

RESEARCH ARTICLE OPEN ACCESS

Using the Dispersion of Apatite (U-Th-Sm)/He Single Grain Ages to Unravel the Burial and Exhumation History of the Foreland Basin of the Central Alps

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Received: 4 March 2025 | **Revised:** 28 July 2025 | **Accepted:** 29 July 2025

Funding: This work was supported by the Geological Institute, RWTH Aachen University and Nagra (National Cooperative for the Disposal of Radioactive Waste, Switzerland).

Keywords: (U-Th-Sm)/He thermochronometry | burial and exhumation | northern Swiss Molasse Basin | single grain ages | time–temperature modelling

ABSTRACT

Dispersed single-grain ages are a common phenomenon in detrital thermochronometry datasets that are often challenging to interpret. In this study, we showcase how a thermochronological forward modelling approach can be applied to such a complex dataset from the distal Swiss Molasse Basin. Despite extensive thermochronological research, the basin's exhumation history, magnitude and driving processes remain a subject of scientific debate. We present a large new apatite (U-Th-Sm)/He (AHe) dataset from a densely sampled deep exploration borehole extending beyond the actual Molasse basin fill into the underlying sedimentary sequence to provide more robust constraints on the exhumation history. AHe ages of over 100 grains range between 4 and 30 Ma in the upper 500 m and between 3 and 80 Ma below 1300 m, respectively. This is counterintuitive as, given the partial resetting of the shallow data, total reset would be expected at depths exceeding approximately 600 m. To arrive at a single consistent thermal history, we use a forward thermochronological modelling approach that allows us to test the influence of different provenance histories and distinguish between cooling phases associated with changes in heat flow vs. changes in exhumation. We find that a total of approximately 1100 m of Neogene exhumation, already starting at around 11 Ma, reconciles all available data. Exhumation continued throughout the Late Miocene but was initially not accompanied by significant cooling, suggesting a compensation by gradual increase of heat flow. At around 5 Ma, heat flow rises sharply towards the anomalously high present-day values of 120 mW m^{-2} . We argue that this discrepancy between the onset of exhumation and the onset of cooling may be responsible for previously differing estimates for the exhumation history of the basin. Furthermore, we infer geodynamic processes as the primary driving force for basin-wide exhumation, as it best explains its early onset.

1 | Introduction

Constraining the burial and exhumation history of foreland basins is important for a variety of research objectives. In general, the timing and magnitude of exhumation provide information on the underlying processes, in particular the influence

of climate, tectonic forces or mantle dynamics (e.g., Bernet and Spiegel 2004; Malusà et al. 2005; Glotzbach, van der Beek, et al. 2011; Von Hagke et al. 2012; François et al. 2014; Lacombe et al. 2019; Stalder et al. 2020). Furthermore, foreland basins serve as geological archives of the tectonic history of the orogen in their hinterland, providing valuable information about

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Summary

- We use apatite (U-Th-Sm)/He thermochronometry (AHe) and date more than 100 single grain ages and conduct thermal history modelling using the Python code PyBasin.
- The tool allows for the inclusion of provenance ages and to define heat flow over time, as well as burial and exhumation phases separately.
- We show that this is key for developing a single consistent model explaining the AHe dataset that reconciles previous studies and contributes to the discussion on exhumation drivers.

the late-stage development of a mountain range (e.g., Brewer et al. 2003; Bernet et al. 2009; Glotzbach, Bernet, et al. 2011; Anfinson et al. 2020). Additionally, foreland basins may contain hydrocarbon resources (e.g., Beaumont and Bout 1985; Beydoun et al. 1992; Gusterhuber et al. 2014), as well as potential targets for geothermal energy production (e.g., Wolfgramm et al. 2007; Moeck et al. 2015), and offer potential sites for radioactive waste disposal (e.g., Nagra 2014).

The most powerful tool to recover foreland basin evolution is stratigraphy. However, the stratigraphic record is incomplete in many of these basins around the world because of late-stage exhumation (Cederbom et al. 2004; Levina et al. 2014; Koshnaw et al. 2017). In these cases, low-temperature thermochronometry stands out as a powerful tool that can be used to recover the late stage of the foreland basin evolution (e.g., Bernet and Spiegel 2004; Spiegel et al. 2004; Mazurek et al. 2006; Van der Beek et al. 2006; Willett and Schlunegger 2010; Cederbom et al. 2011; Luijendijk et al. 2011; Von Hagke et al. 2012; Folguera et al. 2015; Tatzel et al. 2017). The method can provide insights into landscape evolution, including relief development through time (e.g., Ehlers and Farley 2003; Braun et al. 2012; Glotzbach, van der Beek, et al. 2011; Valla et al. 2012), and tectonic processes operating on basins at local or orogenic scale (e.g., Ehlers and Farley 2003; Reiners and Brandon 2006; Mock et al. 2020; Von Eynatten et al. 2021).

Typically, thermochronometric ages of individual grains from the same sampling depth exhibit a spread (i.e., are dispersed), which would not be expected assuming all grains from the same sampling depth and formation witnessed the same temperature history. In the last decade, several studies have addressed this dispersion problem of low-temperature thermochronological data (Brown et al. 2013; Green and Duddy 2018; Fox et al. 2019). The most common reasons for dispersion are the variable pre-depositional burial and exhumation histories (inheritance) of the single grains from the same sample set. This includes sediment recycling expected in many tectonic settings, such as foreland basins (e.g., Fosdick et al. 2015; Flowers, Ketcham, Enkelmann, et al. 2022). Additionally, the thermal history a grain witnessed can be influenced by other factors that are independent from late-stage burial and exhumation, such as changes in heat flow, the local affection by hydrothermal fluids, or even heat from friction during faulting or wildfires (Brown et al. 2013; Green and Duddy 2018; Fox et al. 2019; Flowers,

Ketcham, Enkelmann, et al. 2022). Beyond these exterior influences on single-grain thermochronometric ages, factors concerning properties of the individual grains can also lead to dispersion of apatite (U-Th-Sm)/He (AHe) ages measured from a specific sample set. These are helium diffusion kinetics, crystal zonation or authigenic crystallisation of apatites from hydrothermal fluids (Flowers et al. 2009; Ault and Flowers 2012; Cecil et al. 2014; Djimbi et al. 2015; Fox et al. 2019; Berger et al. 2022), as well as helium implementation from neighbouring crystals (e.g., Murray et al. 2014). Given the variable factors that contribute to the dispersed distribution of AHe ages, corresponding datasets represent challenges for geological interpretation and modelling. Despite these challenges, dispersed datasets also offer opportunities, for example, to determine different single-grain age populations and thereby derive individual time–temperature histories (e.g., Fillon et al. 2013; Cecil et al. 2014; Fox and Shuster 2014).

In this contribution, we present an extensive new detrital apatite (U-Th-Sm)/He data set from a deep exploration borehole in the distal part of the northern Swiss Molasse Basin (Figure 1), leveraging the observed dispersion of single-grain ages. This basin has been intensively investigated previously, but the exhumation magnitude estimated on the basis of thermochronometric data varies widely from 750 to 1600 m (e.g., Mazurek et al. 2006; Cederbom et al. 2011). The onset of the cooling signal related to exhumation is also debated. Existing models show that exhumation could have started as early as 12 Ma or as late as 4 Ma (Von Hagke et al. 2015). Furthermore, the Molasse basin lacks sufficient (U-Th-Sm)/He data to constrain the cooling history at temperatures below approximately 60°C.

The dataset presented herein contains grains from a well-constrained stratigraphic sequence and is derived from differing source areas (Figure 1). This provides an opportunity to cover an extensive range of age populations, significant for explaining the late-stage basin thermal history. Following the analysis of the raw data and exclusion of solely grain size or grain chemistry being responsible for the age dispersion (Section 4.2), we apply a thermal forward modelling approach to reconcile the different previous estimates for exhumation of the Alpine foreland and constrain the underlying driving forces.

2 | Geological Setting

2.1 | Basin Evolution and Sediment Provenance

The foreland basin of the Central Alps, that is, the Swiss Molasse Basin (Figure 1), has witnessed a polyphase tectonic history. Immediately to the north of the basin, the crystalline foreland basement is exposed in the BF, which is characterised by deformation processes related to the Late Palaeozoic Variscan orogeny (Eisbacher et al. 1989; Echtler and Chauvet 1992). Widespread plutonism occurred during the latest collisional phase of the latter between approx. 340 and 330 Ma (Schaltegger 2000). The collision phase was followed by extension in Late Carboniferous and Permian times. Graben and half-graben structures from this period are reported in large parts of Europe (McCann et al. 2008). In the study area, the so-called Constance-Frick Trough locally

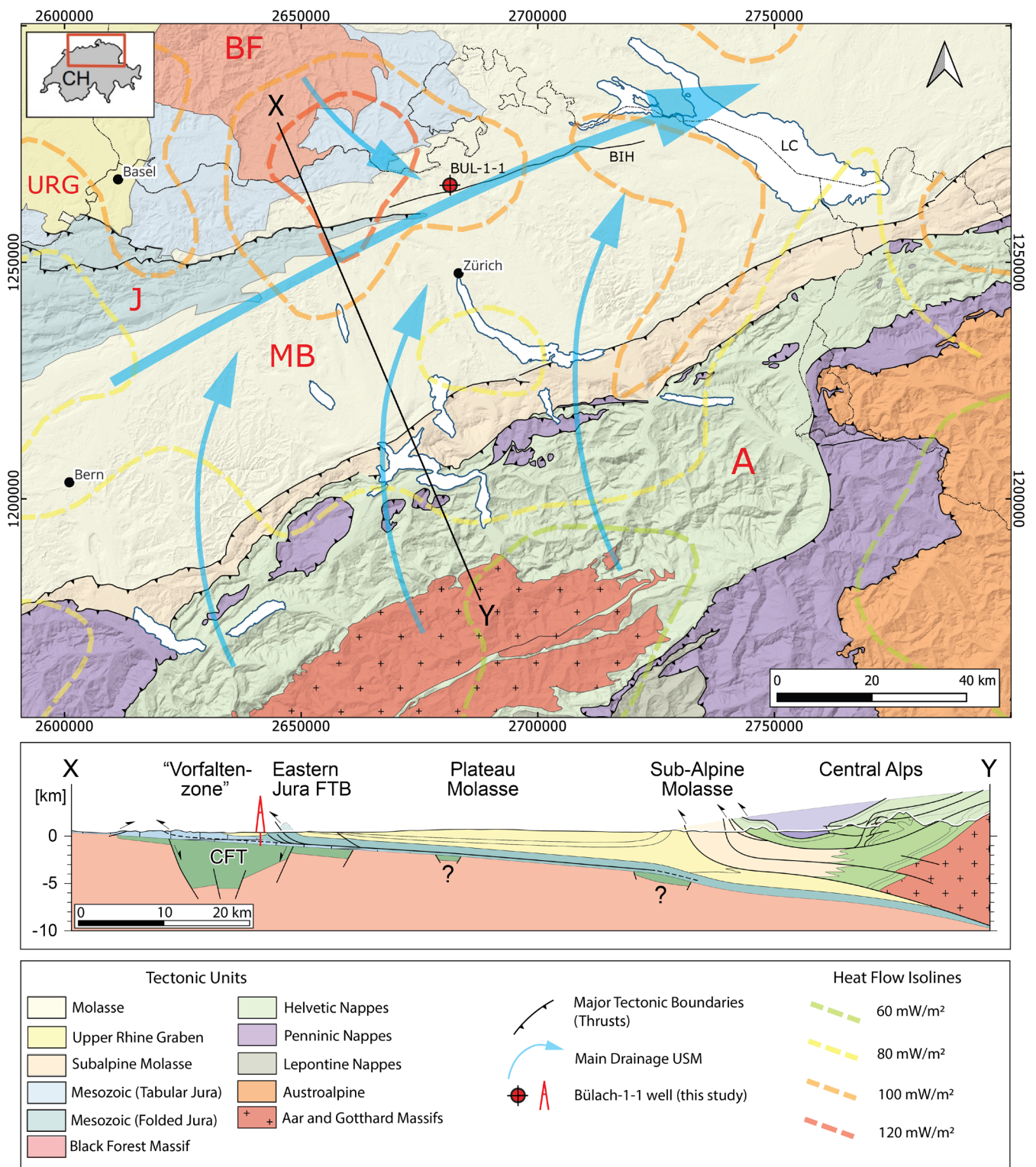


FIGURE 1 | Simplified tectonic map of the study area in northern Switzerland, including a cross-section with the projected position of the Bülach-1-1 well (modified from Madritsch et al. 2024). Drainage directions of the time of USM (Lower Freshwater Molasse, see Section 2.1) deposition ~22 Ma are indicated (blue arrows, after Kuhlmann and Kempf 2002). Heat flow isolines indicate interpolated recent heat flow (simplified after Medici and Rybach 1995). BIH, Baden-Irchel-Herdern Lineament; CFT, Constance-Frick Trough; FTB, Fold-and-Thrust Belt; LC, Lake Constance. Tectonic domains labelled in red: Molasse Basin (MB), Upper Rhine Graben (URG), Alps (A), Black Forest Massif (BF) and Jura Mountains (J).

reaches depths of roughly 5 km (Figure 1; Diebold et al. 1991; Madritsch et al. 2018). The largest part of the trough fill consists of Permian clastic sediments that are Variscan-derived from rocks eroded from the trough shoulders (Matter et al. 1988). This sedimentary sequence is cut by a regional unconformity. Following this Permian–Triassic unconformity, transgression from the north led to the deposition of sandstones, after which a sedimentary sequence of an

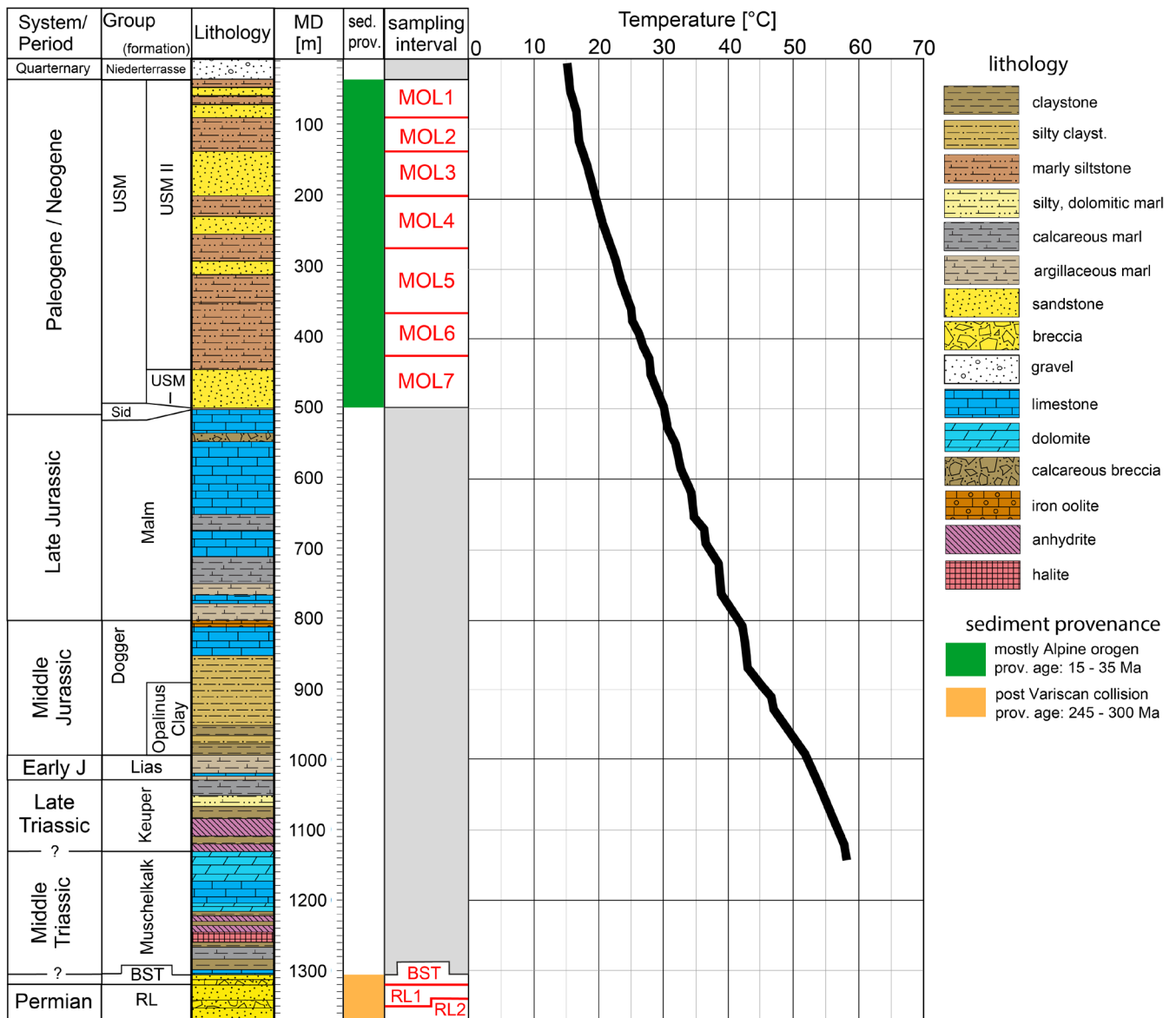


FIGURE 2 | Lithostratigraphic log of the BUL-1 well (modified and simplified after Nagra 2021) and measured borehole temperatures (Nagra 2021). Sediment provenance (source and provenance age) is indicated in green (Alpine, 15–35 Ma) and orange (post-Variscan, 245–300 Ma). Sampling intervals and depth ranges are indicated and named MOL1—MOL7 (MOL: “Molasse”), BST (“Buntsandstein”), RL1 and RL2 (RL: “Rotliegend”).

epicontinental marine environment was deposited (Geyer et al. 2011). The first siliciclastic sediments, belonging to the Lower Triassic Dinkelberg Formation, were probably derived from the continent to the south, reworking Variscan basement rocks of the coastal area. Subsequently, Triassic marls, evaporites and occasional lime- and sandstones are overlain by a thick sequence of Early to Mid-Jurassic claystones, including the Opalinus Clay Formation (Nagra 2014). The latter is overlain by predominately calcareous rocks of Upper Jurassic age that are cut by another regional unconformity, which has been associated with the northward migration of the forebulge of the Alpine foredeep in Late Cretaceous to Paleogene times (Crampton and Allen 1995; Bourgeois et al. 2007; Ziegler and Dèzes 2007). On the basis of vitrinite reflectance and apatite fission track studies, the original thickness of the eroded Cretaceous sediments has been estimated to have ranged from 0 to 1400 m (Schegg and Leu 1998; Mazurek et al. 2006). As a

result of this Mesozoic burial history, the sediments sampled from Permian and Triassic rocks in the context of this study (see Section 2.4 and Figure 2) possibly have resided at shallow depths (i.e., at temperatures mostly lower than 100°C) for most of their post-depositional history. Tectonic activity during the Mesozoic was apparently only subtle, although some extensional reactivation of pre-existing faults may have occurred locally in Triassic and Early Jurassic times (Wetzel et al. 2003; Marchant et al. 2005; Roche et al. 2020). Unfortunately, the entire Mesozoic sequence is characterised by lithologies of very rare apatite occurrences, which strongly limits the applicability of thermochronological dating techniques (Mazurek et al. 2006).

From Eo-Oligocene times onwards into the Miocene, the northernmost Alpine foreland was affected by the evolution of the European Cenozoic Rift System (Hinsken et al. 2007;

Ziegler 1992). Most important for the study area, the opening of the Upper Rhine Graben (URG) resulted in flank uplift in the area of the future BF and deposition of the eroding rocks into the Molasse Basin (MB, Figure 1). The formation of the latter is primarily related to the Alpine orogeny involving the subduction of the oceanic part of the European plate under the Adriatic continental plate and subsequent continent-continent collision after approximately 35 Ma (Pfiffner 1986; Schmid et al. 1996; Sinclair et al. 1991; Burkhard and Sommaruga 1998; Schlunegger et al. 2007). The stratigraphy of the foreland basin is well-established (e.g., Kempf et al. 1997; Schlunegger et al. 1997; Kuhlemann and Kempf 2002; Berger et al. 2005b) and has been studied intensely regarding the link between Alpine mountain building and foreland basin evolution (Sinclair and Allen 1992; Schmid et al. 1996; Sinclair 1997; Mosar 1999; Pfiffner et al. 2002; Schmid et al. 2004; Bousquet et al. 2008; Handy et al. 2010; Willett and Schlunegger 2010; Von Hagke et al. 2012; Hinsch 2013; Ortner et al. 2015; Schlunegger and Kissling 2015; Schlunegger and Castelltort 2016). The sedimentary sequence is separated into two regressive megacycles (Matter et al. 1980; Kuhlemann and Kempf 2002). The lower and upper cycles of the Molasse sediments in this area are subdivided into the Untere Meeresmolasse (UMM, “Lower Marine Molasse”, ca. 31.5–30 Ma (Kempf et al. 1997; Kempf et al. 1999)), Untere Süßwassermolasse (USM, “Lower Freshwater Molasse”, ca. 30–21 Ma, locally until 19 Ma (Kempf et al. 1997, 1999)), Obere Meeresmolasse (OMM, “Upper Marine Molasse”, 21–17 Ma (Bolliger 1999)), and Obere Süßwassermolasse (OSM, “Upper Freshwater Molasse”, 17–13 Ma (Kempf et al. 1997, 1999)). These sediments derive from the emerging Alpine chain, as well as the BF in the northernmost part of the basin (Figure 1, e.g., Kuhlemann and Kempf 2002). Hence, their inherited cooling age (provenance age) prior to burial in the Molasse Basin is expected to be much younger (roughly 15 Ma to 35 Ma) compared to the Permian and Triassic sediments below (roughly 245 to 300 Ma, Figure 2).

The southern part of the Molasse Basin was affected by thrusting of the Alpine orogen in early Miocene times, shortly after the sediments were deposited (Schlunegger et al. 1997; Kempf et al. 1999), and deformation continued until the late Miocene (Von Hagke et al. 2012). At approximately 14 Ma, deformation of the Jura fold-and-thrust belt started as a result of thin-skinned propagation of the Alpine front into the foreland (Laubscher 1978; Burkhard and Sommaruga 1998; Looser et al. 2021). The main folding phase started after about 12 Ma (Sommaruga et al. 1997; Becker 2000; Madritsch et al. 2024). The most prominent structure related to the Alpine foreland deformation in the vicinity of the investigated BUL-1 borehole is the Baden-Irchel-Herdern Lineament (BIH, approx. 1 km to the south of BUL-1; Figure 1, Nagra 2014). It has been interpreted as a middle Miocene normal fault that was inherited from a Permo-Carboniferous precursor structure and later overprinted by late Miocene shortening (Malz et al. 2016). Also, from the middle Miocene onwards, different deformation processes occurred immediately east of the study area, where crustal-scale transtension led to the formation of the Hegau–Lake Constance graben (Egli, Mosar, et al. 2017; Madritsch et al. 2024). This area was also affected by volcanism (Hegau volcanoes), with ages ranging from 15 to 7 Ma (Schreiner 1992). Apatite fission track ages from related ash layers in OSM sediments allowed constraining the

youngest Molasse deposition ages to younger than 11.5, and potentially younger than 9.8 Ma (Rahn and Selbekk 2007; Binder et al. 2023).

2.2 | Late-Stage Basin Exhumation

Different methods have been applied to study the basin's late-stage history, including vitrinite reflectance (Schegg and Leu 1998; Mazurek et al. 2006), wedge mechanics (Willett and Schlunegger 2010; Schlunegger and Mosar 2011; Von Hagke, Oncken, and Evseev 2014) or low-temperature thermochronometry (Mazurek et al. 2006; Cederbom et al. 2011; Von Hagke et al. 2012; Von Hagke, Oncken, Ortner, et al. 2014; Mock et al. 2020). These investigations provided evidence for substantial uplift and exhumation during the latest Neogene (e.g., Schegg and Leu 1998; Cederbom et al. 2004; Mazurek et al. 2006; Willett and Schlunegger 2010; Cederbom et al. 2011; Schlunegger and Mosar 2011; Baran et al. 2014). The extent, timing and driving mechanisms of this exhumation have been a scientific debate ever since. Studying the northern rim of the basin in the close vicinity of the Jura Mountains (Figure 1), Mazurek et al. (2006) argued for 750 to 1050 m of exhumation starting at around 9 Ma. On the contrary, Cederbom et al. (2011), who also included borehole data from the more proximal part of the basin, suggested exhumation of 800 m to 1600 m since approximately 5 Ma. Compiling and interpreting existing data, the onset of exhumation could have been as early as 12 Ma to late-stage exhumation, commencing at 4 Ma (Von Hagke et al. 2015). Drivers that have been suggested for the exhumation include climatic changes (Cederbom et al. 2004; Champagnac et al. 2009), tectonic forcing (Burkhard and Sommaruga 1998; Persaud and Pfiffner 2004; Reinecker et al. 2008), river capture (Kuhlemann and Rahn 2013; Yanites et al. 2013), mantle processes (Schlunegger and Castelltort 2016) or a combination thereof (e.g., Cederbom et al. 2011; Schlunegger and Mosar 2011; Von Hagke et al. 2012).

2.3 | Heat Flow History

Geothermal gradients were presumably high during the Variscan collisional phase, with values ranging between 50 and 130°C/km (Mazurek 1992, 1998). In Mesozoic times, heat flow was likely initially lower (around 70 mW/m²). However, on the basis of thermokinematic modelling and vitrinite reflectance data, several authors have suggested a heating event in Late Jurassic to Early Cretaceous times, accompanied by hydrothermal activity (Timar-Geng et al. 2004; Mazurek et al. 2006; Omodeo-Salé et al. 2021; Villagómez Díaz et al. 2021). Peak heat flow values of about 90 mW/m² are proposed during the Early Cretaceous. The development of heat flow during the tectonically complex Cenozoic evolution of the Molasse Basin is considered to have been dynamic. Initially, high heat flow values have presumably decreased below 70 mW/m² before Miocene times, with geothermal gradients ranging between 15 and 30°C/km (Mazurek et al. 2006; Schegg and Leu 1998). From the Miocene onwards, previous thermochronological models suggested an increase of heat flow to over 100 mW/m² (e.g., Mazurek et al. 2006), peaking in recent times at around 120 mW/m² in the study area

(Medici and Rybach 1995). Indeed, post-glacial hydrothermal fluid flow also affected the region of the northern Swiss Molasse Basin (Sonney and Vuataz 2008). This hydrothermal activity results in heat flow anomalies throughout the basin (Figure 1). Two examples from the study area are the geothermal springs of Baden and Schinznach (easternmost Jura Mountains, Luijendijk 2019).

2.4 | The Bülach-1 Well

The Bülach-1 well (hereafter referred to BUL-1) is located in the distal part of the Molasse Basin, immediately north of the main thrust of the Jura fold-and-thrust belt (Figure 1; Madritsch et al. 2024). The vertical well was drilled on behalf of the National Cooperative for Radioactive Waste Disposal (Nagra) and covers 1370 m of a sub-horizontally layered sedimentary sequence (Figure 2, Nagra 2021). In the following, we briefly present the drilled strata, which are described in detail by Nagra (2021). From bottom to top, the drilled stratigraphy starts with 50 m of siliciclastic Permian Rotliegend sediments belonging to the Weitenau Formation (Figure 2). This formation consists mainly of sandstones with thin claystone intercalations and breccias that were deposited in the Constance-Frick Trough (Diebold and Noack 1997; Madritsch et al. 2018). Lower Permian sediments are capped by the regional upper Permian unconformity. The succession from Lower Triassic to Upper Jurassic strata is completely recovered. 816 m of Mesozoic deposits were drilled, including Triassic siliciclastic rocks, carbonates and evaporites, while being dominated by Jurassic limestones, claystones and marls in the upper part. The regional unconformity on top of the Malm section is marked by approximately 2 m thick Siderolithikum, a reddish-brown claystone with abundant iron-pisoids of Eocene age. The Molasse section starts with 54 m of USM I sandstones, siltstones and silty marls, topped by 448 m of alternations of silty to sandy marls, sandstones, siltstones and argillaceous marls, resembling the USM II formation. Stratigraphically higher Molasse sediments (OMM and OSM) were not drilled because of the location of the well in a river valley. However, these formations are exposed at the surface within a radius of 2 km around the well (Haldimann et al. 2017). The top 34 m of the well comprise Quaternary sandy gravels.

3 | Methods

3.1 | (U-Th-Sm)/He Thermochronometry, Sampling and Measurements

To determine the timing and magnitude of exhumation of the region, we used apatite (U-Th-Sm)/He (AHe) thermochronometry. This method makes use of the decay of radioactive isotopes (^{238}U , ^{232}Th and ^{147}Sm) and the temperature-dependent retention of the decay product (^4He) in apatite crystals (Zeitler et al. 1987; Wolf et al. 1996; Farley 2000; Flowers 2009). The low-temperature sensitivity (closure temperature) of 40°C to 80°C (Zeitler et al. 1987; Wolf et al. 1996) makes AHe thermochronometry a suitable method to understand the cooling of the uppermost 2–3 km of the crust. Exhumation can be

estimated on the basis of the concept of partial retention zones (PRZ, Figure 3a, Fitzgerald et al. 2006). The term refers to the partial retention of helium in the crystal lattice at temperatures just below the closure temperature. Such temperatures are too high to retain all helium in the crystal, but too low for a complete removal over the time it resides in this temperature window. Consequently, because of the geothermal gradient, a vertical section of AHe ages will show a total reset at depths with temperatures well above the closure temperature and a gradual increase in measured AHe ages to shallower depths in the partial retention zone (PRZ) towards helium ages representing complete retention of helium (Figure 3a). For detrital samples, a slight younging towards shallower depths above a PRZ can be expected. This is because provenance ages tend to decrease to the top of the sedimentary section, as the younger sediments have been exhumed and eroded in the orogen more recently. With the onset of rapid cooling, the pattern of a partial retention zone can be preserved when the whole section cooled below the closure temperature of the apatite helium system (exhumed PRZ, Fitzgerald et al. 2006).

We collected samples from the siliciclastic Miocene USM ("Molasse"; MOL), the Triassic Dinkelberg Formation ("Buntsandstein"; BST) and the Permian Weitenau Formation ("Rotliegend"; RL), as these lithologies are possibly apatite bearing (Lucas and Prevot-Lucas 1997). In total, our samples span a vertical distance of 1,370 m. For the BST-RL section, we collected pieces of drill cores over three intervals of roughly 25 m each, guided by internal stratigraphic boundaries. These three intervals are called BST, RL1 and RL2, respectively (Figure 2). We selected fine-grained sandstone sections of the core of about 5 to 30 cm in length. From each interval, we sampled at least 1 meter of half cores. From the Molasse, only cuttings were available for sampling, which were washed and dried at 20°C . We subdivided the Molasse into seven intervals (MOL1—MOL7, Figure 2) on the basis of stratigraphic boundaries, with one sampling interval covering approximately 75 m.

We crushed all samples to release single grains and sieved the crushed material to obtain the 63–250 μm fraction. With this fraction, we performed heavy liquid separation with a sodium-polytungstate solution at 2.82 g/cm^3 . After investigation of the samples with a microscope, we re-separated the MOL samples with a heavy liquid of 3.0 g/cm^3 density. The heavy fraction entered magnetic separation using an Isodynamic Magnetic Separator (Model L-1, S. G. Frantz Co. Inc.). The red sandstone grains of the RL samples were covered by a thin coating of iron minerals. We removed the coating chemically using dithionite (after Tributh and Lagaly 1986). We selected apatites for dating from the final separates with a binocular. The pre-selected grains were further investigated with a transmitted light polarisation microscope with magnifications of $5\times$ to $50\times$. We checked for inclusions by immersing the grains in ethanol. Grain dimensions were measured by taking scaled images of the grains.

Apatite picking, as well as helium extraction and parent isotope measurements, has been carried out at the GÖOchron Laboratories at the University of Göttingen (analytical procedure in Von Eynatten et al. 2021). We measured 45 single grains from the Molasse section and 61 grains from the deeper BST and RL intervals, as precisely listed in Tables 1a and 1b.

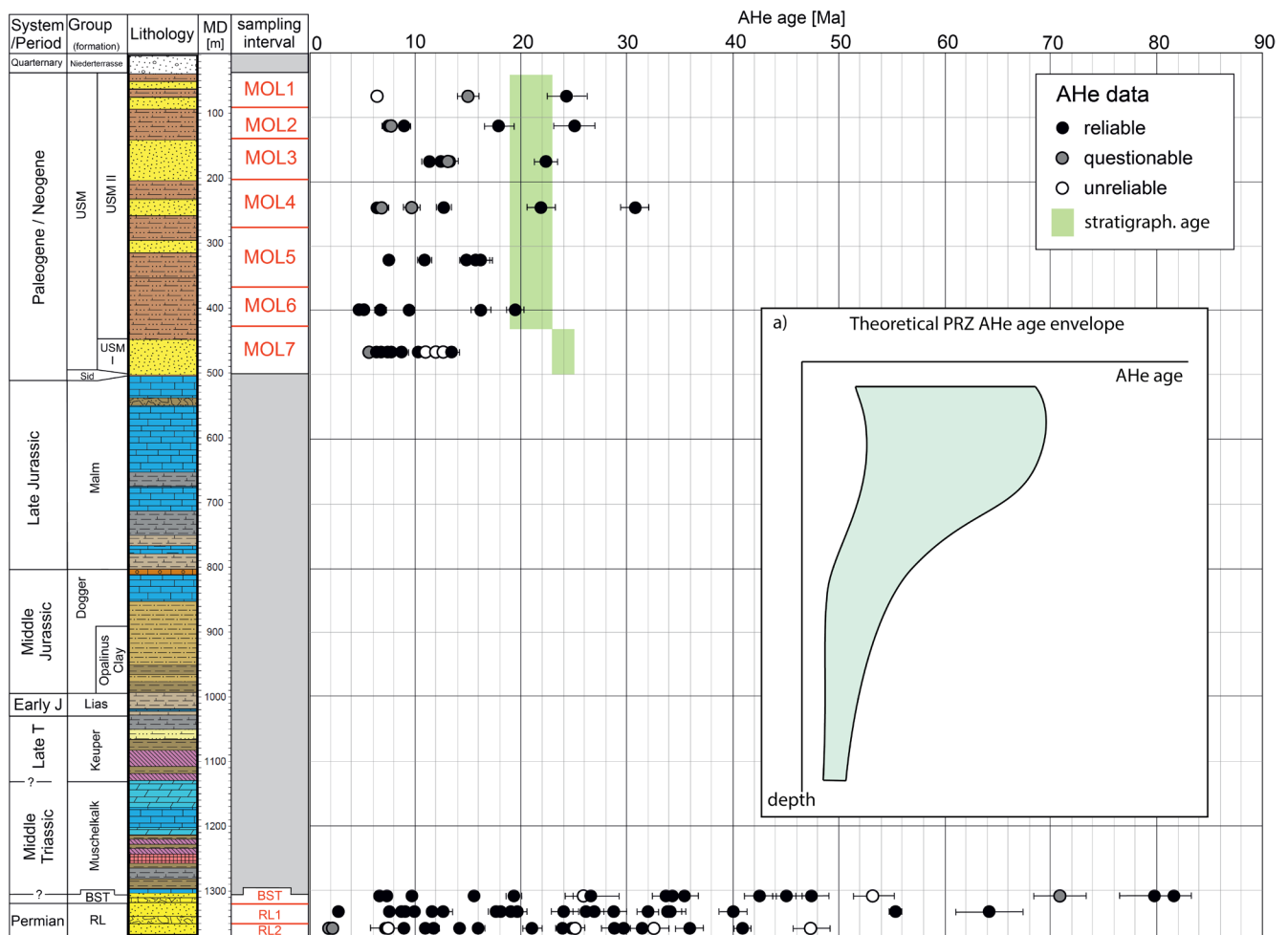


FIGURE 3 | Measured AHe data from the BUL-1 well plotted against sampling depth. Sampling intervals and depth ranges are indicated and named MOL1—MOL7 (Molasse sampling intervals), BST (Buntsandstein sampling interval), RL1 and RL2 (Rotliegend sampling intervals). Error bars represent 2σ on AHe ages, including analytical uncertainty and estimated uncertainty of F_T correction. Light green planes indicate stratigraphic ages. The stratigraphic age range of BST is 249 to 244 Ma, and 270 to 260 Ma for RL (Kämpfen 1972). Colours of data points indicate grains with analytical errors $> 10\%$, $eU < 10$ ppm, first He re-extracts $> 4\%$ or unrealistically high AHe ages (white, not reliable), analytical errors $> 7.5\%$, $eU < 15$ ppm, first He re-extracts $> 3\%$ (grey, questionable), others are called “reliable” (black). See Figure 2 for a lithologic legend. (a) theoretical shape of a partial retention zone (PRZ) in a sedimentary basin setting (or exhumed partial retention zone after Fitzgerald et al. (2006)). All single-grain ages should plot within the greenish-coloured area. Although MOL1–7 samples possibly resemble this pattern, BST, RL1 and RL2 AHe ages contradict a single PRZ interpretation.

In total, we dated 107 grains separated into 10 sampling intervals. We set up three classes on the basis of the reliability of single-grain ages. (1) Unreliable ages are associated with high analytical errors ($> 10\%$, all values are smaller), low uranium concentration (< 10 ppm), substantial amounts of helium measured on the first re-extract ($> 4\%$, in percent of the first extraction) and improbably old ages (concerning one grain with an AHe age > 400 Ma) that are most likely the result of parent isotope-rich insoluble inclusions (white points in Figure 3, bold font in Tables 1a and 1b). (2) Questionable ages of grains with analytical errors $> 7.5\%$, uranium concentrations < 15 ppm and first helium re-extracts $> 3\%$ (grey points in Figure 3, italic font and underscored in Tables 1a and 1b). (3) Reliable ages are all grains above these thresholds. This categorization is meant to assist in interpreting outliers or misfits in case a single unreliable or questionable grain contradicts modelling results. The categorization does not lead to the exclusion of grains from modelling and interpretation. We note that F_T

corrections range between 0.47 and 0.88 (Tables 1a and 1b). Errors associated with F_T corrections are included in thermokinematic models.

3.2 | Modelling

To constrain the thermal history, we applied different modelling approaches. The commonly used modelling tools HeFTy (Ketcham 2005) and QTQt (Gallagher 2012) have already been discussed methodologically and compared to each other in previous studies (Vermeesch and Tian 2014, 2018; Gallagher and Ketcham 2018). We also used both of these modelling tools to analyse our dataset (Supporting Information S1–S5, Frings 2023). Herein, we focus on modelling using the tool PyBasin (Luijendijk et al. 2011; Luijendijk 2020). It follows a forward modelling approach, where the burial history is determined by back-stripping and decompaction of the

TABLE 1a | Results of apatite (U-Th-Sm)/He dating for the Molasse samples from the BUL-1 well.

Sampling interval	Depth (m)	Raw			Corrected age (Ma)	Error (Ma)	Analytical error (%)	U (ppm)	Th (ppm)	Sm (ppm)	He (ncc)	eU (ppm)	Th/U	Sphere radius (μm)
		Grain	age (Ma)	F_T										
MOL1	68	Mol-1 a1	12.2	0.50	24.4	1.9	4.4	30	4	233	0.041	31	0.14	36
		Mol-1 a2	4.4	0.69	6.4	0.4	3.4	9	42	381	0.029	18	4.83	54
		<u>Mol-1 a3</u>	9.5	0.63	15.0	1.0	4.2	<u>14</u>	25	213	0.034	20	1.79	44
MOL2	115	Mol-2 a1	12.8	0.51	25.0	1.9	3.3	48	59	112	0.093	62	1.24	35
		Mol-2 a2	5.0	0.66	7.7	0.5	3.9	27	40	196	0.025	36	1.49	49
		<u>Mol-2 a3</u>	4.3	0.55	7.8	0.9	<u>9.4</u>	<u>14</u>	39	329	0.010	23	2.75	41
		Mol-2 a4	9.3	0.52	17.9	1.3	3.0	30	91	378	0.059	52	2.97	36
		Mol-2 a5	5.6	0.62	9.0	0.5	3.2	24	45	229	0.046	35	1.87	48
MOL3	171	Mol-3 a1	16.2	0.72	22.4	1.1	2.4	53	67	413	0.379	69	1.25	56
		Mol-3 a2	8.8	0.77	11.4	0.7	5.4	15	32	239	0.040	23	2.15	62
		Mol-3 a3	8.7	0.70	12.5	0.6	2.7	32	45	169	0.097	43	1.38	55
		Mol-3 a4	7.7	0.59	13.2	0.9	2.9	32	119	622	0.088	60	3.75	42
		<u>Mol-3 a5</u>	11.0	0.83	13.2	0.8	5.3	<u>15</u>	21	246	0.035	20	1.43	79
MOL4	241	Mol-4 a1	4.2	0.66	6.4	0.4	2.8	24	78	497	0.027	42	3.23	46
		<u>Mol-4 a2</u>	5.5	0.56	9.8	0.7	5.1	<u>15</u>	31	238	0.011	22	2.13	39
		Mol-4 a3	7.7	0.61	12.7	0.7	2.1	70	82	364	0.093	90	1.17	45
		<u>Mol-4 a4</u>	3.8	0.55	6.9	0.6	5.3	<u>10</u>	23	216	0.007	16	2.32	40
		Mol-4 a5	22.1	0.72	30.9	1.3	1.9	57	19	238	0.762	61	0.34	60
		Mol-4 a6	14.8	0.67	22.0	1.3	2.8	141	13	203	0.383	144	0.10	46
MOL5	323	Mol-5 a1	7.2	0.66	10.9	0.6	3.2	47	57	408	0.058	60	1.23	46
		Mol-5 a2	4.3	0.57	7.6	0.5	2.7	62	42	388	0.056	72	0.69	40
		Mol-5 a3	10.1	0.62	16.2	1.0	3.4	24	15	107	0.061	28	0.60	44
		Mol-5 a4	10.3	0.69	15.0	0.7	2.8	96	23	382	0.114	101	0.24	52
		Mol-5 a5	8.9	0.56	15.8	1.3	4.2	22	67	386	0.031	38	3.03	35

(Continues)

TABLE 1a | (Continued)

Sampling interval	Depth (m)	Grain	Raw age (Ma)	F_T	Corrected age (Ma)	Error (Ma)	Analytical error (%)	U (ppm)	Th (ppm)	Sm (ppm)	He (ncc)	eU (ppm)	Th/U	Sphere radius (μm)
MOL6	400	Mol-6 a1	4.6	0.66	6.9	0.5	5.5	20	90	588	0.034	41	4.45	51
		Mol-6 a2	15.0	0.77	19.5	0.8	2.3	62	14	383	0.509	65	0.22	69
		Mol-6 a3	7.3	0.76	9.5	0.3	2.1	92	45	405	0.113	102	0.49	59
		Mol-6 a4	10.0	0.61	16.3	0.9	2.2	49	3	308	0.115	50	0.05	45
MOL7	466	<u>Mol-6 a5</u>	2.3	0.47	4.8	0.4	5.6	<u>13</u>	78	134	0.008	31	6.15	34
		Mol-6 a6	3.0	0.57	5.3	0.4	4.5	55	50	153	0.010	67	0.90	36
		Mol-7 a1	6.7	0.61	11.1	0.9	6.1	9	11	303	0.016	12	1.18	44
		Mol-7 a2	5.3	0.59	7.9	0.5	4.3	23	40	263	0.037	32	1.72	49
		Mol-7 a3	4.8	0.54	8.7	0.7	4.6	34	10	186	0.017	37	0.30	34
		<u>Mol-7 a4</u>	3.4	0.53	5.8	0.5	5.6	<u>15</u>	10	97	0.009	17	0.64	41
		Mol-7 a5	5.9	0.69	8.8	0.5	3.1	25	41	322	0.049	35	1.63	49
		Mol-7 a6	7.0	0.65	10.4	0.6	2.7	83	36	378	0.131	91	0.43	49
		Mol-7 a7	9.8	0.56	12.0	0.5	2.7	52	10	303	0.210	54	0.20	79
		Mol-7 a8	8.2	0.67	12.8	0.8	2.8	56	15	233	0.125	59	0.26	48
		Mol-7 a9	8.9	0.55	13.5	0.7	2.6	75	19	352	0.121	80	0.25	49
		Mol-7 a10	4.5	0.58	7.6	0.5	2.3	187	100	248	0.134	211	0.53	39
		Mol-7 a11	3.5	0.67	6.5	0.5	5.2	40	19	177	0.015	45	0.46	36
		Mol-7 a12	3.6	0.67	6.8	0.5	3.6	70	34	278	0.022	78	0.49	41
		Mol-7 a13	4.8	0.82	7.0	0.3	2.8	66	26	353	0.080	72	0.40	58
		Mol-7 a14	4.2	0.64	6.5	0.4	3.6	20	10	240	0.021	22	0.51	48
		Mol-7 a15	7.6	0.66	13.4	0.8	2.4	51	38	267	0.098	60	0.75	44

Note: Depth is measured depth (MD) in the well, and F_T is alpha particle ejection correction, adjusted for grain geometry. Sphere radius is the radius of a theoretical sphere resembling the grain volume. Grains with analytical errors > 10%, eU < 10ppm, first He re-extracts > 4% or unrealistically high AHe ages are highlighted in bold font, analytical errors > 7.5%, eU < 15ppm, first He re-extracts > 3% in italic font and underscored.

TABLE 1b | Results of apatite (U-Th-Sm)/He dating for the BST and RL samples from the BUL-1 well.

Sampling interval	Depth (m)	Raw			Corrected age (Ma)	Error (Ma)	Analytical error (%)	U (ppm)	Th (ppm)	Sm (ppm)	He (ncc)	eU (ppm)	Th/U	Sphere radius (μm)
		Grain	age (Ma)	F_T										
BST	1318	BST3 a1	39.5	0.87	45.2	1.4	2.5	87	11	304	15.592	90	0.12	119
		BST3 a2	13.8	0.88	15.6	0.5	2.4	68	18	269	1.333	73	0.27	130
		BST3 a3	66.8	0.84	79.9	3.4	3.5	122	15	287	18.353	125	0.12	91
		BST3 a4	40.8	0.86	47.5	1.6	2.5	172	14	193	13.503	175	0.08	107
		BST-3 a6	71.8	0.88	81.7	2.5	2.5	91	15	275	18.490	95	0.16	124
		BST-6 a1	34.5	0.81	42.6	1.5	2.2	55	20	267	2.054	59	0.37	79
		BST-6 a2	5.6	0.75	7.5	0.3	2.3	41	19	231	0.128	46	0.47	63
		BST-6 a3	7.2	0.74	9.8	0.4	2.2	35	58	302	0.167	49	1.64	60
		BST-6 a4	4.7	0.69	6.7	0.3	2.4	36	23	311	0.077	42	0.64	57
		BST-6 a5	14.9	0.77	19.3	0.7	1.9	132	17	269	1.394	136	0.13	67
		BST-6 a6	27.3	0.77	35.4	1.3	1.9	85	17	235	1.747	89	0.21	71
		BST-8 a1	21.8	0.82	26.7	1.0	2.5	65	20	282	1.604	69	0.30	82
		BST-8 a2	20.6	0.79	25.9	1.0	2.5	64	18	296	0.988	68	0.28	73
		BST-8 a4	42.2	0.79	53.3	1.9	2.4	36	21	235	2.399	41	0.58	83
		BST-8 a5	29.0	0.84	34.4	1.1	2.4	40	19	252	2.550	45	0.48	97
		<u>BST-8 a6</u>	59.1	0.83	70.9	2.5	2.4	51	21	346	<u>4.894</u>	55	0.41	91
		BST-8 a7	25.6	0.76	33.8	1.4	2.4	47	52	296	0.814	59	1.10	67

(Continues)

TABLE 1b | (Continued)

Sampling interval	Depth (m)	Raw age (Ma)	Grain	F_T	Corrected age (Ma)	Error (Ma)	Analytical error (%)	U (ppm)	Th (ppm)	Sm (ppm)	He (ncc)	eU (ppm)	Th/U	Sphere radius (μm)
RL1	1335	23.7	RL1-6 a1	0.70	33.9	1.6	2.5	99	24	274	0.949	104	0.25	54
		10.0	RL1-6 a2	0.79	12.7	0.5	2.5	101	25	338	0.595	107	0.24	70
		12.0	RL1-6 a3	0.68	17.8	0.9	2.5	129	13	275	0.471	133	0.10	53
		17.9	RL1-6 a4	0.74	24.1	1.1	2.6	108	12	326	0.539	111	0.11	57
		42.4	RL1-6 a5	0.66	64.2	3.2	2.5	51	33	264	0.715	58	0.65	53
		15.4	RL1-6 a6	0.78	19.8	0.8	2.5	150	13	217	1.571	153	0.08	74
		2.0	RL1-6 a7	0.71	2.9	0.2	3.7	35	23	387	0.030	40	0.67	57
		18.5	RL1-6 a8	0.71	26.2	1.3	2.6	77	16	230	0.409	81	0.21	54
		310.4	RL1-6 a9	0.71	438.4	37.5	<u>7.6</u>	133	35	206	0.885	141	0.27	58
		20.2	RL1-6 a10	0.75	27.0	1.2	2.5	136	25	220	1.010	142	0.18	59
		42.3	RL1-6 a11	0.76	55.4	2.4	2.6	57	14	229	1.091	60	0.25	63
		13.1	RL1-6 a12	0.72	18.2	0.9	2.6	129	17	300	0.450	133	0.13	54
		23.7	RL1-6 a13	0.74	32.1	1.5	2.5	294	22	282	1.901	299	0.08	57
		6.8	RL1-6 a14	0.78	8.7	0.4	2.4	62	22	386	0.209	67	0.36	68
		27.4	RL1-9 a1	0.80	34.1	1.1	1.9	141	12	247	4.675	143	0.08	86
		6.8	RL1-9 a2	0.75	9.1	0.4	2.0	62	15	295	0.282	66	0.24	66
		32.3	RL1-9 a3	0.81	40.0	1.3	1.9	234	19	263	6.547	238	0.08	83
		14.5	RL1-9 a4	0.76	19.1	0.7	1.9	106	27	291	0.740	113	0.25	66
		5.3	RL1-9 a5	0.69	7.7	0.4	2.2	29	78	108	0.066	48	2.67	55
		7.4	RL1-9 a6	0.75	10.0	0.4	2.0	70	23	260	0.241	75	0.33	61
		22.1	RL1-9 a7	0.77	28.9	1.1	1.9	161	16	361	2.093	164	0.10	72
		9.1	RL1-9 a8	0.78	11.7	0.2	1.9	197	25	270	0.882	203	0.13	70

(Continues)

TABLE 1b | (Continued)

Sampling interval	Depth (m)	Grain	Raw age (Ma)	F_T	Corrected age (Ma)	Error (Ma)	Analytical error (%)	U (ppm)	Th (ppm)	Sm (ppm)	He (ncc)	eU (ppm)	Th/U	Sphere radius (μm)
RL2	1360	RL2-8 a1	5.5	0.74	7.5	0.3	2.9	9	17	147	0.052	13	1.89	65
		RL2-8 a2	5.9	0.80	7.3	0.3	2.5	36	26	356	0.228	42	0.74	76
		RL2-8 a3	13.3	0.83	16.0	0.5	2.4	70	28	307	1.791	76	0.41	96
		RL2-8 a4	22.0	0.76	28.9	1.3	2.5	144	28	235	1.581	150	0.20	62
		RL2-8 a5	21.4	0.72	29.8	1.4	2.4	157	52	264	1.639	169	0.33	58
		RL2-8 a6	8.8	0.75	11.8	0.5	2.5	66	25	241	0.441	72	0.38	66
		RL2-8 a7	24.9	0.76	32.6	1.3	2.4	66	26	249	1.629	72	0.39	69
		RL2-8 a8	33.7	0.82	40.9	1.5	2.5	84	20	263	4.007	89	0.23	85
		RL2-8 a9	38.5	0.81	47.4	1.8	2.4	55	33	270	2.695	63	0.60	81
		RL2-8 a11	6.6	0.74	9.0	0.4	2.5	76	28	298	0.360	82	0.37	64
		RL2-8 a12	8.6	0.78	11.0	0.5	2.5	94	28	310	0.432	100	0.30	70
		<u>RL2-8 a14</u>	1.5	0.71	2.1	0.2	5.9	<u>13</u>	25	190	0.008	19	1.90	54
		RL2-8 a16	20.8	0.82	25.2	0.8	2.2	99	44	198	3.546	109	0.45	101
		RL2-8 a17	28.5	0.79	35.9	1.4	2.3	250	46	293	4.011	261	0.18	72
		RL2-8 a18	11.0	0.78	14.3	0.5	2.2	129	47	298	1.059	140	0.37	76
		RL2-8 a19	23.6	0.82	28.9	1.0	2.2	135	54	324	3.004	148	0.40	84
		RL2-8 a21	20.1	0.84	24.0	0.8	2.2	113	38	349	3.815	122	0.33	100
		RL2-8 a22	18.9	0.76	25.0	1.1	2.2	165	44	258	1.169	175	0.27	62
		RL2-8 a23	15.0	0.71	21.1	0.9	2.3	180	36	267	1.215	188	0.20	63
		<u>RL2-8 a24</u>	1.8	0.79	2.3	0.1	3.7	<u>13</u>	34	293	0.019	21	2.57	75
		RL2-8 a25	25.3	0.80	31.5	1.2	2.2	96	37	312	1.922	105	0.38	77

Note: Depth is measured depth (MD) in the well; F_T is alpha particle ejection correction, adjusted for grain geometry. Sphere radius is the radius of a theoretical sphere resembling the grain volume. Grains with analytical errors > 10%, eU < 10ppm, first He re-extracts > 4% or unrealistically high AHe ages are highlighted in bold font; analytical errors > 7.5%, eU < 15ppm, first He re-extracts > 3% in italic font and underscored.

sedimentary fill, whereas exhumation timing and rate, and heat flow at the base of the sediment sequence are defined as parameters. PyBasin allows for the inclusion of different provenance histories and the specification of multiple provenance histories in single sediment samples. Making use of this, we define a range of possible end-member provenance histories, from which PyBasin computes the range of possible resulting AHe grain ages, including the implemented post-depositional thermal history, as well as grain and rock properties (see Section 4.3). The fit of the implemented thermal history model to the data can be tested by comparing the modelled AHe age ranges with the observed AHe ages. Such a modelling approach has been successfully tested on challenging datasets (e.g., Colleps et al. 2022).

4 | Results

4.1 | AHe Dataset

Most of the AHe ages within the Molasse section (i.e., down to depths of 500m) are younger than their stratigraphic age (Figure 3, Tables 1a and 1b). This indicates that these samples were at least partially reset after deposition. In the upper four samples (MOL1-MOL4), single-grain ages exist that are as old or older than their stratigraphic age, whereas in the lower three samples (MOL5-MOL7), all grains are younger than their stratigraphic age (apart from a single grain in MOL6). In all samples, the youngest ages are consistently younger than 10Ma, and in the lowermost two samples, the youngest single-grain ages are as low as 4 and 5 Ma. Overall, this results in less age dispersion in the lower samples (Figure 3).

AHe ages of the BST-RL interval (1306–1370 m) are all younger than their stratigraphic ages (Figure 3). However, the age dispersion is larger than seen in the overlying section, ranging between 3 Ma and 82 Ma at the top BST interval, and between 3 and 64 Ma in the RL intervals. Like in the Molasse section, age dispersion decreases towards the bottom of the section; maximum ages for the RL1 interval are 64 Ma, and for the RL2 interval, the oldest grain measured has an AHe age of 41 Ma.

This age pattern, and particularly the age dispersion and relatively old ages at a depth of >1200 m, are unexpected. Applying the concept of a partial retention zone (PRZ, see Section 3.1 and Figure 3a), the trend of younger AHe ages with depth within the Molasse section would imply a total reset of AHe ages below a depth of approximately 600 m. As introduced above, different explanations of the observed dispersed age pattern can be put forward. Presuming that the BST-RL and the MOL sample sets are characterised by comparable helium diffusion kinetics, the observed age dispersion may result from grain properties such as effective uranium (eU) or grain size (e.g., Farley 2000; Flowers 2009). However, age dispersion could also be because of different grain provenance and hence different thermal histories the sample sets experienced prior to deposition (see Section 2.1). To probe into these hypotheses, we first examine the data regarding element concentrations and grain size for correlations with the AHe ages, before modelling the data under consideration of provenance differences with PyBasin.

4.2 | Correlations of AHe Ages With Grainsize and Chemistry

Correlations between grain sphere radius (Rs, representing grain size) and age cannot be observed for most of the data (Figure 4a). However, these correlations apparently differ between different subsets of the data. Although the grain age of BST grains seemingly depends on sphere radius, this cannot be observed for other subsets. Even the apparent correlation obtained for the BST is strongly dependent on a few grains. When disregarding the two oldest grains of the BST subset, no correlation is implied. Disregarding the two largest grains, the correlation would appear stronger. We also observe that

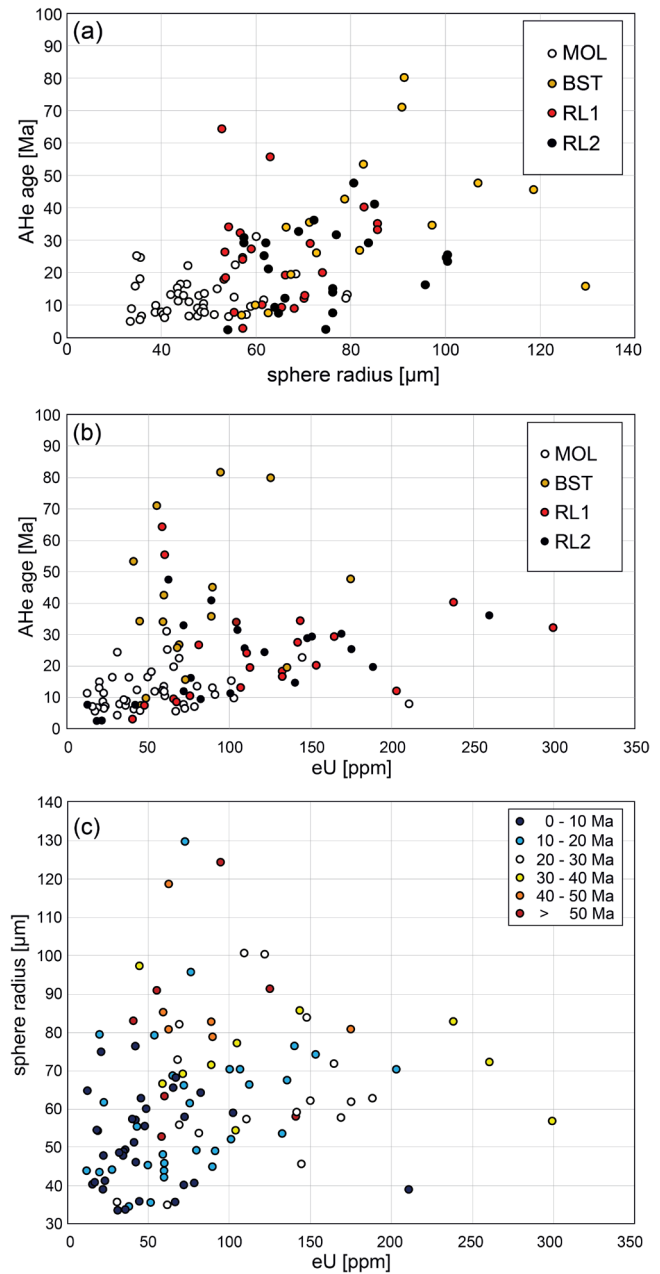


FIGURE 4 | (a) AHe age versus grainsize (sphere radius). (b) AHe age versus effective uranium concentration (eU). (c) Grainsize (sphere radius) versus effective uranium concentration (eU), with the colour code being AHe ages in 10 Ma wide bins.

Molasse grains are relatively small compared to BST-RL grains, and we also measured younger ages for the Molasse grains. However, a relatively small Molasse grain can have a relatively old AHe age, whereas a relatively large BST-RL grain can have a young AHe age. Although we do not rule out effects of grain size on the AHe ages, grain size alone cannot explain the observed age pattern, and if at all, only in a few cases.

Plotting eU versus AHe age shows only weak correlations as well (Figure 4b). Any specific eU value is associated with a wide range of AHe ages. The majority of Molasse grains show eU values ranging from 0 to 100 ppm and AHe ages ranging between 4 Ma and 20 Ma. Despite this general pattern, we observe that Molasse grains with eU values lower than 25 ppm show AHe ages below 16 Ma, whereas AHe ages for grains with eU concentrations of 25 to 80 ppm exhibit the full observed range from 4 to 30 Ma. However, this difference is marked by only a few grains. Similarly, for BST grains, a correlation between eU and AHe age cannot be observed. We note that RL grains with eU values below 50 ppm have AHe ages below 10 Ma. For eU values above 50 ppm, the correlation is much weaker, with the oldest grains exhibiting eU values around 60 ppm. Grains of higher eU show no correlation with ages ranging from around 10 to 40 Ma.

The effects of both parameters, grain size and eU, could counteract each other. As this might be an explanation for the lack of correlation observed for the AHe age versus grain size and versus eU plot, we display all three parameters together. Figure 4c shows a plot of sphere radius versus eU, with colour-coded AHe ages. If counteracting of the effects occurs systematically, the AHe age groups would plot in a diagonal band from low eU—large sphere radius to high eU—small sphere radius. Such a pattern might be interpreted for the youngest AHe age group (0–10 Ma), although datapoints of this group scatter over a broad range. Also, this group is dominated by Molasse grains, which are characterised by small grain sizes, low eU and young AHe ages, which will lead to a bias compared to groups of older AHe ages dominated by other subsets. The three grains with the highest eU of the AHe age group from 30 to 40 Ma align in a diagonal pattern. However, other grains of the group scatter over a wide range of values. Apart from these examples, the AHe age groups show no distinct patterns, scattering over a wide and independent range of eU and sphere radius.

Given these observations, the dispersed AHe age pattern of our dataset cannot be explained by effects of grain size or eU alone. Therefore, we engage in thermal history modelling to further explore the very likely influence of sediment grain provenance in the following.

4.3 | Thermal History Modelling

For thermal history modelling, we focus on the modelling code PyBasin (Luijendijk et al. 2011) that allows for including a range of provenance ages for each sample and includes the option to change heat flow over time. To this end, PyBasin has limitations in using RDAAM (radiation damage accumulation and annealing model; Flowers et al. 2009) as a diffusion model. To compensate for this limitation and to further back our PyBasin

modelling results, we also used the commonly applied modelling codes HeFTy (Ketcham 2005) and QTQt (Gallagher 2012). The modelling approaches and results for these tools are presented in detail in the [Supporting Information S1–S5](#). In summary, using HeFTy and QTQt on our dataset does not produce a model that explains the observed AHe ages and aligns with geological t–T history evidence, even when RDAAM is used as a diffusion model. Further concentrating on PyBasin, we first explore the parameter space towards a best-fit model.

4.3.1 | Exploring Parameter Space With PyBasin

First, we defined a stratigraphic column where rock properties are assigned for each lithology (Tables 2 and 3). We compiled the lithologic composition of each unit using mineralogic data. For rock density, surface porosity and compressibility, we used measurements on cuttings and drill cores provided by Nagra (2021). In addition, thermal conductivity and heat capacity have been measured on drill cores of the Weiach well located approximately 3 km northwest of the Bülach well (see Mazurek et al. 2006, for details). We note that depending on the burial scenario, the porosities may be underestimated; therefore, the thermal conductivity and, in turn, heat flow may have been overestimated. As a helium diffusion model, we used the algorithm of Farley (2000).

PyBasin uses the surface temperature at a certain time to calculate the temperatures with depth using the presumed heat flow at that time and the thermal conductivity of the rocks. Surface temperatures of times before 45 Ma were taken from Mazurek et al. (2006) and adjusted for expected paleo water depths (Figure 5). We assumed a subaquatic temperature profile for tropical regions (Ward 2006) and estimated water depth on the basis of lithofacies characteristics extracted from the lithostratigraphic log of the Bülach well (Nagra 2021). After 45 Ma, we used surface temperatures provided by Mosbrugger et al. (2005). The burial plot shows the subsidence phases determined by stratigraphic ages (Kämpfen 1972) and drilled thicknesses (Figure 5b,f, Nagra 2021). Additionally, erosion events and previous additional sedimentation, that is, sediments that are not preserved, can be defined. For the additional Cretaceous burial and subsequent erosion event, we set 400 m of sedimentation between the Upper Jurassic and 65 Ma, which we defined to be subsequently eroded by an exhumation event from 65 to 50 Ma. Constraints arguing for this estimate are the maximum of 400 m of Cretaceous sediments preserved in synclines of the Jura fold-and-thrust belt (Strasser et al. 2016), as well as the same amount of maximum Cretaceous sediments farther east in the Molasse Basin (e.g., Ortner et al. 2015, 2023). The uppermost Jurassic formation (Felsenkalk/Massenkalk) is preserved in the section drilled in the BUL-1 well, indicating that negligible amounts of Jurassic strata have been eroded (Nagra 2021).

In addition to the burial and exhumation history, the pre-depositional provenance histories of the grains were defined for each stratigraphic unit (Figure 5b, greenish-beige fields). These provenance histories are based on the exhumation history of the Alps (e.g., Hurford 1986; Schmid et al. 2004; Glotzbach, van der Beek, et al. 2011; Egli, Mancktelow, et al. 2017; Herwegh et al. 2017; Kissling and Schlunegger 2018; Herwegh et al. 2020), which represent possible source regions of the Molasse sediments

TABLE 2 | Lithostratigraphic column of the BUL-1 well, with stratigraphic ages, lithologic components and provenance histories used for PyBasin modelling.

Stratigraphic unit	Strat. ages (Ma)		Lithologic components (%)										Provenance histories (Ma)											
	Bottom	Top	Congl.	Sand	Silt	Clay	Limst.	Dolo.	Marl	Anhy.	Hal.	Coal	Start 1	End 1	Start 2	End 2	Start 3	End 3	Start 4	End 4	Start 5	End 5	Start 6	End 6
OSM	17.25	13	0.100	0.300	0.300	0.100	0.200						22	16	24	20	22	16	24	24	20			
OMM	19	17.25	0.100	0.720	0.050	0.050	0.080						23	19	24	21	23	19	24	24	21			
USM_II_top	20	19		0.350	0.200			0.450					23	20	25	22	23	20	25	25	22			
USM_II_tm	21	20		0.350	0.200			0.450					24	21	25	22	24	21	25	25	22			
USM_II_mb	22	21		0.350	0.200			0.450					25	22	26	23	25	22	26	26	23			
USM_II_base	23	22		0.350	0.200			0.450					26	22	27	23	26	22	27	27	23			
USM_I	25	23		0.600	0.250			0.150					28	23	29	26	28	23	29	29	26			
Siderolithikum	42	31			0.100	0.900							150	50	110	50	80	50	150	150	140			
Cretaceous	145	65		1.000									247	153	218	153	198	153	247	247	237			
Fels_Masse	149	145				0.025	0.950		0.025				249	157	221	157	200	157	249	249	239			
Schwarzbach_FM	151	149			0.075		0.675		0.250				250	158	223	158	201	158	250	250	240			
Villigen_FM	153	151					0.850		0.150				251	160	225	160	202	160	251	251	241			
Wildegg_FM	155	153		0.050	0.050		0.300		0.600				252	162	227	162	203	162	252	252	242			
Dogger_top	168	165			0.200	0.250			0.550				262	176	236	176	211	176	262	262	252			
Herrenwis	170	168			0.050		0.850		0.100				264	176	238	176	212	176	264	264	254			
Dogger_Clay	180	170		0.125	0.175	0.475	0.125						270	186	245	186	220	186	270	270	260			
Staffellegg_FM	200	180			0.250	0.050	0.300		0.400				286	205	257	205	235	205	286	286	276			
Klettgau_FM	225	200		0.150	0.100			0.350	0.400				306	229	282	229	255	229	306	306	296			
Baenkerjoch_FM	233	225			0.100	0.250		0.150		0.500			311	236	287	236	261	236	311	311	301			
Schinznach_FM	240	233			0.050	0.050	0.300	0.600					316	242	292	242	266	242	316	316	306			
Zeglingen_FM	242	240				0.150		0.200		0.400	0.250		317	244	294	244	267	244	317	317	307			
Kaiseraugst_FM	244	242		0.125	0.125	0.175	0.050	0.125	0.400				318	245	296	245	268	245	318	318	308			

(Continues)

TABLE 2 | (Continued)

Stratigraphic unit	Strat. ages (Ma)		Lithologic components (%)										Provenance histories (Ma)											
	Bottom	Top	Congl.	Sand	Silt	Clay	Limst.	Dolo.	Marl	Anhy.	Hal.	Coal	Start 1	End 1	Start 2	End 2	Start 3	End 3	Start 4	End 4	Start 4	End 4	Start 4	End 4
Dinkelberg_FM	249	244	0.450	0.350	0.100	0.050		0.050					320	250	300	250	270	250	320	310	320	310	320	310
Weitenau_FM	270	260	0.150	0.650	0.050	0.150							320	265	300	265	270	265	320	310	320	310	320	310

Note: Stratigraphic age ranges were taken from Kämpfen (1972), lithologic composition from Nagra (2021), provenance histories based on tectonic models of the Alps by Egli, Mancktelow, et al. (2017), Glotzbach, Bernert, et al. (2011), Herwegh et al. (2017, 2020), Hurford (1986), Kissling and Schlunegger (2018) and Schmid et al. (2004). Start 1–4 is the time at which cooling below 120°C occurred; end 1–4 is the time at which the sample reached the surface.

(e.g., Spiegel et al. 2001; Kuhlemann and Kempf 2002; Berger et al. 2005a). The paths represent endmembers and result in a range of modelled AHe ages that can subsequently be compared with the observed variation of AHe ages in samples.

Figure 5e,h shows observed AHe data (grey dots) and the computed theoretical ranges following the defined temperature history (light grey area, Figure 5e,h). The ideal fit is reached when the computed ranges cover the measured data in a way that most data are explained. The dataset comprises over 100 single-grain ages with a low number of questionable or unreliable ages (Tables 1a and 1b). Consequently, we assume that we have covered most single-grain age populations present in the samples. During fitting, we allowed for a misfit of the actual data points and the modelled ranges, if this misfit is explained by the error range of the data points.

We used previously published heat flow histories as a basis (Figure 5c; e.g., Mazurek et al. 2006; Omodeo-Salé et al. 2021) and adjusted these for the best fit to our data. The present-day heat flow was adjusted to match the measured borehole temperatures in the BUL-1 well (Figure 5d; Nagra 2021), which showed a relatively high geothermal gradient of about 35°C/km that is consistent with the relatively high temperatures and heat flow in this part of the basin (Figure 1). We reconciled the moderate basal heat flow of 60 mW m⁻², inferred from models of crustal heat flow in the Mesozoic and Cenozoic in the basin (Mazurek et al. 2006; Omodeo-Salé et al. 2021) with the high rates of present-day heat flow by assuming a linear increase in basal heat flow from 10 Ma to the present. This follows previous estimates for the onset of heating (Mazurek et al. 2006).

For the base model shown in Figure 5, we chose exhumation magnitude and timing values that fall in the range of results of previous studies (Mazurek et al. 2006; Cederbom et al. 2011; Omodeo-Salé et al. 2021; see Section 2). We took 1000 m of exhumation magnitude and a start at 7 Ma, lasting until the present day (Figure 5b). Comparing the measured AHe ages (grey dots, Figure 5h) with the predicted AHe age ranges (light grey area, Figure 5h) shows that only parts of the oldest Molasse AHe ages are covered by the predicted ranges. The Molasse AHe ages are subject to additional effects that will be discussed later (Section 5.2). In this scenario, only the younger BST-RL ages up to about 20 Ma are explained. To fit the older ages, the temperature over time needs to be reduced. This reduction can be achieved by assuming lower heat flow values or by a lower maximum burial depth. Recent heat flow is well constrained by the measured temperature profile. Therefore, a heat flow increase towards present times is required for the base model (Figure 5c). Implementing even lower heat flow for the past 10 Myr would require a sharper increase towards recent heat flow values than modelled in this study. This would be at odds with previous work (e.g., Mazurek et al. 2006) and we did not explore this possibility further; instead, we focused on exhumation history. Thus, for this study, adjusting exhumation magnitude and exhumation timing remain as options to reduce the temperature for the successions over the past million years and to improve the fit with the measured AHe data.

Figure 6 shows the scenario of 300 m less additional burial (i.e., 700 m) and subsequent exhumation. This version provides

TABLE 3 | Rock properties of the different lithologies.

Lithology	Density (kg/m ³)	Surface porosity	Compr. (m ⁻¹)	Thermal cond. (W/(m*K))	Heat capacity (J/(kg*K))
Conglomerate	2650	0.20	0.000395	4.80	2800
Sand	2650	0.20	0.000395	4.80	2800
Silt	2700	0.25	0.000395	3.50	2500
Clay	2650	0.15	0.000900	2.50	2100
Limestone	2750	0.05	0.000790	2.90	2600
Dolomite	2850	0.30	0.000790	4.80	2600
Marl	2650	0.20	0.000790	2.70	2400
Anhydrite	2900	0.10		5.50	900
Halite	2100	0.05		5.50	900
Coal	2650	0.05		1.00	900
Water	1025			0.65	4181

Note: Density, surface porosity and compressibility derived from measurements on the BUL-1 drill cores (Nagra 2021). Thermal conductivity and heat capacity from measurements on the Weiach drill cores (Mazurek et al. 2006).

a better fit to the BST-RL ages, whereas it barely changes the predicted AHe age ranges of the Molasse sequence (Figure 6e). However, this scenario also includes a hiatus from 13 to 7 Ma (Figure 6b), which is not expected. Furthermore, regarding the expected AHe age ranges, the model implies a decrease in the oldest Molasse AHe ages towards the surface (Figure 6e). This trend is less distinct or absent in the model with 1000 m of exhumation (Figure 5h). For partially reset ages, the opposite trend is expected. Therefore, we argue that the modelled expected AHe age ranges would indicate that temperatures were below the temperature needed for partial reset in the Molasse section. However, the measured data show that the oldest AHe ages of each Molasse interval are indeed younger at the bottom of the Molasse sequence; hence, partial reset must have occurred. Consequently, the predicted age trend does not fit with the observed trend. We note that both models (1000 and 700 m of additional burial and exhumation) do not provide a good fit to Molasse data. However, the trends in predicted ages show that a model with 1000 m of additional burial and exhumation explains the data better than 700 m.

As a next step, we implement an earlier onset of exhumation to reduce temperatures over the past several million years. Figure 7 shows the result of 1000 m of exhumation and an exhumation starts at 10 Ma. The model provides a good fit to most of the BST-RL data (apart from the three oldest grains) and a slightly better gradient of predicted AHe ages for the Molasse sequence as compared to the scenario with 700 m of exhumation. Also, the duration of hiatus before the onset of exhumation is reduced to 3 Myr compared to 6 Myr in earlier models.

4.3.2 | Best Fit Model

The best fit model finally found for our dataset is shown in Figure 8. As shown above, it was established by constraining

the heat flow and surface temperature history, rock properties and provenance histories, and by adjusting exhumation magnitude and timing by small increments. The old ages in the BST-RL section are predicted well by this model. For the Molasse ages, the model provides the best possible fit to the AHe ages and the age-depth trend, while keeping the fit to BST-RL ages ideal. In this model, a total of 1100 m of exhumation occurred, starting at 11 Ma (Figure 8). Interestingly, maximum temperatures for the deepest sediments were reached during exhumation at about 5 Ma (Figure 8b), as the increasing heat flow has a larger effect than cooling by erosion. Using maps and cross-sections of the Bülach region (Haldimann et al. 2017), the additional section can be explained by the deposition of 50 m of USM II, 100 m of OMM, and 950 m of OSM, respectively.

To test the robustness of our best fit thermal model, we carried out a series of sensitivity tests documented in the Supporting Information S3. In summary, our models turn out to be sensitive to minor changes in exhumation and heat flow. Nevertheless, the spread of samples considered here narrowly confines parameter space. Consequently, we estimate the uncertainty on the exhumation magnitude to be 100 m at maximum, or alternatively, an uncertainty in the exhumation onset of 1 Myr.

5 | Discussion

5.1 | Insights From Thermochronological AHe Data Distribution and Modelling

Our single-grain age dataset from the BUL-1 well (Figure 3) shows remarkable dispersion. As outlined earlier, such dispersion is quite common and can be because of several factors. Amongst these are crystal zonation, grain imperfections or authigenic crystallisation of apatites from hydrothermal fluids (Flowers et al. 2009; Ault and Flowers 2012; Cecil et al. 2014;

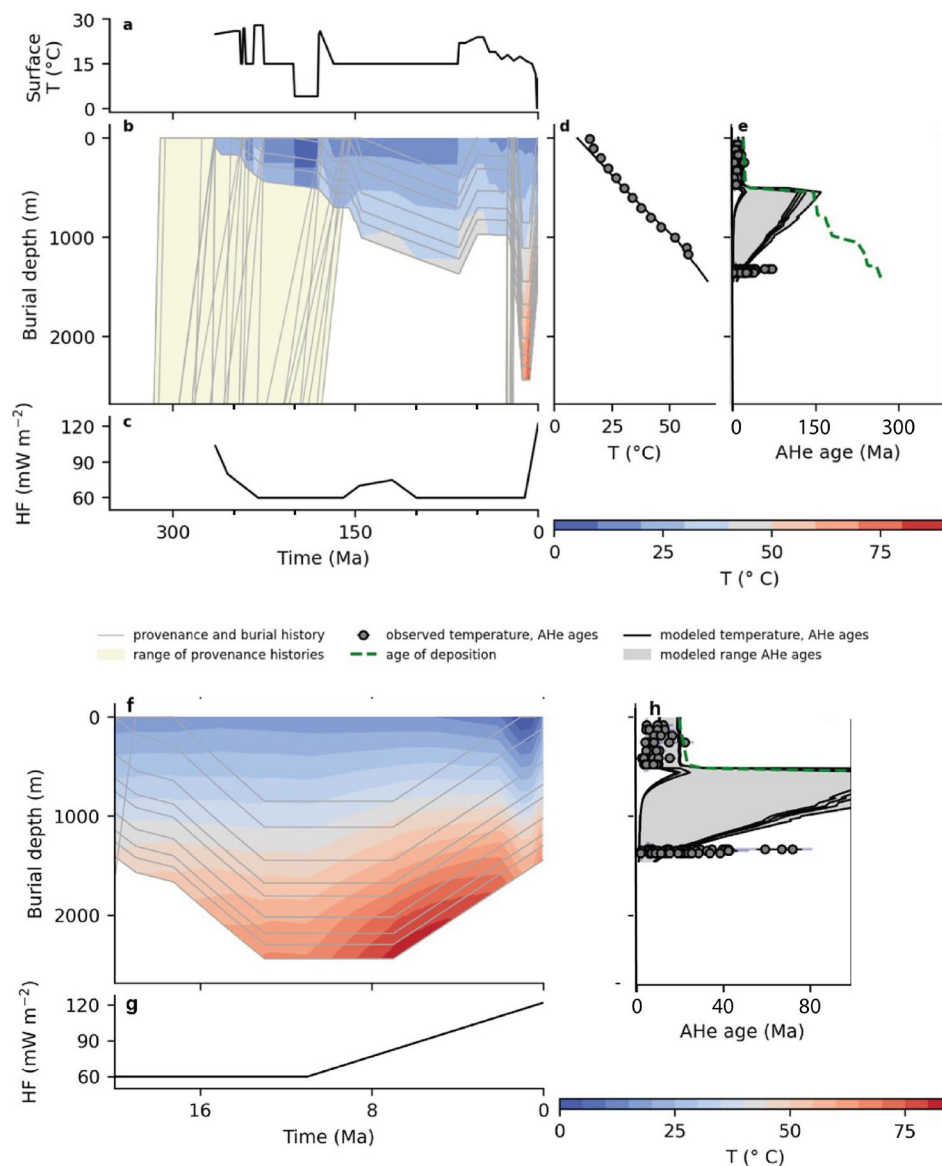


FIGURE 5 | PyBasin model parameters and results for the starting parameter setting (1000 m of burial and exhumation, late-stage exhumation starting at 7 Ma), using all samples (MOL1—RL2). (a) surface temperature over time (after Mazurek et al. 2006; Mosbrugger et al. 2005). (b) burial plot including preserved stratigraphic column (Nagra 2021) and additional sedimentation and erosion on the basis of geological constraints. Colours of the burial plot give temperatures for each depth over time. Greenish-beige areas are provenance history paths. (c) basal heat flow over time on the basis of previous studies and adjusted to provide the best fit to the data. (d) measured borehole temperatures (grey circles) of the Bülach well (Nagra 2021) and modelled temperature profile (black line). (e) AHe age data, deposition age and modelled AHe age ranges. (f) closeup of the burial plot for the last 20 Ma and basal heat flow (g). (h) closeup of the AHe age panel, age axis is cut at 90 Ma.

Djimbi et al. 2015; Fox et al. 2019; Guo et al. 2021; Berger et al. 2022). We cannot fully rule out such effects, but we argue that the overall pattern of much older ages of the BST-RL subset cannot be explained solely by zonation. This is supported by the fact that plots of AHe ages, effective uranium (eU) and grain size do not show distinct clusters that would point to groups of crystals that are similarly affected by zonation (Figure 4). An influence of direct contact with hydrothermal fluids, faulting-related frictional heat, or wildfires at outcrop sites (Brown et al. 2013; Green and Duddy 2018; Fox et al. 2019; Flowers, Ketcham, Enkelmann, et al. 2022) would cause singular very low AHe grain ages. However, for a detrital sample set recovered from a well and with no large faults in the direct

proximity of the sampling spots, such heat-event effects can be ruled out or would be expected to only affect very few grains at maximum. These effects cannot be responsible for the overall pattern observed in the dataset. The influence of implementation of helium into apatite grains from neighbouring minerals can be estimated (Murray et al. 2014) and potentially occurs for the Permian (RL) grains in our sample set, which show an iron mineral coating. The grains are large compared to a thin observed coating, limiting the influence of helium implementation. However, partial abrasion or dissolution of a past thicker coating could have occurred. The AHe ages of (RL) grains align with the Triassic AHe ages (BST), which are not coated and show even older maximum AHe ages. For these reasons, we

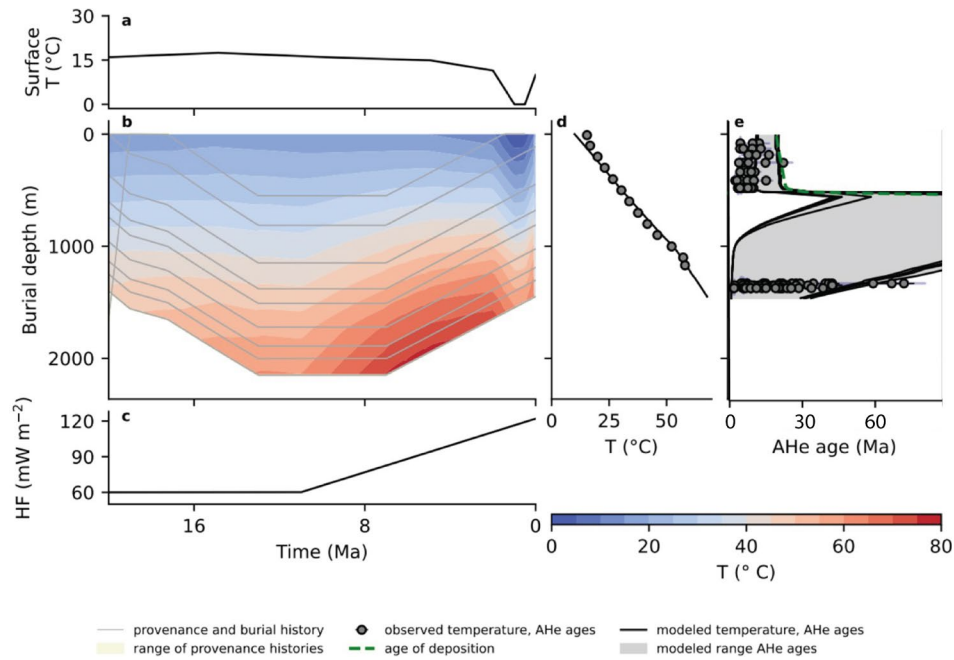


FIGURE 6 | PyBasin model parameters and results (closeup of the last 20 Myr) for 700 m of Molasse burial and subsequent erosion (i.e., 300 m less additional burial and exhumation compared to Figure 5) using all samples (MOL1—RL2). For panel explanations, see Figure 5.

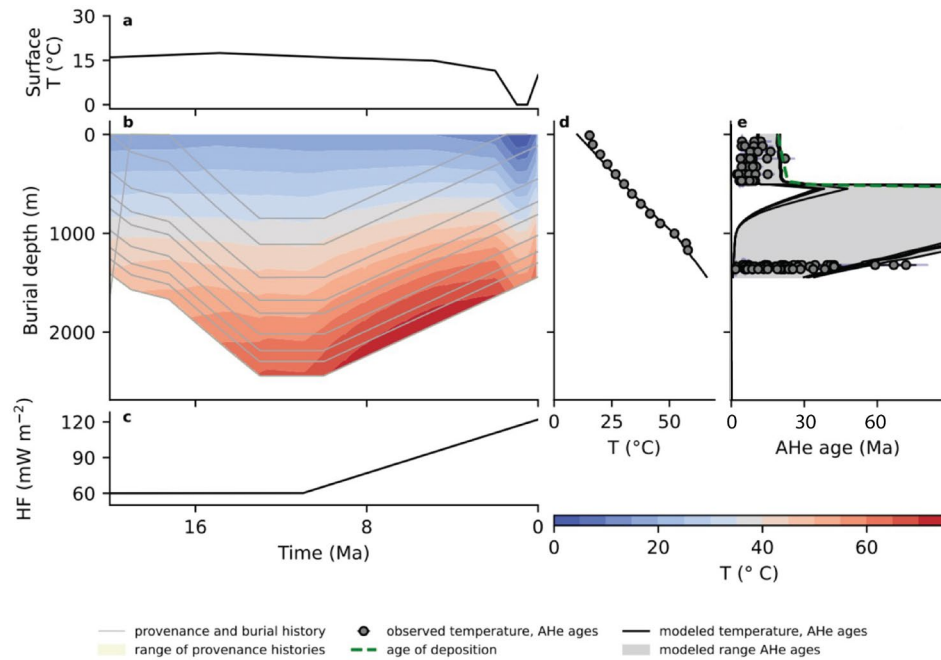


FIGURE 7 | PyBasin model parameters and results (closeup of the last 20 Ma) with 1000 m of burial and exhumation and a late-stage exhumation onset at 10 Ma (3 Ma earlier as compared to Figure 5), using all samples (MOL1—RL2). For panel explanations, see Figure 5.

also estimate the influence of helium implementation to be minor for our samples.

The overall vertical age pattern witnessed within the Molasse section of the age distribution of younger maximum AHe ages to the bottom of the section may be interpreted as an exhumed partial retention zone (PRZ; e.g., Fitzgerald et al. 2006). If an exhumed PRZ is responsible for the age-depth trend, interpreting an envelope that marks such a PRZ would imply a complete reset for the Molasse section at a depth just below

600 m (600 m refers to the section drilled in the BUL-1 well, see Figure 3). The base of the PRZ is commonly estimated to indicate a paleo-temperature of around 70°C for the AHe system (Zeitler et al. 1987; Wolf et al. 1996). Using the recent geothermal gradient of 35°C/km (Nagra 2021), this temperature is expected at a depth of around 1700 m. The difference between this depth and the estimated base of the exhumed PRZ translates into a first-order exhumation estimate of 1100 m, which aligns with our thermal history modelling results (Section 4.3.2). Below the depth of total reset inferred by

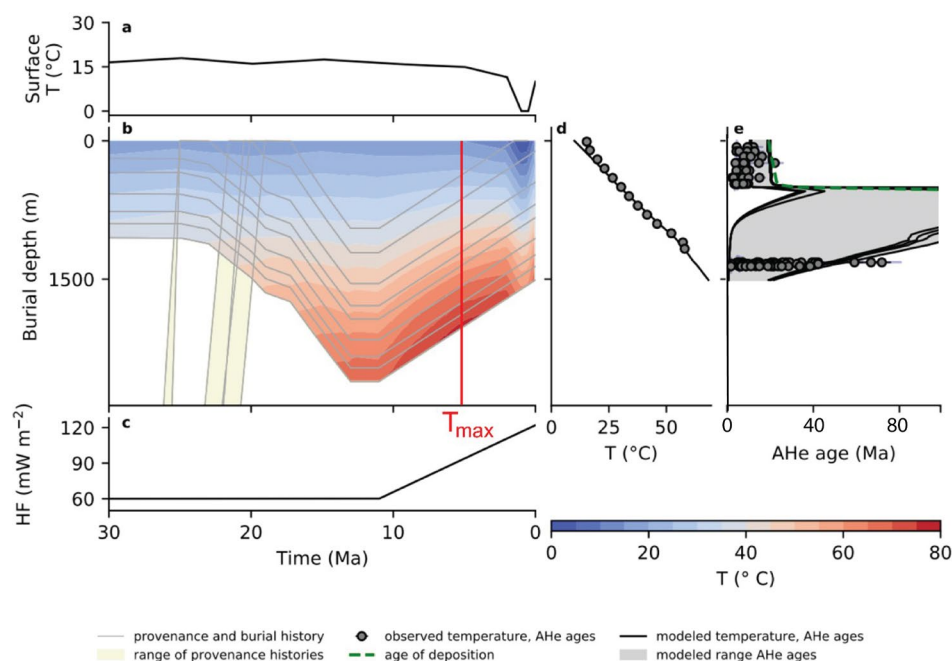


FIGURE 8 | PyBasin model parameters and results (closeup of the last 30 Ma) for the best fit model with 1100 m of additional burial and exhumation and a late-stage exhumation onset at 11 Ma using all samples (MOL1—RL2). For panel explanations, see Figure 5. $T_{\max}$ (at 5 Ma) marks the time at which maximum temperatures for the deepest sequences are calculated.

the Molasse data, BST-RL AHe ages can be as old as 80 Ma. This apparent discrepancy challenges the interpretation of a PRZ for the Molasse section. However, this can be reconciled in case the grains in the BST-RL are sensitive to a different closure temperature. The older provenance ages of the BST-RL ages indicate more helium accumulation, which consequently shifts the bottom of the PRZ to a greater depth (Luijendijk et al. 2011). In this way, the BST-RL PRZ base may be shifted to depths below 1400 m (referring to the present-day borehole section). Results from thermal modelling, including provenance ages (Figure 8) show a shift of the predicted AHe age range between the Molasse and BST-RL sample sets, which supports this interpretation.

5.2 | Implications of Modelling Results on the Thermal History of the Northern Swiss Molasse Basin

The constraints of the thermal history of the northern Swiss Molasse Basin are based on a set of measurable parameters, including the recent temperature profile (Figure 2), rock properties (Tables 2 and 3) and the thermochronometric dataset (Figure 3) compiled in this study. On the basis of these boundary conditions, we narrowed down provenance, heat flow and burial and exhumation histories to find a consistent thermal history. In the following, we discuss key parameter constraints made and discuss implications arising from the final thermal history models.

Modelling shows that provenance history is key for understanding the dispersion of ages. The older ages of the BST-RL sequence compared to the Molasse section can only be explained by the Permo-Carboniferous provenance ages

of BST-RL grains and the long time spent at relatively low temperatures prior to Molasse sedimentation (Figure 8). Although we did not directly model this with PyBasin, we note that the older provenance ages probably also affect helium kinetics during reheating and cooling in the Molasse Basin stage (e.g., Shuster et al. 2006; Flowers 2009; Flowers, Ketcham, Macdonald, et al. 2022). Most Molasse grains have young provenance ages as they originate from the exhuming Alpine chain shortly before deposition in the basin (e.g., Hurford 1986; Schmid et al. 1996; Spiegel et al. 2001; Kissling and Schlunegger 2018). However, in the study area, a possible additional source region that might yield much older provenance ages is the Black Forest (Figure 1; e.g., Timar-Geng et al. 2004; Spiegel et al. 2007; Meyer et al. 2010). Paleogeographic and paleo-drainage studies show that possible source regions for Molasse sediments of the study area are the Alpine chain from as far west as the recent location of Lake Geneva and the Black Forest region (Figure 1; e.g., Spiegel et al. 2001; Kuhlemann and Kempf 2002; Berger et al. 2005a).

We arrived at a single internally consistent thermal model, but the best-fit model leaves the youngest AHe ages of the Molasse sequence unexplained (Figure 8). Consequently, for resolving the observed pattern of two different partial retention zones, provenance age alone is not sufficient. One possible solution might be exceptionally high closure temperatures for the BST-RL grains that are not implemented in modelling algorithms. In recent years, more attention has been focused on the relevance of radiation damage in increasing the AHe closure temperature, reaching temperatures $>100^{\circ}\text{C}$ (e.g., Flowers, Ketcham, Macdonald, et al. 2022). Using RDAAM as a diffusion model, we evaluate HeFTy and QTQt models. However, these models failed to accurately represent a coherent T-t history aligned with the geological evidence (Supporting Information). Radiation damage (Flowers 2009) and

helium concentration (Shuster et al. 2006) are considered proxies for closure temperature. We propose that inconsistencies are not a problem of understanding the AHe system and diffusion models. Instead, we argue for a more complex thermal history than could be resolved with previous data sets.

For the best fit to our data, we need a sharp increase in basal heat flow from 60 mW/m² at 11 Ma to reach its present-day magnitude of 122 mW/m². Starting this heat flow increase earlier would cause temperatures at maximum burial to be too high to explain the AHe ages measured for the BST-RL sequence. Such a sharp increase has been proposed in previous studies (e.g., Mazurek et al. 2006; Omodeo-Salé et al. 2021). The initially low heat flow values of around 60 mW/m² are expected in active foreland basins, as subsidence causes cooling because of the blanketing effects of the deposited sediments (e.g., Wangen 1995). The switch to exhumation after burial will contribute to the increase of heat flow, as exhumation is associated with an opposite heating effect compared to burial. This cannot, however, fully account for the high recent heat flow values of more than 120 mW/m². Hence, the recent increase in heat flow after the burial phase needs an additional underlying geologic process. Complex heat flow settings have been recognised for other basins globally. For the North Slope of Alaska, local heat flow variations by a factor of three have been detected, which are explained by basin-scale groundwater flow (Deming et al. 1992). A comparable setting is interpreted for the Western Canada Sedimentary Basin (Weides and Majorowicz 2014). Uniform basal heat flow effects of hydrothermal systems are reported for the Foreland Basin south of the Alps in northern Italy (Pola et al. 2020). Furthermore, it has been shown that such basin-wide fluid flow systems may influence thermochronological data (Duddy et al. 1994; Ravenhurst et al. 1994; Duddy et al. 1998). However, these examples discuss spatial, not temporal variations of heat flow. Amadori et al. (2023) analysed the thermal history of this region (Tertiary-Piedmont Basin, NW Italy) and recognised heating because of mantle upwelling. Temporal heat flow changes have also been analysed for German Variscan basins; heat flow values between 50 and 70 mW/m² are proposed for burial in Carboniferous times, with values up to 110 mW/m² during Early Devonian rifting, associated with volcanism (Littke et al. 2000). For our study area, Looser et al. (2025) documented changes in fluid flow and heat flow over the past 200 Myr. These studies show that substantial and rapid changes in heat flow are a realistic and common scenario for basin settings, including the northern Swiss Molasse Basin.

For the region of the northern Swiss Molasse Basin, several potential drivers for heat flow changes through time exist. First, between 15 and 7 Ma, the Hegau volcanic system was active, as witnessed by ash layers in the Molasse sediments (Hofmann 1975; Schreiner 1992; Rahn and Selbekk 2007). Volcanic activity, possibly associated with a mantle source, may have caused local flux of hydrothermal fluids and generally elevated geothermal gradients. Furthermore, elevated heat flow may be caused by Plio-Pleistocene reactivation of basement faults related to the European Cenozoic rift system (Giamboni et al. 2004; Mazurek et al. 2006), or fluid flow and heat transport along basement faults (Griesser and Rybach 1989; Mazurek et al. 2006). These effects have also been proposed for other foreland basins worldwide (e.g.,

Deming et al. 1992; Weides and Majorowicz 2014; Pola et al. 2020).

5.3 | Comparison of the Constraints on Burial History With Previous Studies

We consider the BUL-1 well to be representative of the region of the northern Swiss Molasse Basin, as lithostratigraphy and measured recent borehole temperatures are broadly comparable with those of other nearby wells (e.g., Mazurek et al. 2006; Von Hagke et al. 2015; Nagra 2022a, 2022b). No major tectonic structures of considerable offset and/or throw directly affect the well, which rules out local exhumation contributions. The young Molasse AHe ages compared to older BST-RL ages raise the question of whether sampling for this study missed the older age population. However, the AHe data measured in our study fall into the range expected from previous data. Fission-track data from the nearby Weiach well show maximum single-grain fission-track ages of 75 Ma for Molasse samples, with a central age of about 40 Ma (Mazurek et al. 2006). Comparable, but mostly even younger ages are observed in the basin centre (Cederbom et al. 2011). The closure temperature of fission-track dating is much higher than for AHe dating. Consequently, AHe ages from the same samples would be younger. We note that examples exist where apatite fission-track ages are older than apatite (U-Th-Sm)/He ages (e.g., Green et al. 2006), but this is not the case for the existing data from the northern Swiss Molasse Basin. Consequently, we expect that our sampling campaign did not miss older AHe ages and that the measured range represents all existing age populations. The previous studies obtained different results on exhumation magnitude and timing of the northern Swiss Molasse Basin. Mazurek et al. (2006) argue for 750 to 1050 m since 9 Ma, whereas Cederbom et al. (2011) argue for 800 to 1600 m since 5 Ma. The data allow for a broad spectrum of exhumation scenarios with onset ages ranging from 4 Ma to about 12 Ma (Von Hagke et al. 2015). We present a model considering all data with a best fit of about 1100 m of exhumation since about 11 Ma. These timing constraints are not only comparable to the results of Mazurek et al. (2006), but our models also explain the difference in the interpretation offered by Cederbom et al. (2011). The authors state that they assume constant heat flow over time to arrive at their thermochronological model and conclusion. However, our modelling results suggest that increasing heat flow starting at 11 Ma is required to match current borehole temperatures and the relatively old AHe ages in the borehole. Because of this, maximum temperatures are reached around 5 Ma, despite exhumation starting at 11 Ma (Figure 8). This is also supported by clumped isotope data from vein carbonates and cements in Jurassic rocks, which indicate that no cooling occurred before 4.5 Ma (Looser et al. 2021). Hence, we agree with Cederbom et al. (2011) and Looser et al. (2021) that cooling started around 5 Ma. Exhumation, however, may have started already earlier and was contemporaneous with uplift and exhumation reported from the Subalpine Molasse (Von Hagke et al. 2012; Von Hagke, Oncken, Ortner, et al. 2014; Mock et al. 2020), and thrusting in the Jura Mountains (Laubscher 1978; Philippe et al. 1996). We note that exhumation starting at 11 Ma is not in conflict with sedimentation ages reported from the Hegau region, which

may be younger than 11 Ma (Rahn and Selbekk 2007; Binder et al. 2023). Early exhumation, as witnessed in our data set, may be associated with the beginning of folding of the Jura Mountains, resulting in small-scale uplift and topographic highs within the basin. This may have caused local erosion and redeposition of Molasse sediments, whereas in other parts of the basin, sedimentation continued without interruption. Only later, that is, after deposition of the youngest Molasse sediments, basin-wide exhumation may have started.

5.4 | Drivers of Exhumation

Previous studies proposed climatic, tectonic, or geodynamic factors as drivers of exhumation in the region of the northern Swiss Molasse Basin (see Section 2.2). We evaluate these contributors in light of our constrained exhumation history of approximately 1100 m starting at 11 Ma. Tectonic events affecting the study area are related to the folding of the Jura fold-and-thrust belt, including basin uplift (passive transport above a thin-skinned décollement). Associated localised faulting (Laubscher 1978; Jordan et al. 2015) likely played a significant role in the observed exhumation. Jura folding started in its southern parts possibly as early as 14 Ma (Looser et al. 2021), or around 12 Ma at the latest (Sommaruga et al. 1997; Burkhard and Sommaruga 1998; Becker 2000). Since then, deformation propagated northwards and may still be active; however, in a thick-skinned fashion (Giamboni et al. 2004; Madritsch et al. 2010). Structures possibly associated with early Jura folding are present in the study area and were drilled in the Bülach well (Nagra 2021; Madritsch et al. 2024). We speculate that this tectonic exhumation accounts for the starting phase of exhumation we propose from 11 Ma on (Figure 8). However, the magnitude of tectonic contribution is likely limited, as structures drilled and mapped in reflection seismic data account for a maximum of 100 m of exhumation (Malz et al. 2016; Haldimann et al. 2017; Nagra 2019, 2021).

Climatic drivers are twofold. First, exhumation associated with Quaternary glaciations, glacial scouring, incision of subglacial channels and isostatic rebound after glaciation affected the study area (Haldimann et al. 2017; Buechi et al. 2018; Pollhammer et al. 2022). However, it cannot account for exhumation recorded by our data, which has been ongoing well before the Quaternary glaciation. Second, global climate change and increased erosion since approximately 5 Ma have been proposed for mountain belts worldwide (e.g., Molnar and England 1990; Herman et al. 2013), including the Alpine foreland (Cederbom et al. 2004, 2011; Champagnac et al. 2009). However, the global increase in erosion rates associated with climate change has been questioned (e.g., Willenbring and von Blanckenburg 2010; Ganti et al. 2016; Schildgen et al. 2018). Similarly, thermochronological data of the Molasse Basin do not necessarily require a single exhumation pulse at 5 Ma (Von Hagke et al. 2012, 2015). Our data set does not support an exhumation pulse starting as late as 5 Ma because BST-RL ages would require a total reset in this case. Although we cannot completely rule out climate as a driver, its influence is not required to explain the existing data.

Lastly, geodynamic processes such as slab rollback, delamination or breakoff can lead to foreland basin uplift and heat flow variations, particularly in the Alpine foreland (e.g., Lyon-Caen

and Molnar 1989; Lippitsch 2003; Genser et al. 2007; Von Hagke et al. 2012; Schlunegger and Kissling 2015; Herwegh et al. 2017; Baran et al. 2014). Such processes are characterised by regional uplift and exhumation lasting over several million years. Typically, they are associated with volcanic activity and drainage reorganisation (e.g., Braun et al. 2014; Robl et al. 2017). In the northern Alpine Foreland Basin, both volcanic activity as well as drainage reorganisation of the Rhine River have been observed since the late Miocene (Hofmann 1975; Schreiner 1992; Rahn and Selbekk 2007; Kuhlemann and Rahn 2013; Yanites et al. 2013).

We propose a model where all drivers may have been active, albeit to varying degrees. Early exhumation from 11 Ma may have been associated with early Jura folding (Madritsch et al. 2024). The timing of exhumation does not coincide with a climatic event, but it roughly fits with the timing of Hegau volcanism (Hofmann 1975; Schreiner 1992; Rahn and Selbekk 2007). Consequently, we relate this phase to geodynamic processes. These may also be responsible for later drainage reorganisation and further exhumation. A last cooling phase may be associated with glacial processes; however, their contribution to exhumation is not resolved with our data set.

6 | Conclusions

We presented an extensive new thermochronological dataset from a deep exploration well in the distal Swiss Molasse Basin and showcased how a strongly dispersed and initially counter-intuitive single-grain age distribution can be used to better constrain the much-debated exhumation history of this basin. Using an iterative forward modelling approach and including rock properties, exhumation history, changes in basal heat flow and especially provenance ages allowed us to establish a consistent thermal history for the study area.

Along the investigated BUL-1 well, partially reset samples from the upper Molasse basin section indicate a total reset below 600 m, whereas ages from Permo-Triassic samples at more than 1300 m depth are not completely reset. We conclude that the most likely reason for the observed age dispersion is a difference in provenance histories between shallow and deep units, with the deeper samples retaining some of their inherited old provenance ages, whereas the shallow samples have much younger provenance histories. Modelling the data with the tool PyBasin allowed us to incorporate this aspect and ultimately indicates 1100 ± 100 m of late Miocene exhumation beginning as early as 11 Ma (± 1 Ma). Our models also indicate that cooling and exhumation do not necessarily need to start at the same time. In our best-fit model, cooling starts as late as 5 Ma, despite an earlier onset of exhumation, because rising heat flow since 11 Ma causes heating that exceeds the cooling by exhumation until 5 Ma. We suggest that the main drivers of the reconstructed exhumation are deep-seated dynamic topography processes related to mantle dynamics, for example, slab-breakoff. Such processes can explain an exhumation onset earlier than 5 Ma. Tectonic forcing of exhumation associated with the onset of folding in the Jura fold-and-thrust belt could have played an initial role by potentially contributing to the slow onset of exhumation from 11 Ma. Reorganisation of the Rhine River drainage system

from 4.2Ma is considered to represent a symptom of the ongoing geodynamically driven exhumation process. The role of Miocene climate change on basin-wide exhumation seems to be minor. The possible effect of Quaternary glaciations on exhumation is not resolved by our data and requires additional studies using different methods.

Our study demonstrates that the consideration of single-grain ages of many grains in dense sampling intervals using apatite helium thermochronometry is a suitable method to reconcile complex burial and exhumation histories in foreland basin settings. Although dispersed single-grain ages present a modelling challenge, determining different grain age populations provides the opportunity to derive individual time–temperature histories of these grain age populations. This can be beneficial for untangling the late common temperature history all grains share. Possible refinements of our approach are the integration of additional thermochronometers (e.g., fission-track dating), absolute dating (e.g., U–Pb) and paleo-drainage analyses to enhance the understanding of grain age populations and their provenance histories.

Acknowledgements

This study is partly funded by Nagra within the SANDS project. We thank István Dunkl and GÖochron Laboratories (University of Göttingen) for measurements and for helpful discussions. Elco Luijendijk is thanked for support with PyBasin software and discussion on the results. Meinert Rahn (ENSI), Angela Landgraf, Jens Becker (both Nagra) and Nathan Looser (then ETH Zürich) are thanked for fruitful discussions. Laura Zieger (RWTH Aachen) is thanked for discussions concerning basin modelling. We also acknowledge Kerry Gallagher for constant advice on the QTQt modelling approach. We thank two anonymous reviewers of earlier versions of this manuscript, as well as Chiara Amadori for her constructive review of the latest version. Special thanks go to Peter van der Beek for three very constructive reviews of different versions of the manuscript that enormously helped to improve it. We finally give thanks to the editor, Sarah Falkowski, for handling the manuscript and for her very helpful suggestions for improving the manuscript. Open Access funding enabled and organized by Projekt DEAL.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

Data included in manuscript tables.

Peer Review

The peer review history for this article is available at <https://www.webofscience.com/api/gateway/wos/peer-review/10.1111/bre.70056>.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. **Supporting Information S1:** bre70056-sup-0001-SupplementaryFile.docx. **Data S1:** bre70056-sup-0002-DataS1.xlsx.