. 2). They are restricted to the granitoid basement of the external massifs and do not occur 462 anywhere else in the Central Alps, which makes them an excellent provenance proxy (Stutenbecker et 463 al., 2017). A further distinction among garnets supplied by the different plutons (e.g. the Central Aar 464 granite from the Aar massif, the Rotondo granite from the Gotthard nappe or the Mont Blanc granite 465 from the Mont Blanc massif) is not possible based on garnet major element geochemistry alone 466 (Stutenbecker et al., 2017).

#### 467

#### 4.5 Implications for the evolution of the Alpine orogen

468 Previous provenance studies have identified meta sedimentary detritus in the youngest (ca. 21-14 My 469 old) Molasse and located its source in the unroofing sedimentary cover of the Lepontine dome (von 470 Eynatten, 2003). This was strongly supported by the very young detrital zircon fission track ages (youngest peak at 19.5±0.9 My, Spiegel et al., 2000) that match the exhumation patternzircon fission 471 472 track ages of the Lepontine dome\_(e.g. Hurford, 1986; Bernet et al., 2009). However, garnet compositions in the youngest Molasse sandstones are not comparable to Lepontine garnets sampled in 473 474 this study nor to any detrital garnet found in the main rivers draining the Lepontine dome today (Andò 475 et al., 2014).

Instead, the occurrence of grossular- and spessartine-rich garnets in the 14 My old Molasse mark a 476 477 distinct provenance change compared to the 19 My old deposits that was not noticed in previous 478 studies (Schlunegger et al., 1998; Spiegel et al., 2000; von Eynatten, 2003). Garnets of this particular 479 composition are described from the Permo-Carboniferous plutons intruded into the crystalline 480 basement of the Aar and Mont Blanc massifs and the Gotthard nappe (Steck and Burri, 1971). Such 481 particular chemical composition provides a unique sedimentary fingerprint (Stutenbecker et al., 2017). 482 Their occurrence in the youngest Molasse sediments has important implications for the tectonic 483 evolution of the orogen. Until now, the surficial exposure of the external massifs in the Central Alps 484 was thought to post date Molasse deposition. This interpretation relies principally on the absence of 485 pebbles of external massif origin (e.g. Aare granite) in the foreland basin (Trümpy, 1980). However, 486 many alpine granites closely resemble each other, especially if present as altered pebbles in the 487 Molasse deposits, and hence it is difficult to discount a specific source only on this basis. Further 488 support of late surficial exposure of the external massifs comes from structural reconstructions (e.g. Pfiffner, 1986; 2017), that have located the top of the crystalline basement similar to the modern 489

- 490 topography, based on a relatively flat-lying contact between the crystalline basement and the overlying
- 491 Mesozoic sedimentary cover (Fig. 7a). According to this model and the published exhumation rates of
- 492 0.5-0.7 km/My (Michalski and Soom, 1990; Glotzbach et al., 2010), the top of the basement must have
- 493 been buried 7-10 km below the surface 14 Ma ago. However, Nibourel et al. (2018) have recently
- 494 proposed a revised geometry of the contact between crystalline basement and overlying cover, which
- 495 allows ca. 8 km of additional crystalline basement on top of the present day topography (Fig. 7b). The
- 496 presence of external massif sourced garnets in the youngest Molasse deposits provides independent
- 497 evidence that parts of the crystalline crust comprised in the external massifs were already at the
   498 surface at ca. 14 Ma. Assuming the aforementioned average exhumation rates, 7-10 km of crystalline
- 499 basement would have already been exhumed (and subsequently eroded) during the past 14 My, which
- 500 is in good agreement with the geometric reconstructions by Nibourel et al. (2018).
- 501 <u>The resulting implications for the paleogeography and drainage evolution of the Central Alps, and in</u>
   502 <u>particular for the direct hinterland of the Napf fan, are summarized in Fig. 8.</u>
- 503 Although detrital garnet chemistry suggests exclusively contributions of amphibolite facies sources 504 during the latest Oligocene (~25 My), this methodology cannot distinguish between the diverse amphibolite facies rocks present in the Central Alps (e.g. alpine metamorphic rocks in the Lepontine 505 506 nappes vs. Paleozoic metamorphic rocks in the Austroalpine nappes). The related zircon fission track 507 data are mostly >100 My old with a small, younger, but badly constrained age peak of  $41\pm9$  My 508 (Spiegel et al., 2000). This would favor a dominant input from the Austroalpine nappes, which yield cooling ages older than ca. 50 My (e.g. Bernet et al., 2009, Gemignani et al. 2017), rather than from 509 510 the Lepontine nappes, which is characterized by zircon fission track ages younger than ca. 30 My (e.g. 511 Hurford, 1986). During this time, the drainage divide is probably located close to the Insubric line 512 (e.g. Schlunegger et al., 1998), but north of the Bergell pluton (Fig. 8a), whose detritus is exclusively 513 found in the retroforeland in the south (Gonfolite Lombarda, Giger and Hurford, 1989; Carrapa and Di 514 Giulio, 2001). 515 Garnets in the 19 My-old Molasse indicate a mixed contribution of sources that could be located in the Lepontine nappes as well as the Prealps Romandes (Fig. 8b). This is supported by the related young 516 517 (<30 My) zircon fission track ages (Spiegel et al. 2000) and the abundant flysch pebbles (Schlunegger 518 et al., 1998), respectively. A potential contribution from ophiolites, as suggested by Spiegel et al. 519 (2002), could be supported by the few eclogite facies garnet grains found in the 19 My old sample. 520 Their source could be located in the Penninic nappes in the hanging wall of the Rhone Simplon line or 521 in the Penninic nappes located between the Lepontine and the Austroalpine nappes (Fig. 8b). The 522 Lepontine dome was probably drained both towards the north and the south (Fig. 8b), because old
- 523 basement detritus with young cooling ages (~30 My, derived from K/Ar on white mica) was found in
- 524 the Gonfolite Lombarda group in the southern retroforeland (Giger and Hurford, 1989).
- 525 Finally, the garnet data suggests a dominant contribution from the external massifs and/or the Gotthard
- 526 <u>nappe at around 14 My ago (Fig. 8c). We suggest that the drainage divide was essentially located at its</u>
- 527 <u>current position (Fig. 8c, d), but this warrants corroboration from other deposits in the foreland and the</u>
- 528 <u>retroforeland.</u>

### 5. <u>Conclusions</u>

Garnet geochemistry is a useful tool to further constrain the provenance of sediments-sandstones in orogens such as the Central Alps. We have demonstrated that it is possible to distinguish detrital garnets using a combination of garnet classification schemes (Mange and Morton, 2007; Tolosana-Delgado et al., 2018) and case-specific comparison with available <u>A</u>elpine source rock compositions (Stutenbecker et al., 2017). For the Miocene deposits of the Swiss Molasse basin, we were able to (1) confirm the provenance shift possibly related to the exhumation of the Lepontine dome between 25 and 19 My ago as suggested by previously-studies (e.g., von Eynatten, 2003) and (2) to identify an

- 537 additional provenance shift between ca. 19 and 14 My ago that had not been noticed before. The This
- 538 latter shift before 14 My ago shift is related to the erosion of granites from the external crystalline
- 539 massifs, which provides a minimum age for their surficial exposure and corroborates their recently
- 540 revised structural geometry. (Fig. 7b). We conclude that the exposure of the crystalline basement
- happened already ca. 14 My ago, which is several million years earlier than previously assumed. In
   contrast to most previous studies, conclude that parts of the crystalline basement must have been
- 543 <u>exposed already ca. 14 My ago.</u>
- 544 <u>Data availability</u>
- 545 <u>The data (chemical composition of garnets from Molasse sandstones and source samples) can be found</u>
- 546 <u>online: https://figshare.com/articles/Detrital\_garnet\_chemistry\_from\_the\_Molasse\_basin/8269742/1</u>
   547 <u>doi: 10.6084/m9.figshare.8269742.v1.</u>
- 548 <u>Author contribution</u>
- 549 LS designed the project. AM helped during field work and sample collection. PT and PL gave advice
- 550 for sample preparation, supported the microprobe measurements and data acquisition at the University
- of Bern. LS prepared the manuscript with contributions by all co-authors.
- 552 <u>Competing interests</u>
- 553 The authors declare that they have no conflict of interest.
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814 Table 1: Sample locations and characteristics of the Molasse sandstones from the <u>Honegg-Napf fan.</u>

815 <u>Abbreviations used: UFM = Uupper Ffreshwater Molasse, UMM = Uupper mMarine Molasse, LFM</u> 816 <u>=  $\frac{1}{Lower}$  fFreshwater Molasse.</u>

Sample	Sampling	Lithostratigraphy	Magnetostratigraphic	Magnetostratigraphic	
name	location	(Matter, 1964;	section (Schlunegger	age (Schlunegger et	
		Schlunegger et al. 1996)	et al. 1996)	al. 1996)	
LS2017-3	47.00566	UFM, Napf beds	Fontannen section	ca. 14 <del>Ma<u>My</u></del>	
	7.971325				
LS2018-5	46.93913	UMM, Luzern	Schwändigraben	ca. 19 <del>Ma<u>My</u></del>	
	7.950800	formation	section		
LS2016-18	46.77463	LFM, Thun formation	Prässerebach section	ca. 25 <del>Ma</del> My	
	7.732383				

1

817	Table 2:	Sample	locations	and	characteristics	of	potential	source <u>s</u>	(tributary	sampling	approach)
818	<del>rocks</del>										

Sample	Sampling	River catchment	Metamorphic grade	Lithological unit
name	location			
LS2018-12	46.72026	Ärgera,	Not metamorphic	Gurnigel flysch
	7.24548	ca. 30 km <sup>2</sup>		(detrital garnets)
LS2018-40	46.39026	Valle di Foioi,	Alpine amphibolite	Orthogneiss, Antigorio
	8.54124	ca. $3 \text{ km}^2$	-facies	nappe, Lepontine dome
LS2016-43	46.43955	Valletta di Fiorina,	Alpine amphibolite	Paragneiss, Lebendun
	8.50115	ca. 8 km <sup>2</sup>	-facies	nappe, Lepontine dome

- 819 Table 3: Minimum, maximum and average <u>Average</u> contents (including standard deviation in
- 820 brackets) of the five common garnet endmembers in the Molasse sedimentssandstones, the fluvial
- 821 <u>samples from the Lepontine gneisses and the Gurnigel flysch (this study) and three potential source</u>
- 822 <u>rocks from the literature: External crystalline massif granites (Stutenbecker et al., 2017), eclogite</u>
- 823 <u>facies rocks (Chinner & Dixon, 1973; Ernst & Dal Piaz, 1978; Oberhänsli, 1980; Sartori, 1990;</u>
- 824 <u>Thélin et al., 1990; Reinecke, 1998; Cartwright & Barnicoat, 2002; Angiboust et al., 2009; Bucher &</u>
- 825 Grapes, 2009; Weber & Bucher, 2015), and granulite facies rocks (Hunziker & Zingg 1980). For the
- 826 *full dataset we refer to Stutenbecker (2019).*

Sample	Almandine	Andradite	Grossular	Pyrope	Spessartine
	(%)	(%)	(%)	(%)	(%)
25 My	70 (12)	2 (5)	9 (7)	9 (5)	9 (8)
n=110					
19 My	65 (16)	3 (13)	16 (12)	9 (8)	5 (6)
n=88					
14 My	50 (12)	2 (2)	32 (11)	6 (5)	9 (9)
n=77					
<b><u>Valle di Foioi (</u>Antigorio</b>	<u>67 (10)</u>	<u>1 (1)</u>	<u>11 (12)</u>	<u>10 (6)</u>	<u>10 (10)</u>
orthogneiss)					
<u>n=45</u>					
Valletta di Fiorina (Lebendun	<u>64 (5)</u>	<u>0(1)</u>	<u>22 (4)</u>	<u>8 (3)</u>	<u>5 (3)</u>
paragneiss <u>)</u>					
<u>n= 56</u>					
<u>Årgera (</u> Gurnigel flysch <u>)</u>	<u>69 (12)</u>	<u>2(1)</u>	<u>9 (7)</u>	<u>14 (8)</u>	<u>6 (9)</u>
<u>n=75</u>					
Goneri and Wysswasser	<u>34 (16)</u>	<u>0 (0)</u>	<u>35 (14)</u>	<u>4 (5)</u>	<u>21 (10)</u>
rivers (eExternal crystalline					
massif granites <u>)</u>					
<u>n=212</u>					
Eclogite facies	<u>56 (8)</u>	<u>0(1)</u>	<u>23 (6)</u>	<u>16 (10)</u>	<u>3 (5)</u>
<u>n=147</u>					
Granulite facies	<u>67 (8)</u>	<u>0 (0)</u>	<u>4 (1)</u>	<u>25 (10)</u>	<u>4 (4)</u>
<u>n=18</u>					

- 827 Table 4: Results from classification following Mange & Morton (2007) and Tolosana-Delgado et al.
- 828 (2018). Using the linear discriminant method of Tolosana-Delgado et al. (2018) garnets were was 829 attributed to one single class if the probability for that class was  $\geq 50$  %. Several grains were assigned
- 830 *mixed probabilities with <50 % per class; these are listed separately below.*

	Mange & Morton (2007)				Tolosana-Delgado (2018)		et al.	
TypesafterMange&Morton (2007)	25 My	19 My	14 My	Classes after Tolosana-Delgado et al. (2018)	25 My	19 My	14 My	
Ci-type (high- grade metabasic)		5 %	15 %	Eclogites (Class A)		1 %		
B-type (amphibolite facies)	96 %	84 %	80 %	Amphibolites (Class B)	71 <u>92</u> %	81 %	78 %	
A-type (granulite facies)	3 %	8 %		Granulites (Class C)		9 %	5.5 %	
D-type (metasomatic)	1 %	3 %	5 %	Igneous (Class E)	<u>7</u> <del>5</del> %	3 %	12 %	
				Mixed probabilities Classes B-C	1 %	1 %		
				Mixed probabilities Classes A-B-C		5 %	4.5 %	



833

- 834 Fig. 1: Simplified tectonic map of the Central Alps after Schmid et al. (2004) highlighting the location
- of alluvial fan deposits within the northern <u>aA</u>lpine foreland basin as well as the most important
- source rock units in the hinterland. The <u>Honegg-Napf fan</u>, marked by the black rectangle, is located in
  the central part of the Swiss foreland basin (SFB). <u>For cross section X-X'' see Fig.<del>ure 87</del></u>.
- 838 <u>Abbreviations used: AR = Aiguilles-Rouges massif, BD = Belledonne massif, DB = Dent Blanche</u></u>
- 839 nappe, HN = Honegg-Napf fan, MB = Mont Blanc massif, GN = Gotthard nappe, PE = Pèlerin fan,
- 840 PF = Pfänder fan, <del>HN = Honegg-Napf fan,</del> RH = Rigi-Höhronen fan, SKH = Speer-Kronberg-Hörnli
- 841 *fan, SZ = Sesia zone.<del>, PF = Pfänder fan.</del>*



842

843 Fig. 2: a) Metamorphic map of the Central Alps (Bousquet et al., 2012) showing the distribution and 844 grade of *alpine Alpine* metamorphism. Note the increase from north to south from lower greenschist-845 to eclogite- facies conditions. b) In-situ bedrock zircon fission track ages according to a compilation of Bernet et al. (2009). Note the predominantly young (<30 My) cooling ages in the area around the 846 847 Lepontine dome and the external massifs in contrast to the predominantly old (>50 My) cooling ages 848 in the Austroalpine nappes to the east. The river network (blue) and the thick black outlines of selected geological units (external massifs, Prealps Romandes and Dent Blanche nappe, cf. Fig. 1) are used to 849 850 facilitate the orientation and the comparison with  $Fi_{gure}$ . 1. Abbreviations used:  $PE = P \stackrel{\circ}{e} lerin fan$ , 851 <u>HN = Honegg-Napf fan, RH = Rigi-Höhronen fan, SKH = Speer-Kronberg-Hörnli fan, PF = Pfänder</u> 852 fan.



853

Fig. 3: Compilation of published compositional data in the Honegg-Napf fan. Heavy mineral and rock fragment data from the sand grain size after von Eynatten (2003), pebble petrography after Schlunegger et al. (1998), epidote isotope ratios after Spiegel et al. (2002) and zircon fission\_-track (FT) data after Spiegel et al. (2000). The two pink lines represent the dominant provenance changes as discusses in the text. <u>Abbreviations used: LMM = 4Lower Mmarine Molasse, LFM = L4ower</u> <u>fFreshwater Molasse, UMM = #Upper mMarine Molasse, UFM = #Upper fFreshwater Molasse, ZTR</u> <u>= zircon-tourmaline-rutile-index, sil. = siliceous.</u>



862 Fig. 4: Ternary plots followingGarnet-the\_classification scheme of Mange & Morton (2007). (a-c) Detrital garnet compositions in the 25, 19 and 14 My-old Molasse deposits (this study). Garnet 863 provenance changes in Molasse sandstones are marked by an increasing grossular content with 864 865 decreasing age. Source rock data from (d) Lepontine gneisses (this study), (e) the Gurnigel flysch (this study), (f) external massif granitoids (Stutenbecker et al., 2017), (g) eclogite-facies rocks (Chinner & 866 Dixon, 1973; Ernst & Dal Piaz, 1978; Oberhänsli, 1980; Sartori, 1990; Thélin et al., 1990; Reinecke, 867 1998; Cartwright & Barnicoat, 2002; Angiboust et al., 2009; Bucher & Grapes, 2009; Weber & 868 869 Bucher, 2015), (h) granulite-facies rocks from the Ivrea zone in the Southern Alps (Hunziker & Zingg, 870 1980).



871 Molar proportion of grossular
872 Fig. 5: Shift of garnet compositions between the 25 My-, 19 My and 14 My old Molasse samples,
873 plotted as r<u>R</u>elative frequency of the four most common endmembers almandine, grossular,
874 spessartine and pyrope in the three detrital samples from the Molasse basin. While spessartine and
875 pyrope contents are similar among the three samples, the proportion of almandine decreases and the
876 proportion of grossular increases with decreasing age.
877



880 881 the Honegg-Napf fan. Situation during a) the late Oligocene (~25 My), b) the early Miocene (~19 My), 882 c) the middle Miocene (~14 My) and d) today (after Schmid et al., 2004). The color coding in a-c) 883 corresponds essentially to the color coding in d) (,-see Fig. 1 for detailed legend). However, we have 884 summarized the lower, middle and upper Penninic nappes and the Dent Blanche nappe (pink color) as well as the carbonate and flysch nappes of the Helvetic nappes and the Prealps Romandes (blue 885 color). -Abbreviations used: AA = Aar massif, BP = Bergell pluton, GN = Gotthard nappe, HN =886 887 Honegg-Napf fan, IL = Insubric line, RH = Rigi-Höhronen fan, RSL = Rhone-Simplon lineament,888 *SKH* = *Speer-Kronberg-Hörnli fan.* 



890 Fig. 7: Cross sections from X to X''<u>in Figur.e 1</u> through the Aar massif simplified after Pfiffner

891 (2017) and Nibourel et al. (2018). For trace of cross section see Fig. 1. (a): The reconstructed top of 892 the crystalline basement in the Aar massif is located ca. 1-2 km higher than the present-day

topography according to Pfiffner (2017). (b): In a revised version by Nibourel et al. (2018) the contact

894 between the basement and the overlying Helvetic cover nappes is reconstructed to be steeper,

895 *resulting in ca. 8 km of (now eroded) crystalline crust on top of the present-day topography.*