Miocene basement exhumation in the Central Alps recorded by detrital garnet geochemistry in foreland basin deposits

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Abstract

The Neogene evolution of the European Alps was characterized by the exhumation of crystalline basement, the so-called external crystalline massifs. Their exhumation presumably controlled the evolution of relief, distribution of drainage networks and generation of sediment in the Central Alps. However, due to the absence of suitable proxies, the timing of their surficial exposure, and thus the initiation of sediment supply from these areas, are poorly constrained.

The northern Aalpine foreland basin preserves the Oligocene to Miocene sedimentary record of tectonic and climatic adjustments in the hinterland. This contribution analyses the provenance of 25 to 14 My-old alluvial fan deposits by means of detrital garnet chemistry. Unusually grossular- and spessartine-rich garnets isare 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 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.

1. Introduction

Tectonic processes ~~influence~~ drive the evolution of relief in mountain chains and consequently control the development of the drainage network, sediment supply and deposition in the foreland basin. The Central European Alps and their northern foreland basin, formed through the collision of the European and the Adriatic continents since the Eocene (Schmid et al. 1996~~,~~; Handy et al. 2010), are a classic example of such interactions (e.g. Schlunegger et al., 1998; Pfiffner et al., 2002; Vernon et al., 2008, 2009; Baran et al., 2014; Fox et al., 2015). The exhumation of large slices of mid-crustal rocks from the European plate, the so-called external crystalline massifs, occurred ~~during a late-stage orogenic event~~relatively late in the Alpine evolution, probably during the late Miocene, although the exact timing is not well constrained. The external crystalline massifs are today characterized by high relief, intense glaciation and some of the highest denudation rates ~~measured~~ in the Alps (up to 1.4 mm/y), 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 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 increased precipitation rates in the Pliocene (Cederbom et al., 2004) or enhanced glacial erosion in the Pleistocene (Fox et al., 2015; Herman et al., 2013). ~~The areas exhumed during this event are today characterized by high relief, intense glaciation and some of the highest denudation rates measured in the Alps (up to 1.4 mm/y), which all contribute to their importance as a sediment source (Kühni and Pfiffner, 2001; Wittmann et al., 2007; Stutenbecker et al., 2018).~~

Peak metamorphism of lower to upper greenschist-facies conditions occurred between 17 and 22 Ma in all northern external crystalline massifs (Mont Blanc, Aar massifs and the Gotthard nappe, Challandes et al., 2008; Rolland et al., 2008; Cenki-Tok et al., 2014; Nibourel et al., 2018). Their subsequent exhumation has been investigated using thermochronology ~~by a number of studies~~ (e.g. Schaer et al., 1975, Wagner et al., 1977; Michalski and Soom, 1990; Vernon et al., 2009; Glotzbach et al., 2010). ~~While~~ Whereas some studies concluded that exhumation was episodic (e.g. Vernon et al., 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 the onset of glaciation in the Pleistocene and their possible effect on the exhumation, erosion and sediment accumulation rates (e.g. Kuhlemann et al., 2002; Herman et al., 2013; Schildgen et al., 2017). In contrast, the ~~Paleogene and~~ early Neogene exhumation history ~~is~~received comparably little attention ~~less well studied~~. In particular, ~~T~~the timing of the first surficial exposure of the external massifs ~~has, however,for example, has~~ never been constrained, because estimates of their total thickness have not ~~yet~~ been established yet. In most geometric reconstructions (e.g. Pfiffner, 1986~~,~~; Pfiffner, 2017; Schmid et al., 2004), the contact between the crystalline basement and the overlying Mesozoic cover is assumed to be relatively flat, and the top of the crystalline basement is hypothesized 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 rock on top of the present-day topography (Nibourel et al., 2018).

This study aims to constrain the timing of exposure, and thus the beginning of sediment supply from the external crystalline massifs~~,~~ by determining the provenance of the foreland basin deposits. Sedimentary rocks~~s~~ preserved in the northern peripheral foreland basin of the Central Alps, the Swiss part of the Molasse basin, are a well-studied archive recording tectonic and climatic adjustments in the central orogen between ca. 32 and 14 My ago (Schlunegger et al., 1993, 1996; Kempf et al., 1999; Spiegel et al., 2000; Kuhlemann and Kempf, 2002; von Eynatten, 2003; Schlunegger and Kissling, 2015). So far, the provenance of the Molasse deposits has been investigated using optical heavy 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 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 of settings (Spear, 1994; Mange and Morton, 2007). Furthermore, it is a common heavy mineral in (orogenic) sediments and sedimentary rocks (Garzanti and Andò, 2007) and is relatively stable during transport and diagenesis (Morton and Hallsworth, 2007). In the Central Alps, detrital garnet has 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 detrital garnet geochemistry can help identifying additional provenance changes in the Miocene Molasse deposits that have gone unnoticed so far provide additional provenance information to unravel the Miocene history of the Molasse deposits and its tectonic forcing 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.

1.1 Geological Setting

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

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

The hinterland of the central Swiss foreland basin comprises, from north to south, potential source 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 carbonate~~s~~ and Cretaceous-Eocene flysch), interpreted as the accretionary wedge of the A~~a~~lpine 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 carbonate~~s~~).

(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 Carboniferous to Permian granitoid rocks and an overlying metasedimentary cover. They were buried within the Alpine nappe stack ~~in~~ during the Oligocene (Cenki-Tok et al., 2014), 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 stricto* because of its allochthonous nature, will be included in~~to~~ the term "external crystalline massifs" from here on, because the timing and the rates of exhumation are comparable (Fig. 2b, Glotzbach et al., 2010).

(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 ophiolite~~s~~ of the A~~a~~lpine Tethys as well as the continental crust of Briançonnais, a microcontinent located within the ~~a~~Alpine 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).

1.2 Compositional trends in the Honegg-Napf fan

~~Rocks from the~~The Central Alps are generally considered as the major sediment source of all proximal Molasse basin deposits, ~~while~~ and compositional changes in the foreland are thought to directly reflect tectonic and erosional processes in the immediate A~~a~~lpine 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 Honegg-Napf fan, located in the central part of the basin. ~~,~~ It ~~which is the~~ most likely ~~to archive~~preserves 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-tourmaline-rutile ~~(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 ~~a~~Alpine nappe stack and probably extended further west during this time (Schlunegger et al., 1998; Spiegel et al., 2000; von Eynatten, 2003).

(Phase 2) 25-21 My ago: Around 25 My ago, the occurrence of epidote as well as an increase in ~~granitic~~ granitoid rock fragments mark a major compositional change in the foreland. The presence of characteristic colorful granite pebbles suggests an origin from the Austroalpine Bernina nappe (Matter, 1964). Sediments of this phase clearly reflect the ~~down~~ cutting down into crystalline basement and are 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-Mischabel nappe and argue that parts of the epidote could originate from Penninic ophiolites as well, thus suggesting that erosion might have ~~already~~ reached down into the Penninic nappes already by then. Spiegel et al.~~,~~ (2002) argued against this Penninic contribution based on the $^{87}Sr/^{86}Sr$ and $^{143}Nd/^{144}Nd$ isotopic signatures of the epidote.

(Phase 3) 21-14 My ago: At ~21 My, metamorphic rock fragments occur in the sediments, ~~while~~ whereas the heavy mineral assemblages remain epidote-dominated and overall similar to the second phase. Zircon fission track ages are exclusively Cenozoic (age~~s~~ peaks between ~32 and ~19 Ma). In contrast to the first two phases, the sediment composition allows several, partially contradicting interpretations. Whilst petrographic~~al~~ and mineralogic~~al~~ data might suggest recycling and sediment mixing (von Eynatten, 2003), young $^{40}Ar/^{39}Ar$ cooling ages in white mica (von Eynatten, 2003; von Eynatten and Wijbrans, 2003) and ~~exclusively young~~ a population of zircons with a fission track central age~~s~~ of 19.5±0.9 My ~~-~~(Spiegel et al., 2000) point to an additional, newly exhumed source ~~that these authors~~ identifie~~d~~ 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 scenario, in which the erosional front shifted northwards into the flysch nappes of the Prealps Romandes. A mixture of both sources seems possible. Furthermore, the isotopic signature of detrital epidotes suggests a contribution of mantle source rocks between ca. 21 and 19 My ago, which could point to a contribution by Penninic ophiolites (Spiegel et al., 2002). However, this is not reflected in the heavy mineral spectra (von Eynatten, 2003) that ~~, which~~ do not contain typical ophiolite minerals such as Cr-spinel.

~~In none of these scenarios were t~~The external crystalline massifs have not been 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 ~~sediments~~deposits. 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).

2. Sampling strategy and methodology

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 Honegg-Napf fan deposits located ca. 40 kilometers to the ~~East~~ east and ~~Southeast~~ southeast of Berne in the central part of the Swiss Molasse basin. The exact sampling sites were chosen based on the availability of published petrographical, chemical and mineralogical data (von Eynatten, 2003) as well as magnetostratigraphic calibration (Schlunegger et al., 1996).

It is possible to compare potential source compositions to the detrital ones, b~~B~~ecause 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 A~~a~~lpine chain today~~, it is possible to 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., 1999; Cartwright and Barnicoat, 2002; Bucher and Bousquet, 2007; Angiboust et al., 2009; Bucher and Grapes, 2009; Weber and Bucher, 2015).

In addition, three river sand samples were collected from small monolithological catchments (3-30 km^2) draining potentially garnet-bearing ~~potential~~ source rocks that were previously not, or only partially, considered in the literature. We prefer this "tributary sampling approach" (first-order sampling scale according to ~~see e.g. Stutenbecker et al., 2017~~Ingersoll, 1990) over *in-situ* sampling of specific source rocks, because small monolithological catchments are more likely to comprise all garnet varieties of the targeted source rock and to average out spatial variations of the source rock properties, e.g. mineral size or fertility (Malusà et al., 2016). ~~differences in garnet fertility~~. The 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 Lepontine dome (Fig.1). Sample characteristics are summarized in Table 1 and Table 2. For detailed lithological descriptions of the ~~sampled~~ sampling sites in the Honegg-Napf area, see Schlunegger et al. (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 µm, 63-125 µm, 125-250 µm and >250 µm. The fractions of 63-125 µm and 125-250 µm were further processed in sodium polytungstate heavy liquid at 2.85 g/cm^3 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.

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 ¼ µm diamond suspensions and graphite-coated. Major element oxides were analyzed using a JEOL JXA-8200 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 voltage of 15 k~~K~~eV, electron beam current of 15 nA, beam diameter of 1µm, 20 s peak acquisition time for Si, Ti, Al, Fe, Mn, Mg, Ca and 10 s for both backgrounds. Natural and synthetic standard olivine (SiO_2, MgO, FeO), anorthite (Al_2O_3, CaO) ilmenite (TiO_2) and tephroite (MnO) were used for calibration by applying a CITIZAF correction (Armstrong, 1984). Garnet compositions were measured as close as possible to the geometric centers of the grains, unless the area was heavily fractured or 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).

Molecular proportions were calculated from the measured main oxide compositions on the base of 12 anhydrous oxygen atoms. ~~Because ferric and ferrous iron were not measured separately (FeO = Fe_{total}),~~ T~~t~~he Fe^{2+}/Fe^{3+} ratio was determined based on charge balance (Locock, 2008), because ferric and ferrous iron were not measured separately (FeO = Fe_{total}). Garnet endmember compositions were subsequently calculated using the Excel spreadsheet by Locock (2008). ~~Garnet is a solid solution between different endmembers, the most common ones being almandine ($Fe_3Al_2Si_3O_{12}$), grossular ($Ca_3Al_2Si_3O_{12}$), pyrope ($Mg_3Al_2Si_3O_{12}$), spessartine ($Mn_3Al_2Si_3O_{12}$) and andradite ($Ca_3Fe_2Si_3O_{12}$).~~ The relative proportions of the~~se~~ endmember components almandine ($Fe_3Al_2Si_3O_{12}$), grossular ($Ca_3Al_2Si_3O_{12}$), 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 can vary substantially depending on the metamorphic or magmatic history of the protolith (Deer et al., 1992; Spear, 1994). The data were plotted and classified using the ternary diagram of Mange and 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 al., 2014).

3. Results

Most of the detrital garnets are dominated by ~~the~~ Fe-rich almandine ~~endmember~~ with varying amounts of grossular, pyrope, spessartine and andradite (Fig. 4). Other endmembers (e.g. uvarovite) are negligible. ~~Minimum, maximum and average~~Average endmember contents are summarized in Table 3; for the full dataset we refer to Stutenbecker (2019). Garnet compositions do not differ significantly between the two analyzed grain size fractions of the same sample, although ~~some~~ slight variations are visible ~~in the ternary plot~~ (Fig. 4): i~~I~~n sample LS2016-18 (25 My, Fig. 4a) garnet~~s~~ of the 125-250 µm fraction ~~tend is to be~~ more enriched in pyrope ~~with respect to~~than garnet~~s~~ of the 63-125 µm fraction. In sample LS2018-5 (19 My, Fig. 4b) 4 "outliers" that are very pyrope- and grossular-rich (n=2) or grossular- and andradite-rich (n=2) occur only in the 63-125 µm grain size fraction. Furthermore, 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 grains (n=8) of high almandine and low grossular content that are absent in the 125-250 µm fraction.

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

~~According to the ternary classification plot of Mange and Morton (2007), t~~The 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~~ dominant contribution by amphibolite-facies source rocks (Table 4). Minor ~~portions~~ amounts are ~~derived from~~classified as C-type (high-grade metabasic), A-type (granulite facies) and D-type (metasomatic) ~~sources~~garnet. The 25 My-old sandstone contains almost exclusively B-type garnet (92%, Table 4). The 19 My-old sandstone shows a larger spread with some A-, C- and D-type garnet (Fig. 4b, Table 4). The 14 My-old sandstone contains B-, C- and D-type garnet (Fig. 4c, Table 4). Classification through linear discriminant analysis (Tolosana-Delgado et al., 2018) yields a similar trend with generally high proportions of amphibolite-facies source rocks (class B-garnets, >70 %, Table 4). Some grains (5 %, 3 % and 12 % in the 25 My-, 19 My- and 14 My-old ~~samples~~deposits, respectively) were classified as igneous garnet (Table 4).

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

~~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.~~

~~All source rocks, except for the external crystalline massif granites, supply almandine-dominated (i.e. >50 % almandine component) garnet. The andradite content in all source rock garnets is very low, but they contain varying amounts of grossular, spessartine and pyrope.~~ Garnets from the Lepontine 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 crystalline massifs (Fig. 4f) are unusually rich in grossular (35 %) and spessartine (21 %), and the almandine content is much lower than in the other source rock garnets (34 %). Eclogite facies garnets have high grossular (23 %) and pyrope (16 %) contents (Fig. 4g). Granulite facies garnets (Fig. 4g) have on average the highest pyrope content of all source rock garnets (25 %).~~

4. Discussion

4.1 ~~Origin of amphibolite-facies garnets~~Late 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. meta-sedimentary 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; 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 nappe (Spillmann, 1993; Spillmann and Büchi, 1993), the middle Penninic Briançonnais basement (Sartori et al., 2006) or the polycyclic basement of the external massifs (von Raumer et al., 1999). In fact, the Gurnigel flysch, a Late Cretaceous to Eocene flysch nappe in the Prealps Romandes that did not undergo Alpine metamorphism (Fig. 2a), contains abundant almandine-rich B-type garnets (Fig. 4e).

Zircon fission track ages from sandstones of the same age are mostly >100 My old with a smaller and 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. 6~~7~~a), which yield related cooling ages >50 My (Fig. 2b; e.g. Bernet et al., 2009), rather than from the Lepontine dome, which is characterized by zircon fission track ages <30 My (Fig. 2b; e.g. Hurford, 1986). The presence of granite pebbles

attributable to the Austroalpine Bernina nappe (Matter, 1964; Schlunegger et al., 1998) would further support an Austroalpine rather than a Lepontine provenance.

The drainage divide was probably located close to the Insubric line (e.g. Schlunegger et al., 1998), but north of the Bergell pluton (Fig. 6~~7~~a), 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 source rocks ("B-type"). Garnets derived from amphibolite facies rocks ("B-type") seem to be the most frequent ones in all three considered samples. In the Central Alps, amphibolite facies conditions of alpine age were only reached in the Lepontine nappes (Fig. 2). However, many gneisses in the area 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 massifs (von Raumer et al., 1999). In fact, the Gurnigel flysch, a Late Cretaceous to Eocene flysch nappe in the Prealps Romandes that did not undergo alpine metamorphism (Fig. 2), contains almost exclusively almandine-rich B-type garnets (Fig. 4e).~~

~~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 ago towards a metasedimentary source with a young cooling history. These authors relate this shift to the erosion of the sedimentary cover of the Lepontine dome. Source rock samples taken within the Lepontine dome from the crystalline basement (Antigorio nappe orthogneiss) and the meta-sedimentary cover (Lebendun nappe paragneiss) contain generally almandine-rich garnets, but those from the paragneiss tend to be richer in grossular than those from the orthogneisses (Fig. 4). Because the amount of grossular-rich garnet is higher in the 19 My-old sample compared to the 25 My-old sample, the data could support an origin from the Lepontine meta-sedimentary cover.~~

4.2 ~~Origin of granulite facies garnets~~Early 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 ~~occurence~~occurrence of predominantly young (<30 My) zircon fission track ages (Fig. 3) ~~that~~ in agreement with~~match~~ the ~~the~~ young cooling ages of the Lepontine dome (Fig. 2~~c~~b; Bernet et al., 2009). Due to the overlap of amphibolite-facies garnets, it cannot be excluded ~~Alpine~~ 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. 6~~7~~b), because old basement detritus with young cooling ages (~30 My, derived from K-Ar on white mica) was found in the Gonfolite Lombarda group in the southern retroforeland (Giger and Hurford, 1989).

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. 2a). Furthermore, there is evidence for pre-Mesozoic granulite-facies metamorphism in some rocks in the Southern Alpine Ivrea zone south of the Insubric line (Hunziker and Zingg, 1980), in the Sesia Zone (Fig. 1; Engi et al., 2018; Giuntoli et al., 2018) and in the Dent Blanche nappe (Fig. 1; Angiboust et

al., 2009). It is unlikely that erosion reached ~~so~~ that far to the South during the Miocene, because the Penninic and probably also the exhuming Lepontine nappe stack would have acted as a topographic barrier to the fluvial drainage network (Fig. 6~~7~~b). However, it was proposed that the flysch ~~sediments~~ deposits preserved in the Prealps Romandes were partially fed by these units during the Late Cretaceous and the Eocene (Wildi, 1985; Ragusa et al., 2017). This interpretation is supported by the Gurnigel flysch sample (Fig. 4e), which contains garnet of granulite-facies type that are similar to those found in the Ivrea zone (Table 3, Fig. 4h). A recycled flysch origin is supported further by the abundance of flysch sandstone pebbles in Molasse strata of the same age (Schlunegger et al., 1998).

A potential, but minor contribution from ophiolites, as suggested by Spiegel et al. (2002), could be supported by the two eclogite-facies garnet grains found in the 19 My-old sample (Fig. 4b) that match eclogite-facies garnets from Alpine ophiolites (Table 3, Fig. 4g). Eclogite-facies garnets ~~are known from~~occur both metamorphic rocks of the Penninic Alpine ophiolites (e.g. Bucher and Grapes, 2009; Weber and Bucher, 2015, Fig. 2a), but also from Paleozoic (?) gneisses of the middle Penninic Briançonnais basement (Sartori, 1990; Thélin et al., 1990). Both sources are not distinguishable (Fig. 4g), but would have probably been located in relative close geographic proximity, either in the Penninic hanging wall south of the Simplon fault (Zermatt area) or in the Penninic nappes located between the eastern rim of the Lepontine and the adjacent Austroalpine nappes (Arosa zone; Fig. ~~7~~6b).

4.3 ~~Origin of eclogite-facies garnets~~Middle 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).

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 dome today (Andò et al., 2014). Instead, the detrital garnet signature of the 14 My-old sample mirrors almost exactly the compositional range of garnets from the external crystalline massifs (Table 3, Fig. 4c, 4f). In the external crystalline massifs, these garnets grew in Permo-Carboniferous plutons under Alpine greenschist-facies metamorphic conditions (Steck and Burri, 1971, Fig. 2a). They are restricted to the granitoid basement of the external massifs and do not occur anywhere else in the Central Alps, which makes them an excellent provenance proxy (Stutenbecker et al., 2017). A further distinction among garnets supplied by the different plutons (e.g. the Central Aar granite from the Aar massif, the Rotondo granite from the Gotthard nappe and the Mont Blanc granite from the Mont Blanc massif) is not possible based on major element garnet geochemistry alone (Stutenbecker et al., 2017).

Until now, the surficial exposure of the external massifs in the Central Alps was thought to post-date Molasse deposition. This interpretation relies principally on the absence of pebbles of external massif origin (e.g. Aare granite) in the foreland basin (Trümpy, 1980). However, many Alpine granite bodies closely resemble each other mineralogically and texturally, especially if present as altered pebbles in the Molasse deposits, and hence it is difficult to discount a specific source only on this basis. Further support of late surficial exposure of the external massifs comes from structural reconstructions (e.g. Pfiffner, 1986; Pfiffner, 2017), that have located the top of the crystalline basement at an elevation that is similar to the modern topography, based on a relatively flat-lying contact between the crystalline basement and the overlying Mesozoic sedimentary cover (Fig. 7~~8~~a). According to this model and the published exhumation rates of 0.5-0.7 km/My (Michalski and Soom, 1990; Glotzbach et al., 2010), the top of the basement was buried 7-10 km below the surface 14 Ma ago.

However, Nibourel et al. (2018) recently proposed a revised geometry of the contact between crystalline basement and overlying cover, which allows ca. 8 km of additional crystalline basement on

top of the present-day topography (Fig. 78b). The presence of external massif-sourced garnets in the youngest Molasse deposits provides independent evidence that parts of the crystalline crust comprised in the external massifs were already at the surface at ca. 14 Ma (Fig. 67c). Assuming the aforementioned average exhumation rates, 7-10 km of crystalline basement would have already been exhumed and subsequently eroded during the past 14 My, which is in good agreement with the geometric reconstructions by Nibourel et al. (2018).

We suggest that the drainage divide was shifted northwards due to the exhumation of the Gotthard 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.

4.4 Origin of "igneous" garnets

Of the garnets from the youngest, 14 My old Molasse sample, 12 % can be classified as igneous ("Class E", Table 4) according to Tolosana-Delgado et al. (2018). Their high grossular and very low pyrope content distinguishes them clearly from all the other, generally more almandine-rich, garnets. 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 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-Carboniferous plutons under alpine greenschist facies metamorphic conditions (Steck and Burri, 1971, Fig. 2). They are restricted to the granitoid basement of the external massifs and do not occur anywhere else in the Central Alps, which makes them an excellent provenance proxy (Stutenbecker et al., 2017). A further distinction among garnets supplied by the different plutons (e.g. the Central Aar granite from the Aar massif, the Rotondo granite from the Gotthard nappe or the Mont Blanc granite from the Mont Blanc massif) is not possible based on garnet major element geochemistry alone (Stutenbecker et al., 2017).

4.5 Implications for the evolution of the Alpine orogen

Previous provenance studies have identified meta-sedimentary detritus in the youngest (ca. 21 14 My old) Molasse and located its source in the unroofing sedimentary cover of the Lepontine dome (von 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 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 this study nor to any detrital garnet found in the main rivers draining the Lepontine dome today (Andò et al., 2014).

Instead, the occurrence of grossular- and spessartine-rich garnets in the 14 My old Molasse mark a distinct provenance change compared to the 19 My old deposits that was not noticed in previous studies (Schlunegger et al., 1998; Spiegel et al., 2000; von Eynatten, 2003). Garnets of this particular composition are described from the Permo-Carboniferous plutons intruded into the crystalline basement of the Aar and Mont Blanc massifs and the Gotthard nappe (Steck and Burri, 1971). Such particular chemical composition provides a unique sedimentary fingerprint (Stutenbecker et al., 2017).

Their occurrence in the youngest Molasse sediments has important implications for the tectonic evolution of the orogen. Until now, the surficial exposure of the external massifs in the Central Alps was thought to post-date Molasse deposition. This interpretation relies principally on the absence of pebbles of external massif origin (e.g. Aare granite) in the foreland basin (Trümpy, 1980). However, many alpine granites closely resemble each other, especially if present as altered pebbles in the Molasse deposits, and hence it is difficult to discount a specific source only on this basis. Further 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

topography, based on a relatively flat-lying contact between the crystalline basement and the overlying Mesozoic sedimentary cover (Fig. 7a). According to this model and the published exhumation rates of 0.5-0.7 km/My (Michalski and Soom, 1990; Glotzbach et al., 2010), the top of the basement must have been buried 7-10 km below the surface 14 Ma ago. However, Nibourel et al. (2018) have recently proposed a revised geometry of the contact between crystalline basement and overlying cover, which allows ca. 8 km of additional crystalline basement on top of the present day topography (Fig. 7b). The presence of external massif-sourced garnets in the youngest Molasse deposits provides independent evidence that parts of the crystalline crust comprised in the external massifs were already at the surface at ca. 14 Ma. Assuming the aforementioned average exhumation rates, 7-10 km of crystalline basement would have already been exhumed (and subsequently eroded) during the past 14 My, which is in good agreement with the geometric reconstructions by Nibourel et al. (2018).

The resulting implications for the paleogeography and drainage evolution of the Central Alps, and in particular for the direct hinterland of the Napf fan, are summarized in Fig. 8.

Although detrital garnet chemistry suggests exclusively contributions of amphibolite-facies sources 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 nappes vs. Paleozoic metamorphic rocks in the Austroalpine nappes). The related zircon fission track data are mostly >100 My old with a small, younger, but badly constrained age peak of 41±9 My (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 the Lepontine nappes, which is characterized by zircon fission track ages younger than ca. 30 My (e.g. Hurford, 1986). During this time, the drainage divide is probably located close to the Insubric line (e.g. Schlunegger et al., 1998), but north of the Bergell pluton (Fig. 8a), whose detritus is exclusively found in the retroforeland in the south (Gonfolite Lombarda, Giger and Hurford, 1989; Carrapa and Di Giulio, 2001).

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 (<30 My) zircon fission track ages (Spiegel et al. 2000) and the abundant flysch pebbles (Schlunegger et al., 1998), respectively. A potential contribution from ophiolites, as suggested by Spiegel et al. (2002), could be supported by the few eclogite-facies garnet grains found in the 19 My old sample. Their source could be located in the Penninic nappes in the hanging wall of the Rhone-Simplon line or in the Penninic nappes located between the Lepontine and the Austroalpine nappes (Fig. 8b). The Lepontine dome was probably drained both towards the north and the south (Fig. 8b), because old basement detritus with young cooling ages (~30 My, derived from K/Ar on white mica) was found in the Gonfolite Lombarda group in the southern retroforeland (Giger and Hurford, 1989).

Finally, the garnet data suggests a dominant contribution from the external massifs and/or the Gotthard nappe at around 14 My ago (Fig. 8c). We suggest that the drainage divide was essentially located at its current position (Fig. 8c, d), but this warrants corroboration from other deposits in the foreland and the retroforeland.

5. Conclusions

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 A~~a~~lpine 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~~ previous~~ly studies~~ (e.g., von Eynatten, 2003) and (2) to identify an

additional provenance shift between ca. 19 and 14 My ago that had not been noticed before. ~~The~~ This ~~latter shift before 14 My ago~~ shift is related to the erosion of granites from the external crystalline massifs, which provides a minimum age for their surficial exposure and corroborates their recently 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 exposed already ca. 14 My ago.~~

Author contribution

LS designed the project. AM helped during field work and sample collection. PT and PL gave advice 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.

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Table 1: Sample locations and characteristics of the Molasse sandstones from the Honegg-Napf fan. Abbreviations used: UFM = Upper Freshwater Molasse, UMM = Upper Marine Molasse, LFM = Lower Freshwater Molasse.

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

*Table 2: Sample locations and characteristics of potential source*s *(tributary sampling approach)* ~~*rocks*~~

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

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

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

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

	Mange & Morton (2007)				Tolosana-Delgado et al. (2018)		
Types after Mange & 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~~ 92 %	81 %	78 %
A-type (granulite-facies)	3 %	8 %		Granulites (Class C)		9 %	5.5 %
D-type (metasomatic)	1 %	3 %	5 %	Igneous (Class E)	~~75~~ %	3 %	12 %
				Mixed probabilities Classes B-C	*1 %*	*1 %*	
				Mixed probabilities Classes A-B-C		*5 %*	*4.5 %*

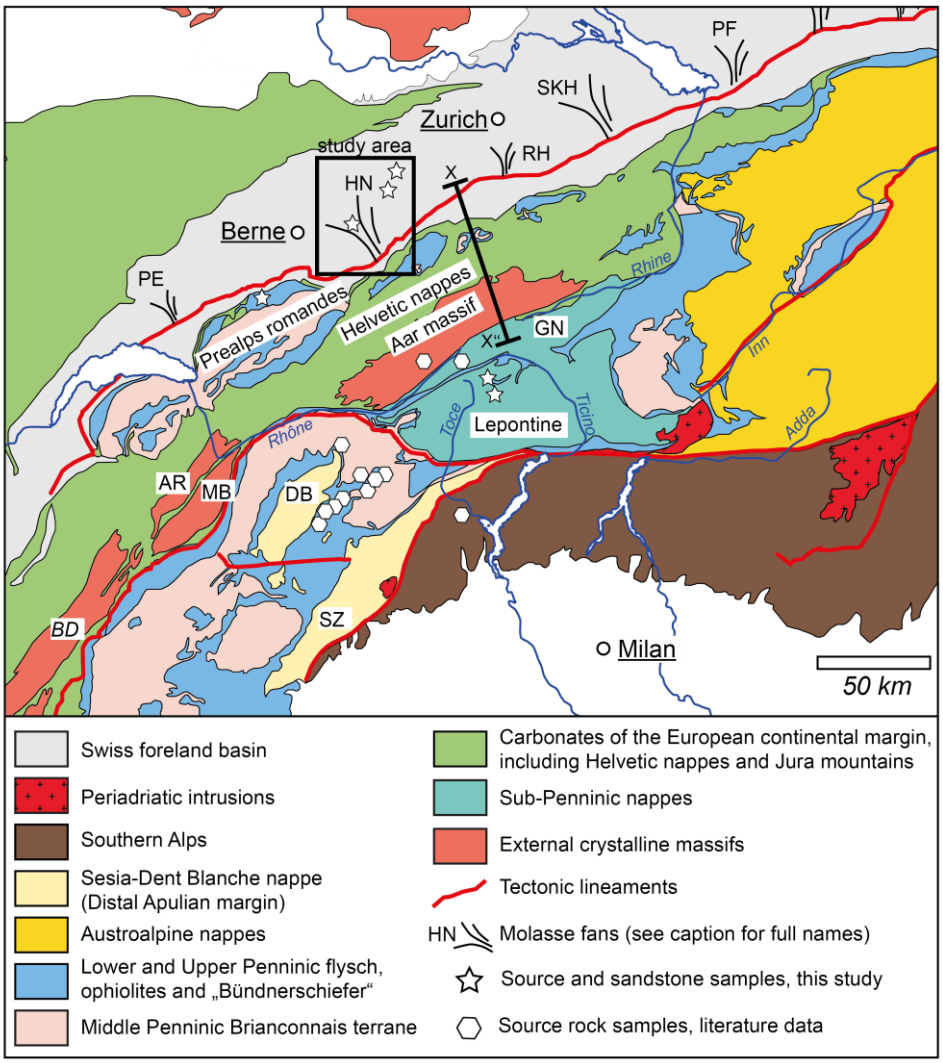

Fig. 1: Simplified tectonic map of the Central Alps after Schmid et al. (2004) highlighting the location of alluvial fan deposits within the northern ~~a~~Alpine foreland basin as well as the most important source rock units in the hinterland. The Honegg-Napf fan, marked by the black rectangle, is located in the central part of the Swiss foreland basin (SFB). For cross section X-X'' see Fig.~~ure 8~~7. Abbreviations used: AR = Aiguilles-Rouges massif, BD = Belledonne massif, DB = Dent Blanche nappe, HN = Honegg-Napf fan, MB = Mont Blanc massif, GN = Gotthard nappe, PE = Pèlerin fan, PF = Pfänder fan, ~~HN = Honegg-Napf fan,~~ RH = Rigi-Höhronen fan, SKH = Speer-Kronberg-Hörnli fan, SZ = Sesia zone.~~, PF = Pfänder fan.~~

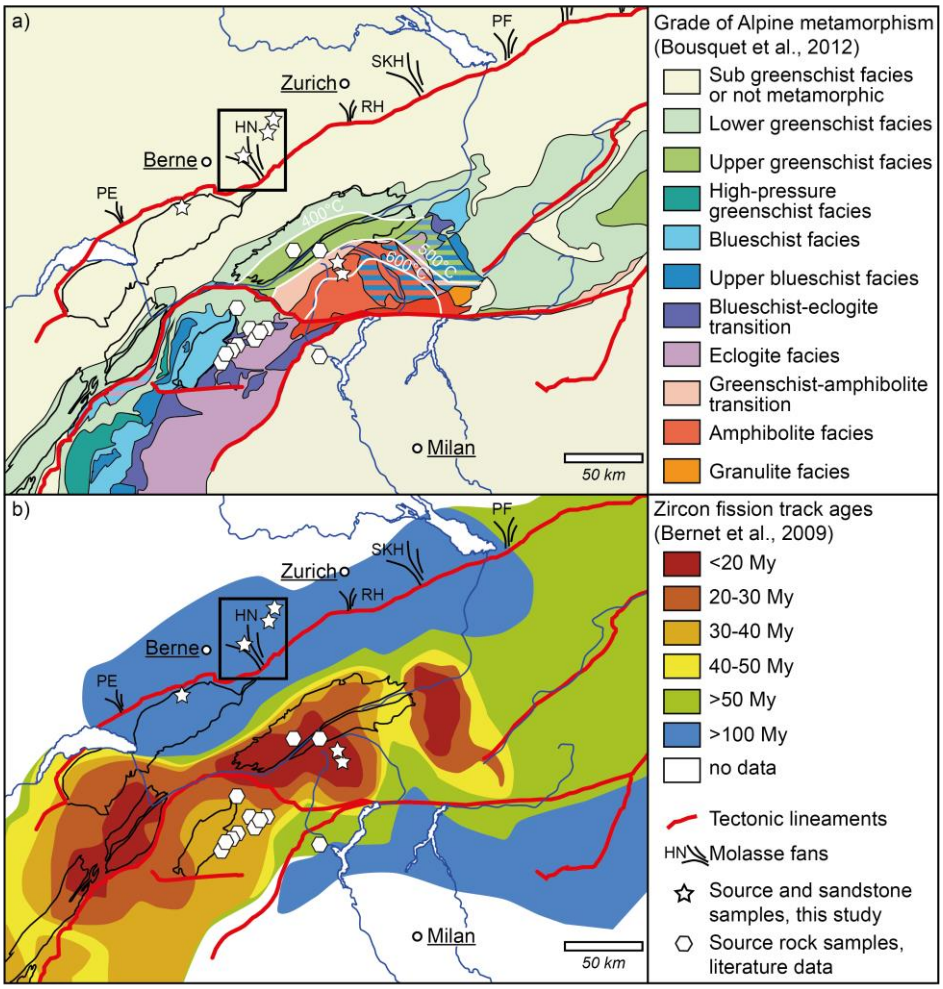

Fig. 2: a) Metamorphic map of the Central Alps (Bousquet et al., 2012) showing the distribution and grade of ~~alpine~~ Alpine metamorphism. Note the increase from north to south from lower greenschist- to eclogite-facies conditions. b) In-situ bedrock zircon fission track ages according to a compilation of Bernet et al. (2009). Note the predominantly young (<30 My) cooling ages in the area around the Lepontine dome and the external massifs in contrast to the predominantly old (>50 My) cooling ages 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 facilitate the orientation and the comparison with Fi~~gure~~. 1. Abbreviations used: PE = Pèlerin fan, HN = Honegg-Napf fan, RH = Rigi-Höhronen fan, SKH = Speer-Kronberg-Hörnli fan, PF = Pfänder fan.

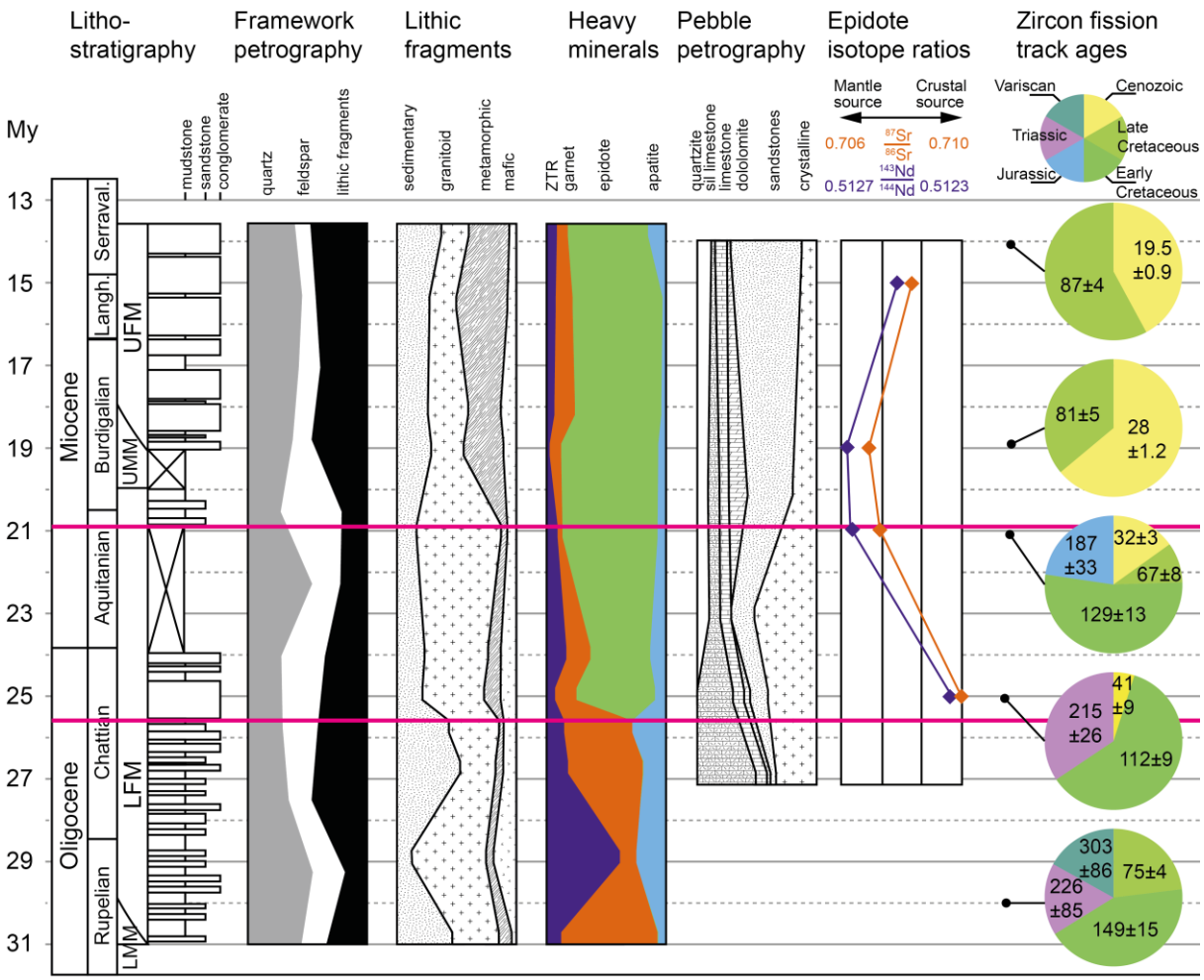

Fig. 3: Compilation of published compositional data in the Honegg-Napf fan. Heavy mineral and rock fragment data from the sand grain size after von Eynatten (2003), pebble petrography after Schlunegger et al. (1998), epidote isotope ratios after Spiegel et al. (2002) and zircon fission -track (FT) data after Spiegel et al. (2000). The two pink lines represent the dominant provenance changes as discusses in the text. Abbreviations used: LMM = ~~l~~Lower M~~m~~arine Molasse, LFM = L~~l~~ower ~~f~~Freshwater Molasse, UMM = ~~u~~Upper ~~m~~Marine Molasse, UFM = ~~u~~Upper ~~f~~Freshwater Molasse, ZTR = zircon-tourmaline-rutile-index, sil. = siliceous.

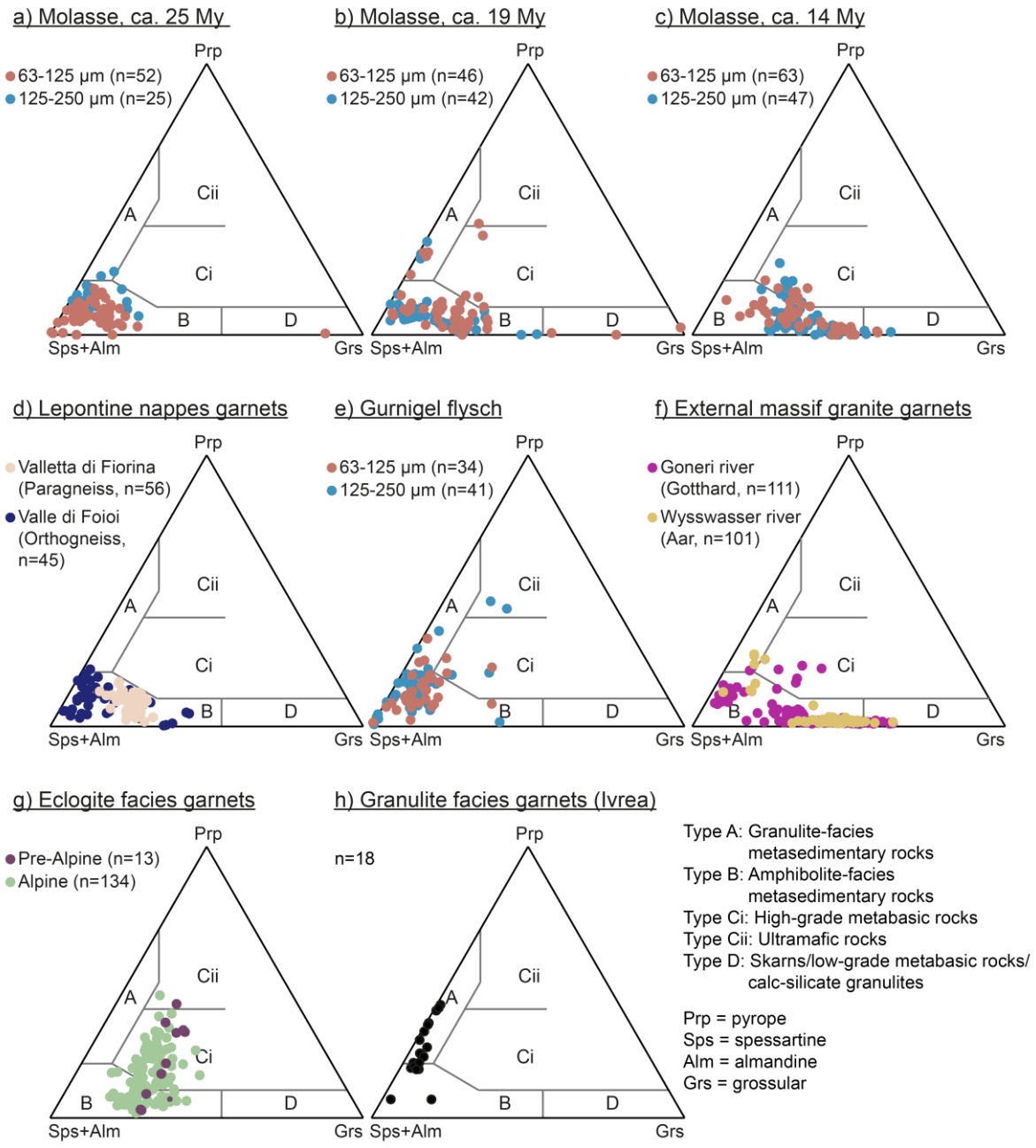

Fig. 4: ~~Ternary plots following~~Garnet ~~the~~ classification scheme of Mange & Morton (2007). (a-c) Detrital garnet compositions in the 25, 19 and 14 My-old Molasse deposits (this study). ~~Garnet provenance changes in Molasse sandstones are marked by an increasing grossular content with decreasing 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 & Dixon, 1973; Ernst & Dal Piaz, 1978; Oberhänsli, 1980; Sartori, 1990; Thélin et al., 1990; Reinecke, 1998; Cartwright & Barnicoat, 2002; Angiboust et al., 2009; Bucher & Grapes, 2009; Weber & Bucher, 2015), (h) granulite-facies rocks from the Ivrea zone in the Southern Alps (Hunziker & Zingg, 1980).

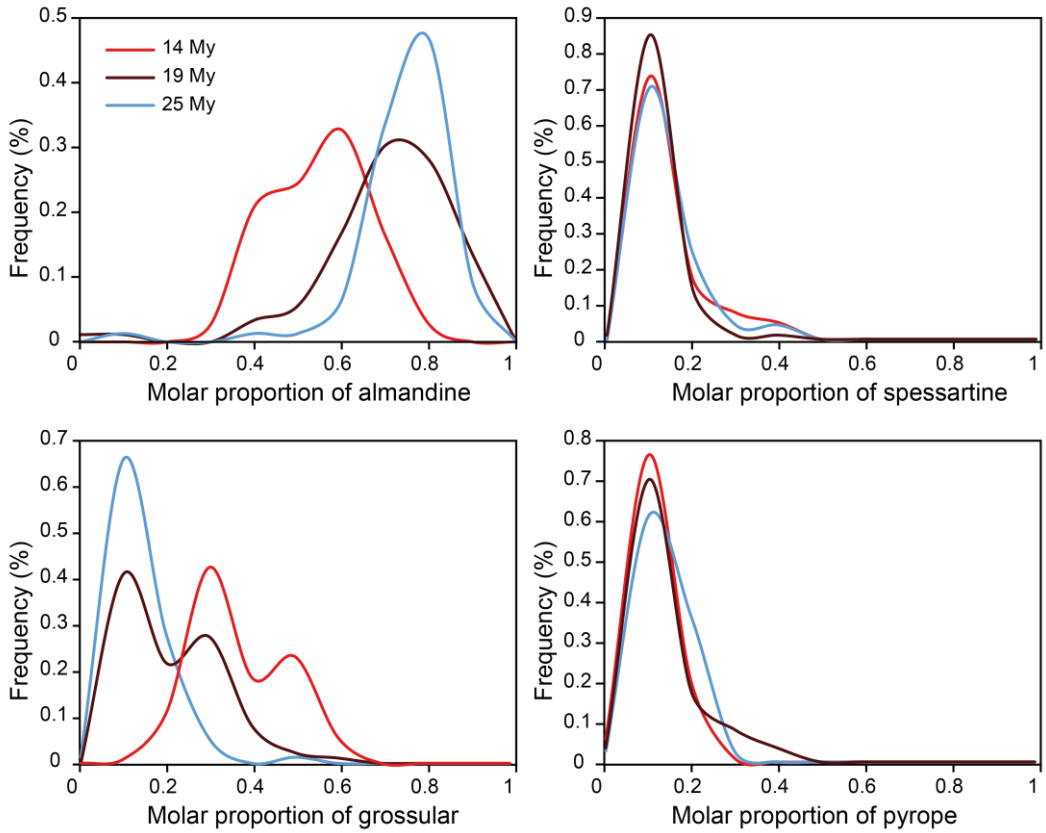

Fig. 5: ~~Shift of garnet compositions between the 25 My-, 19 My- and 14 My-old Molasse samples, plotted as r~~Relative frequency of the four most common endmembers almandine, grossular, spessartine and pyrope in the three detrital samples from the Molasse basin. ~~. While spessartine and pyrope contents are similar among the three samples, the proportion of almandine decreases and the proportion of grossular increases with decreasing age.~~

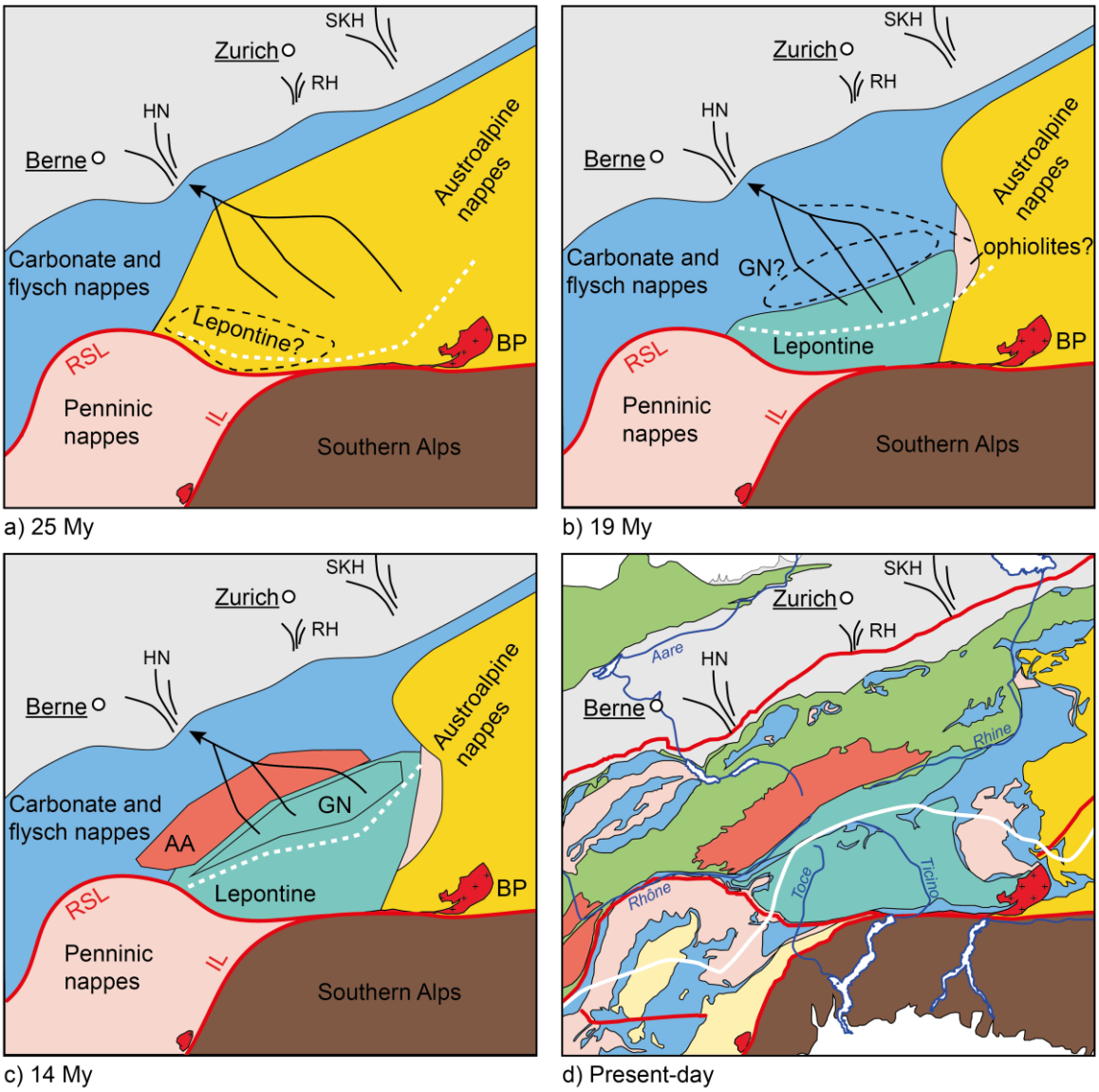

Fig.~~ure~~ ~~7~~6: Paleogeographic reconstruction of the Central Alps, and in particular of the hinterland of the Honegg-Napf fan. Situation during a) the late Oligocene (~25 My), b) the early Miocene (~19 My), c) the middle Miocene (~14 My) and d) today (after Schmid et al., 2004). The color coding in a-c) corresponds essentially to the color coding in d) (~~,~~see Fig. 1 for detailed legend). However, we have 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 color). ~~.~~Abbreviations used: AA = Aar massif, BP = Bergell pluton, GN = Gotthard nappe, HN = Honegg-Napf fan, IL = Insubric line, RH = Rigi-Höhronen fan, RSL = Rhone-Simplon lineament, SKH = Speer-Kronberg-Hörnli fan.

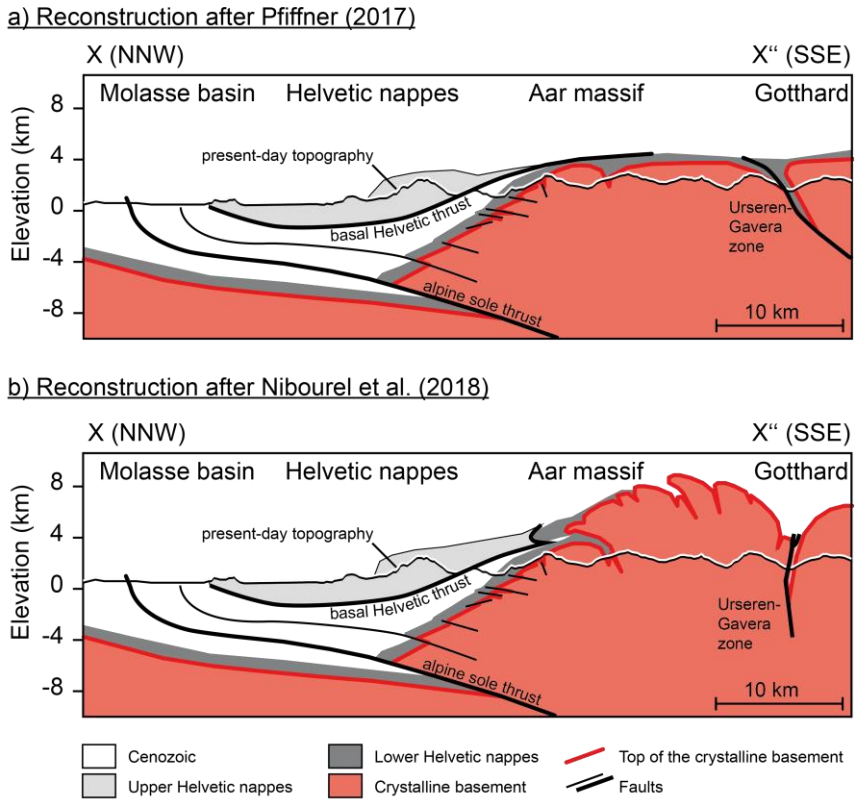

Fig. 7: Cross sections from X to X'' in Figu~~r~~.e 1 through the Aar massif simplified after Pfiffner (2017) and Nibourel et al. (2018). ~~For trace of cross section see Fig. 1.~~ (a): The reconstructed top of the crystalline basement in the Aar massif is located ca. 1-2 km higher than the present-day topography according to Pfiffner (2017). (b): In a revised version by Nibourel et al. (2018) the contact between the basement and the overlying Helvetic cover nappes is reconstructed to be steeper, resulting in ca. 8 km of (now eroded) crystalline crust on top of the present-day topography.