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Absolute dating of the youngest sediments of the Swiss Molasse basin by apatite fission track analysis

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Key words: Molasse basin, Upper Freshwater Molasse, Hegau volcanoes, ash layer, apatite composition, fission track dating

ABSTRACT

A set of ash layer samples within the uppermost Upper Freshwater Molasse (OSM) sediments (N and E of Frauenfeld, Switzerland) was dated by apatite fission track (FT) means. The ages indicate an early Tortonian (perhaps latest Serravallian) eruption and sedimentation age of 11.5 ± 0.3 Ma. The age is in agreement with time constraints by Mammalian relicts which point to MN7-8. Due to the position of the ash layers close to the erosional gap and overlying Quaternary cover, the age represents a maximum age for the cessation of OSM sedimentation in the Swiss Molasse Basin. However, the end of Molasse sedimentation in this region had not stopped before the cover of OSM sediments by volcanic ash layers at the Höwenegg volcano (southern Germany), an event further constrained by an apatite FT age of 9.8 (-0.7/+0.8) Ma from a hornblende-bearing ash layer at Höwenegg. An isolated bentonitic ash layer occurring 25 km to the WSW of the main set of dated ashes (near Humlikon) has an age component identical to the OSM ash layers near Frauenfeld. The age suggests a source for this material within the Hegau, but is too young to be related to the volcanic activity at the Kaiserstuhl.

The apatites from the ash layer samples show two distinct compositional populations, one very close to a Cl end member and one with apatites of equal proportions of Cl and OH end member. These populations are interpreted to have possibly originated from at least two distinct igneous sources for the ashes, separated by their eruption site or eruption time or both. The distinct compositional data on the volcanic apatites may provide a basis to clarify their origin in future work.

Introduction

The Oligo-Miocene Molasse Basin has accumulated the erosive material from the Alpine orogeny (Kuhlemann & Kempf 2002). The sedimentary record in the basin mirrors the orogenic evolution of the Alps and their foreland and can thus be used to gain information on the tectonic stage of the orogeny (Schlunegger et al. 1997; Schlunegger & Willett 1999; Bernet et al. 2004). The sedimentary succession is divided into two megacycles of upward shallowing conditions with two marine sequences (Lower and Upper Marine Molasse), each followed by a sequences of mainly terrestrial deposits (Lower and Upper Freshwater Molasse). In a proximal position, i.e. close to the Alpine border, the thickness of the basin sequences amounts to more than 4 kilometres (e.g. in the borehole of Linden, Vollmayr 1983). Towards the distal margin of the basin, the deposits are reduced in thickness to less than 1.5 kilometres (e.g. borehole of Herdern, Vollmayr 1983). By magnetostratigraphic and biostratigraphic methods the time range of the two sequences was bracketed to 33-20 Ma for the lower megasequence and to 19-12 Ma for the upper (Kuhlemann & Kempf 2003). However, the top of the sedimentary record has been eroded and today forms a diachronous hiatus to the Quaternary cover. While the hiatus in the western Swiss Molasse basin includes large parts of the upper sedimentary megacycle, its extent decreases towards the east to a missing section of appr. 10 myr duration. Evidence on the amount of missing section comes from clay mineralogy and coal rank data from boreholes within the Molasse basin (e.g. Monnier 1982; Schegg & Leu 1998), however, models on the evolution of the Molasse basin (and thus on the evolution of the Alpine orogeny) are somewhat contradictory (e.g. Cederbom et al. 2004; Mazurek et al. 2006; Timar-Geng et al. 2006), in particular with respect to the end of Molasse sedimentation and the moment, when the Molasse basin became inverted and subsidence and sedimentation were replaced by exhumation and erosion. Based on fission track (FT) borehole data, Cederbom et al. (2004) derived an inversion time at around 5 Ma, which is concordant with the time that Willett et al. (2006) suggest for the cessation of orogenic activity in the Alps. In contrast, Mazurek et al. (2006), based on FT and coal rank data favour an inver-

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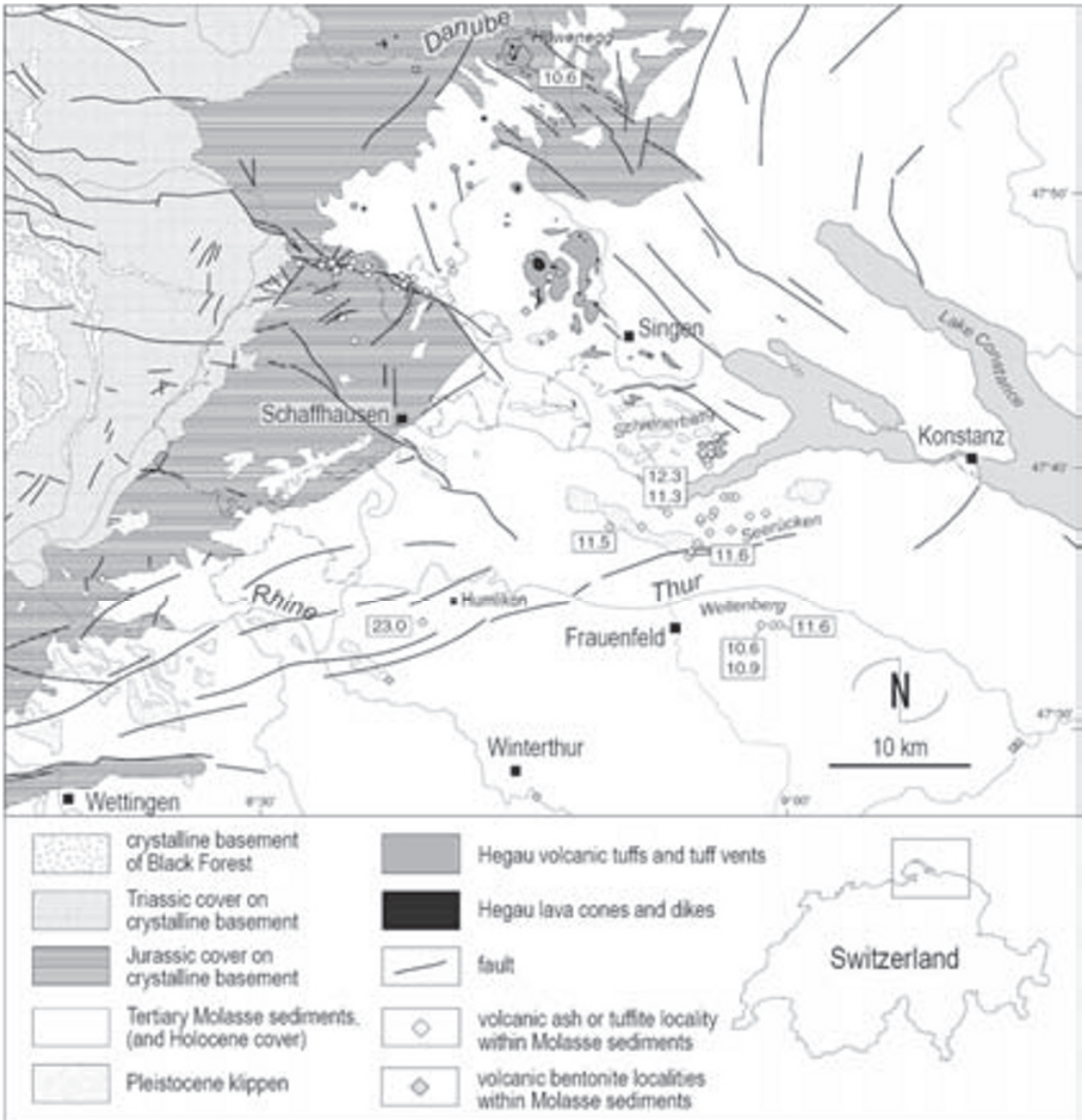

Fig. 1. Simplified geologic map of the northern Swiss Molasse basin and south of the Hegau area (adapted from Schreiner 1992 and Müller et al. 2002) with known occurrences of ash layers and tuffites (white diamonds, Hofmann 1958, 1959, 1967 1975) and bentonite (grey diamonds, Hofmann et al. 1975; Pavoni & Schindler 1981) in the OSM. Apatite fission track ages (in Ma) from volcanic ash layers are indicated in boxes next to their sampling localities.

sion point at 10 Ma, i.e. only a few myr after the sedimentation of the youngest remaining Molasse sediments. In order to timewise bracket and understand the causes for this gap, it is a prerequisite to know the present-day temporal extend of the hiatus, even though the age of the youngest Molasse sediments preserved might depend on the local erosion level.

During the time of the sedimentation of the Upper Freshwater Molasse (OSM, 17-13 Ma, Kempf & Matter 1999), Miocene volcanic activity produced a number of layers that are used as key horizons between the erosive products from the Alps. A set of four bentonite layers is described from the OSM basis (Büchi 1956, 1957, 1958; Pavoni 1956, 1958, 1960; Hofmann et al. 1975, Pavoni & Schindler 1981) and a suite of five ash layers from its top (Hofmann 1955, 1959, 1975). Only the bentonite layers have been dated to now (Gentner et al. 1963; Fischer 1988; Gubler et al. 1992). As a result, the time control on the age of the uppermost OSM sediments in the north-eastern Swiss Molasse basin is currently based on findings of mammal relicts (Bolliger 1998; Kälin 2003) and their comparison with international mammalian stratigraphic zones (Mein 1999).

The aim of this paper is to present absolute age data on the youngest widely distributed ash layer and thus on the lower

Tab. 1. Sample localities and fission track data of volcanic ash layers of the Upper Freshwater Molasse of Northern Switzerland and the Hegau area.

Sample No Locality coord. (km)/alt. (m)	Mineral and No. Crystals	Spontaneous ρ_s (N_s)	Induced ρ_i (N_i)	$P\chi^2$	Dosimeter ρ_d [a] (N_d)	Central FT Age (Ma) (-1σ/+1σ)	Mean Track Length	S.d. of distribution (No. Tracks)
MR P 314 Wilen 711.20/ 275.15/580	apatite (30)	0.006 (120)	0.126 (2565)	94%	1.434 (7221)	11.6 (-1.0/+1.2)	15.25	1.07 (100)
MR P 318 Heldhof 716.99/269.40/610	apatite (20)	0.012 (112)	0.248 (2377)	100%	1.427 (7187)	11.6 (-1.1/+1.2)	14.92	1.10 (83)
MR P 319 Wittobel-1 715.67/269.48/615	apatite (14)	0.007 (77)	0.157 (1728)	68%	1.420 (7154)	10.9 (-1.2/+1.4)	15.09	0.99 (21)
MR P 320 Wittobel-2 715.67/269.48/615	apatite (31)	0.005 (126)	0.107 (2892)	100%	1.414 (7121)	10.6 (-0.9/+1.0)	14.90	1.01 (37)
MR P 321 Freudenfels-1 709.06/277.30/560	apatite (42)	0.011 (406)	0.210 (8020)	79%	1.414 (7120)	12.3 (-0.7/+0.7)	14.90	1.03 (83)
MR P 322 Freudenfels-2 709.06/277.30/560	apatite (52)	0.013 (444)	0.281 (9500)	86%	1.407 (7087)	11.3 (-0.6/+0.6)	15.06	1.09 (100)
MR P 325 Daxmühle, Höwenegg 698.13/307.17/685	apatite (50)	0.009 (214)	0.211 (4974)	<1%	1.427 (7189)	10.6 (-0.7/+0.8)	14.20	1.13 (49)
MR P 329 Humlikon 692.05/269.75/515	apatite (38)	0.012 (274)	0.124 (2951)	<1%	1.440 (7250)	23.0 (-1.5/+1.6)	13.25	1.55 (54)
MR P 343 Nussbaumen 705.10/276.33/595	apatite (57)	0.006 (437)	0.134 (9449)	80%	1.450 (7299)	11.5 (-0.6/+0.6)	14.77	0.96 (32)

Notes:
(i) Track densities are ($x10^7$ tr cm^{-2}), [a]=($x10^5$ tr cm^{-2}) numbers of tracks counted (N) shown in brackets;
(ii) analyses by external detector method using 0.5 for the $4\pi/2\pi$ geometry correction factor;
(iii) ages calculated using dosimeter glass CN-5 for apatite with $\zeta_{CN5} = 344 \pm 5$
(iv) $P(\chi^2)$ is probability for obtaining χ^2 value for v degrees of freedom, where v = no. crystals - 1.
(v) track length data are given in 10^{-6}m, S.d = 1σ standard deviation.

boundary of the sedimentary gap between OSM and Quaternary cover. By applying the fission track (FT) method on volcanogenic apatite, a maximum age for the end of Molasse sedimentation in Switzerland is provided. Apatite electron microprobe (EMP) data reveal the compositional fingerprint of the apatites to be an important tool to decipher between different eruption events and perhaps igneous sources for the ashes.

Geologic Setting and previous age constraints

Due to oblique exhumation and erosion in the Swiss Molasse basin, the youngest Molasse sediments, the Upper Freshwater Molasse (OSM) group, are restricted to the central and eastern parts of the Swiss Molasse basin and the Tabular Jura. On the erosive gap, overlying Quaternary sediments have a minimum age of 1.8 Ma (corresponding to the mammal stratigraphic member MN17, Bolliger et al. 1996). OSM sediments in eastern Switzerland mainly comprise coarse conglomerates of the Hörnli fan, intercalated by minor marls and freshwater carbonates (Kempf & Matter 1999).

For the ash layers in the uppermost OSM, an origin from the Hegau volcanic province (Fig. 1) has been proposed (Hofmann 1975). Radiometric age data from the Hegau volcanic rocks cover a time range from 15 to 7 Ma (for a compilation of selected radiometric ages, see Schreiner 1992). Published age constraints show several inconsistencies such as for example differing ages for samples from within the same volcanostratigraphic unit and age data in contradiction to the observed volcanic succession. The oldest volcanic lithologies in the study area are isolated occurrences of bentonite that are contemporaneous to the bentonites in the lower OSM. The eruption site for these bentonites is not known. Large-scale volcanic activity in the Hegau started with the eruption of up to 100 m thick tuff layers ("Deckentuffs"). The ascent of the magmas might have been triggered by the approaching Alpine front, activating an already existing grid of vertical fault zones in the crystalline basement (Illies 1975), as is suggested by some of the slightly younger olivine melilitite volcanoes being aligned in a N-S direction. After the eruption of large volumes of pyroclastic tuffs (Deckentuffs, at 15-12 Ma) and the formation

Tab. 2. Representative electron microprobe analyses of apatites from volcanic ash layers in the Swiss Upper Freshwater Molasse and Hegau. For analytical conditions, see text. All Fe has been calculated as Fe^{2+}, OH was calculated as 1-Cl-F.

Sample no.	MRP 314	MRP 318	MRP 319	MRP 320	MRP 321	MRP 322	MRP 325	MRP 329	MRP 343
mineral	apatite	apatite	apatite	apatite	apatite	apatite	apatite	apatite	apatite
CaO	55.0	54.8	54.6	54.8	55.1	54.5	55.0	55.8	54.8
Na_2O	0.17	0.28	0.19	0.24	0.18	0.07	0.23	0.00	0.24
SrO	0.23	0.20	0.20	0.20	0.20	0.58	0.30	0.17	0.47
FeO	0.17	0.19	0.16	0.06	0.18	0.02	0.14	0.01	0.08
MnO	0.00	0.08	0.06	0.04	0.03	0.05	0.05	0.00	0.10
K_2O	0.02	0.00	0.00	0.01	0.01	0.01	0.00	0.00	0.00
P_2O_5	39.8	40.3	41.0	41.2	40.8	42.1	42.2	43.0	42.0
SiO_2	1.19	0.69	0.75	0.67	0.77	0.15	0.24	0.03	0.31
Cl	2.78	3.05	2.00	1.84	2.73	2.30	2.23	3.33	2.17
F	0.12	0.16	0.13	0.09	0.12	0.04	0.16	0.00	0.05
OH	0.04	0.05	0.03	0.03	0.04	0.03	0.04	0.05	0.03
total (wt%)	**99.5**	**99.8**	**99.1**	**99.2**	**100.1**	**99.9**	**100.6**	**102.5**	**100.2**
Number of anions on the basis of 25 (O, OH, Cl, F)									
Ca	9.913	9.859	9.798	9.830	9.847	9.717	9.723	9.715	9.732
Na	0.057	0.090	0.063	0.079	0.058	0.021	0.073	0.000	0.078
Sr	0.023	0.020	0.019	0.020	0.019	0.056	0.029	0.016	0.045
Fe	0.023	0.026	0.022	0.008	0.025	0.003	0.020	0.001	0.011
Mn	0.000	0.011	0.009	0.005	0.004	0.007	0.007	0.000	0.014
P	5.673	5.732	5.820	5.836	5.760	5.929	5.900	5.919	5.896
Si	0.201	0.115	0.126	0.112	0.129	0.025	0.040	0.006	0.052
Cl	0.793	0.869	0.569	0.521	0.771	0.647	0.625	0.918	0.609
F	0.066	0.087	0.067	0.050	0.063	0.022	0.081	0.000	0.024
OH*	0.141	0.044	0.363	0.430	0.166	0.331	0.293	0.082	0.368
total	**16.890**	**16.853**	**16.856**	**16.891**	**16.842**	**16.758**	**16.791**	**16.657**	**16.829**

of local cones with olivine melilitite vent fillings (13-9 Ma), a restricted number of phonolithic lavas rose along new vents; some of them may not have reached the surface (Schreiner 1992).

At the volcanic centre of Höwenegg (Fig. 1), a crater lake evolved and the occurrence of Hipparia in the lake sediments (Forstén 1985) documents that volcanic activity lasted into MN9 (11.1 to 9.7 Ma, Mein 1999). Because ash layers at the Höwenegg volcanic centre cover OSM sediments, this locality is classically considered as representing the end of Molasse sedimentation for this area (Trümpy 1980), while sedimentation continued further to the NE (Lemcke et al. 1953; Kuhlemann & Kempf 2002). Since their formation, many of the volcanic cones have been eroded away, including a sedimentary cover in the range of 100–200 m (Schreiner 1992). Today, the partially eroded volcanic vents make up the highest mountains in the Hegau.

Volcanic ash layers have been described from the Seerücken and Wellenberg (Fig. 1), two elongated hills to the north and east of the town of Frauenfeld (Hofmann 1959, 1975). Isolated occurrences of volcanic material of similar stratigraphic position were reported from Humlikon (Hofmann 1967), Anwil (Stumm 1964), and the Tabular Jura (Hofmann 1961). Five volcanic ash layers were distinguished in the field (A–E according to Hofmann 1959, 1–4 according to Hofmann 1975), with only a few dm to m of OSM sediments between the individual ash layers, and only 10 to 30 m beneath the uppermost preserved OSM sediments. The volcanic ash layers occur as small beds of up to 0.3 m thickness within a marly to sandy fine-grained sequence with frequent layers of thick gravel-rich sequences.

Heavy mineral analysis revealed the omnipresence of volcanogenic apatites, frequent magnetite and rare biotite (Hofmann 1959, 1975). Field observations indicate that some of the ash layers represent mixtures (“tuffites”) with non-volcanic material such as crystalline and sedimentary components that were added during the ascent of the igneous products, during sedimentation and perhaps re-sedimentation at the surface. In addition, many outcrops have sharp colour boundaries at the top, while towards the bottom they show a smooth colour transition to mostly light layers with frequent calcareous boulders (tuffaceous limestone). Among the outcrops of the Seerücken and Wellenberg bentonite layers are absent, although some of the volcanic ashes and tuffite layers contain minor amounts of smectitic material.

No radiometric age data exist for these ash layers, but the surrounding OSM sediments are attributed to the mammal zone MN7-8 (Bolliger 1998; Kälin 2003). Zircon ages from the bentonites further south (U-Pb data in Gubler et al. 1992 from the area around Zürich and Bischofzell) cover a very narrow time episode between 14.2 and 15.3 Ma (corresponding to MN5, Mein 1999), which is in agreement with nearby findings of mammalian relicts (Bolliger 1998). Between the uppermost bentonitic level (bentonite level of Bischofszell, Hofmann et

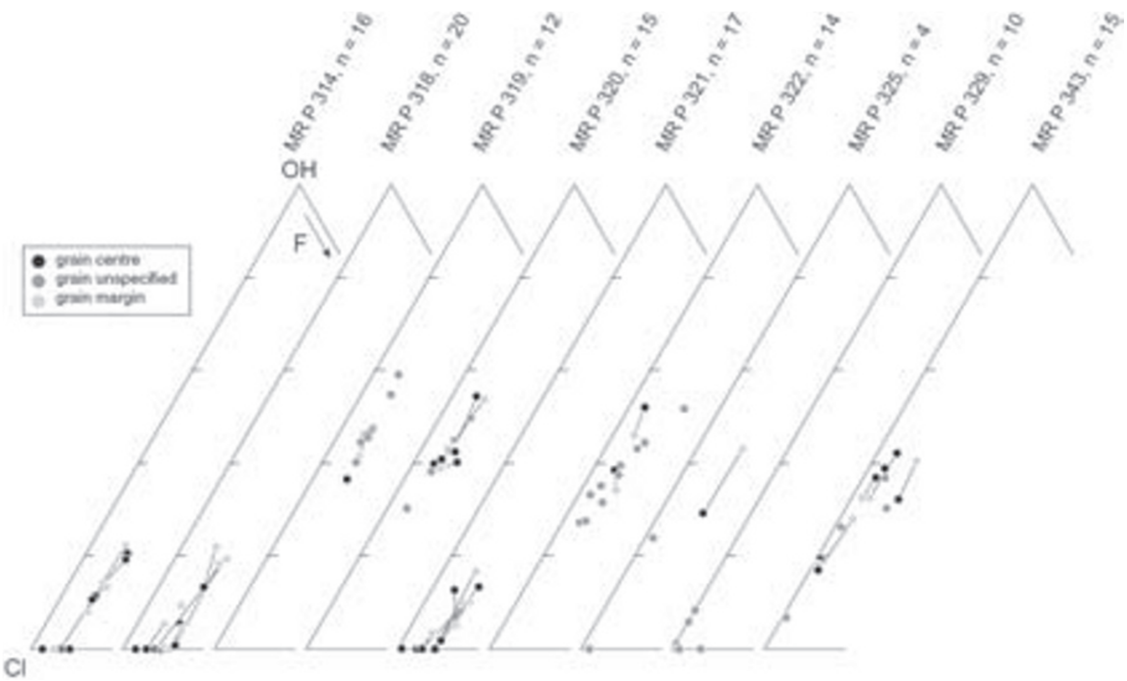

Fig. 2. Electron microprobe analyses of apatites from volcanic ash layers in the OSM of the Swiss Molasse Basin and Hegau area plotted in an F-Cl-OH end member ternary system. Analyses of a single grain's core and border are linked with lines. For representative analyses, see Table 2.

al. 1975) and the first occurrence of volcanic ashes, a thickness of appr. 300 to 400 m of OSM sediments can be estimated for the NE Molasse basin (Kälin 2003).

Sampling and methods

The presented data set focuses on samples from the ash layer B (Hofmann 1959, corresponding to layer 2 in Hofmann 1975), which is the uppermost widely distributed layer. At two localities (Wittobel, Wellenberg, and Freudenfels, Seerücken) two specimens were taken within the same outcrop but from two succeeding ash layers and half a metre apart. The data set (Table 1) is completed by two samples from localities of related origin, namely the previously dated Hornblende tuff from the Höwenegg volcano (MR P 325, radiometric age in Lippolt et al. 1963 and mapped unit in Schreiner 1992), representing the northernmost olivine melilitite volcanic centre of the Hegau (Fig. 1), and a tuffitic bentonite layer from OSM sediments near Humlikon (MR P 329, locality marked by Hofmann 1967). Apatites from ash layer B near Wilen (MR P 314) were taken from the collection of F. Hofmann (sample FH1715).

For each specimen, 2–4 kg of un- to weakly consolidated rock material were collected, the clay and silt fraction was removed by washing and the residual mineral fraction 63–315 µm was treated by standard mineral separation procedures. Zircons were absent in most heavy mineral fractions, suggesting that most specimen were not contaminated by detrital material (e.g. from crystalline rocks or from Alpine erosive material, e.g. Spiegel et al. 2001). Separated apatites were mounted and polished with epoxy on glass slides, etched at 21 °C for 20 s with 5N HNO_3 to reveal the spontaneous fission tracks. Mounts were then covered with U-free white mica sheets and sent to irradiation at the FRM-II reactor in Garching, together with CN5 dosimeter glasses. After irradiation, the white micas were etched for 45 minutes in 40% HF to reveal the induced tracks. Fission tracks were recorded and confined track lengths measured using transmitted light at 1600x magnification on a Zeiss Axioplan microscope, equipped with a computer-driven stage and a digitizing tablet at the Geoscience Department of Basel University. Age errors were minimized by counting all suitable grains on up to two mounts. Central ages (Galbraith & Laslett 1993) and 1σ errors (Table 1) were calculated using the IUGS-recommended zeta calibration approach (Hurford and Green 1983). Statistical χ^2 tests were applied to search for internal variation assuming the single grain ages of mono-population samples to have a Poissonian distribution. Failure of this test ($P(\chi^2) < 5\%$) may indicate the presence of detrital apatites.

Apatite aliquots of each sample were mounted on separate glass slides, polished and coated with carbon. Chemical analyses of apatite were performed using a CAMECA SX-100 electron microprobe, equipped with five WD spectrometers and one ED detector at the Department of Geosciences, University of Oslo, Norway. Natural and synthetic minerals were used for element calibration. The operating conditions were 10 nA and 15 kV, with a defocused beam in order to minimise sodium loss. Na and K were counted first. Na was counted for 10 s; all other elements were counted for 20 s.

Results

With the exception of the sample from the bentonitic ash layer near Humlikon (MR P 329), apatite fission track ages fall within a narrow mean age range of 10.6 to 12.3 Ma with errors in the range of 5–12% (1σ). With two exceptions (MR P 325 and 329), mean track lengths are long and cluster tightly between 14.77 and 15.25 μm. All samples except MR P 325 (Höwenegg) and 329 (Humlikon) pass the χ^2 test, and thus indicate the lack of contamination with non-volcanic apatites or any age heterogeneity caused by compositional effects.

The majority of the volcanogenic apatites are full of etchable dislocation features that may be interpreted as the result of fast crystal growth during ascent and cooling. Some grains indicate the existence of an inner core with no-to-few etched dislocation tubes that presumably crystallized prior to magma ascent. Fission track etch pits are in most cases very large and suggest a Cl-rich composition (Donelick 1993). EMP analyses (Table 2, Fig. 2) reveal the existence of two distinct compositional populations: a Cl-rich population with $X_{OH} < 0.25$ and $X_F < 0.10$ is found in three ashes of typical red colour. A population with approximately equal parts of Cl and OH end member (with X_{OH} varying from 0.3 to 0.6 and $X_F < 0.1$) is found at several other ash layers of varying colour. At Wittobel, the upper reddish ash layer (MR P 319) shows the same apatite composition as the underlying greyish tuffitic marl (MR P 320). At Freudenfels, the upper chocolate brown ash layer reveals an apatite composition different from the slightly older reddish ash layer located below and separated from the upper ash layer by 0.5 m marly silts. In both cases, the different ash layers have the same FT age with respect to their 1σ age error. Observations from the outcrops reveal that the two compositional groups cannot be correlated with colour or any other observable feature of the ash layers.

In both compositional populations internal zoning is indicated by comparison of core and rim analyses (Fig. 2). For the Cl-rich apatites a majority of measured grains indicate an evolution from Cl end member towards higher hydroxyl contents, with a few grains showing opposite trends. For the Cl-OH apatites, most measured grains become more hydroxyl-rich towards the core. Accordingly, the variation in OH-content observed in both compositional populations suggests changing chemical composition during apatite crystallization. The opposite zoning trends in the two populations underlines the assumption that the two apatite populations cannot be linked to a single eruption event.

Analysed apatites from MR P 325 cover a compositional range encompassing both the Cl-rich and the Cl-OH population, which may be attributed to a common source of the

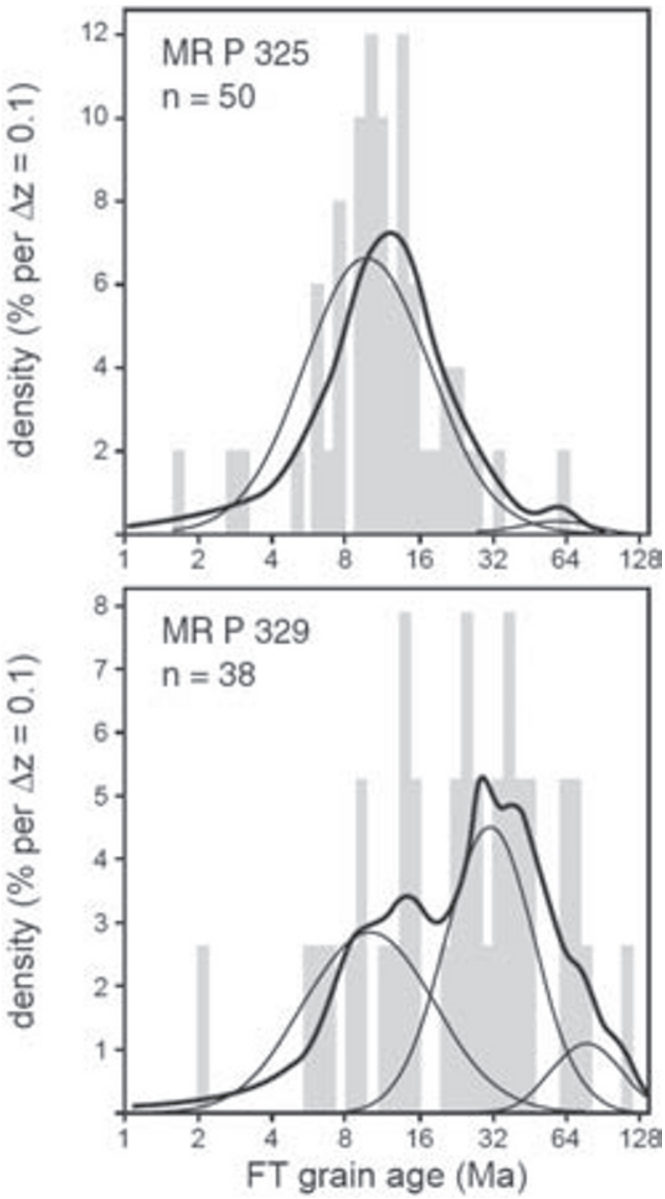

Fig. 3. Binomial fit decomposition (Brandon 1996) of apatite fission track single grain ages from two ash layers with non-volcanic detrital apatites. Gray bars represent histograms of single grain ages (1 unit = 1 grain); thick black curves correspond to probability density functions; thin black curves are single population Gaussian curves. For fitted decompositional data, see Table 3.

Höwenegg volcanic products with those of the source of the investigated OSM ash layers B. Note that the chemical variation in the MR P 329 apatites ($X_{OH} < 0.1$) is less than half of the range observed within other Cl-rich apatite samples. This very restricted compositional variation may either be attributed to a common source with those ash layers B with Cl-rich apatites or may be interpreted to represent a third apatite population not detected in the other OSM ash layers.

Tab. 3. Data from binomial fit decomposition of samples MR P 325 (Höwenegg) and MR P 329 (Humlikon). The reduced χ^2 is a measure of probability, independent of the number of populations (with a reduced $\chi^2 = 1$ corresponding to a good fit).

Sample No	population 1 age (± 1σ)	population 1 % of grains	population 2 age (± 1σ)	population 2 % of grains	population 3 age (± 1σ)	population 3 % of grains	reduced χ^2
MR P 325	9.8 (-0.7, +0.8)	98	58.0 (-13.9, +18.2)	2	–	–	0.992
MR P 329	10.1 (-1.4, +1.6)	44	31.1 (-3.0, +3.3)	47	77.7 (-14.5/+17.8)	9	1.071

Discussion

Sources of allogenic apatite contamination

According to Baadsgaard & Lerbekmo (1982), four possible sources of age contamination should be considered when dating ash and bentonite layers:

1. Dated minerals may contain old inherited cores as a result of a more complex crystallization or igneous history. This problem is of no concern, because the closure temperature of the apatite fission track method is sufficiently low to unambiguously link the closure of the apatite FT system to the moment of eruption.
2. The dated minerals may be mixed with material during ascent, including upper crustal and sediment cover material. This is a serious point of concern, because Hofmann (1975) and Keller (1984) noted the presence of crystalline basement boulders in several of the Hegau volcanic ashes, and fractured pieces of Jurassic limestone are e.g. ubiquitous in the sampled Höwenegg tuff (MR P 325). The position of the crystalline basement surface below the Hegau volcanic vents can be estimated on the basis of existing borehole data (Schreiner 1992) to be less than 1 km at Höwenegg and approximately 2.5 km below sea level next to the lower Lake Constance. This depth is not expected to have changed markedly since the time of eruption. Thus, the uppermost part of the crystalline basement is, and was, situated within or above the partial annealing zone (estimated to range from 60 to 120 °C, Wagner & van den Haute 1992). While the apatite yield from the sediments above the crystalline basement may be negligible, crystalline boulders from the uppermost few km may contain apatites with accumulated tracks at the time of Hegau volcanism. It is expected, however, that such apatites would typically be of F-rich composition (Deer et al. 1992) and show the presence of shorter tracks due to their longer residence within the partial annealing zone. In addition, strong age differences between volcanogenic and basement apatites would lead to a fail of contaminated samples in the χ^2 test.
3. The volcanic ash layers may contain detrital material from the surrounding sediments (and perhaps older ash layers) that were mixed into the ash layers during sedimentation. Such contamination may be visible by either compositional differences, rounding of the grains and/or strong variation among single grain ages leading to a fail in the χ^2 test.
4. Volcanic layers may undergo chemical weathering, which may change the age of a dated layer. This point is not relevant because the dated apatites show no alteration.

Post-sedimentary rejuvenation of the apatite FT data (as e.g. observed in Cederbom et al. 2004) can be excluded. Mean track length values prove that track did not undergo any shortening after fast cooling during ascent and eruption. The only weakly consolidated ash layers did not experience any significant burial after sedimentation. For the Hegau area and the partially eroded volcanic vents, Schreiner (1992) estimated a missing section of 100–200 metres.

From 9 samples, 7 pass the χ^2 test, while only the samples from Höwenegg and Humlikon fail and thus may indicate the presence of more than one age population. The same two samples also show distinctly lower mean track lengths that are not compatible with very fast cooling during a volcanic event. Thus their calculated mean ages (Table 1) have no geologic meaning and should not be linked to a volcanic event. In order to separate the youngest volcanogenic age population the single grain age profiles of MR P 325 and 239 were analysed by a binomial fit procedure (Brandon 1996). For the Höwenegg, contamination of the mean age is caused by one single grain age, and without this grain a corrected mean age of 9.8 (-0.7, +0.8) Ma for the young volcanic population is derived (Fig. 3, Table 3). Grain ages from the Humlikon sample can best be divided into three populations with the youngest population age being 10.1 (-1.4, +1.6) Ma. By their euhedral grain shape, frequency of etched dislocation tubes and Cl-rich composition, this population is clearly of volcanic origin. Within 1σ error, the youngest peak of the Humlikon sample is contemporaneous to many other ages of ash layer B samples, while the Höwenegg age is distinctly younger. All volcanic ages fall into the time range for the eruption of the olivine melilitites in the Hegau (Fig. 4).

Age of ash layer B

The dated ash layers B reveal a narrow time range that covers the latest Serravallian and earliest Tortonian (Gradstein et al. 2004). Its age is compatible with earlier suggestions on the age of the youngest OSM sediments in the Swiss Molasse basin (e.g. Trümpy 1980, Bürgisser 1980, Bolliger 1998, Kempf & Matter 1999; Kälin 1993, 2003). Due to the fact that only a few metres of OSM sediments are between individual ash layers and all dated layers from the Seerücken and Wellenberg were

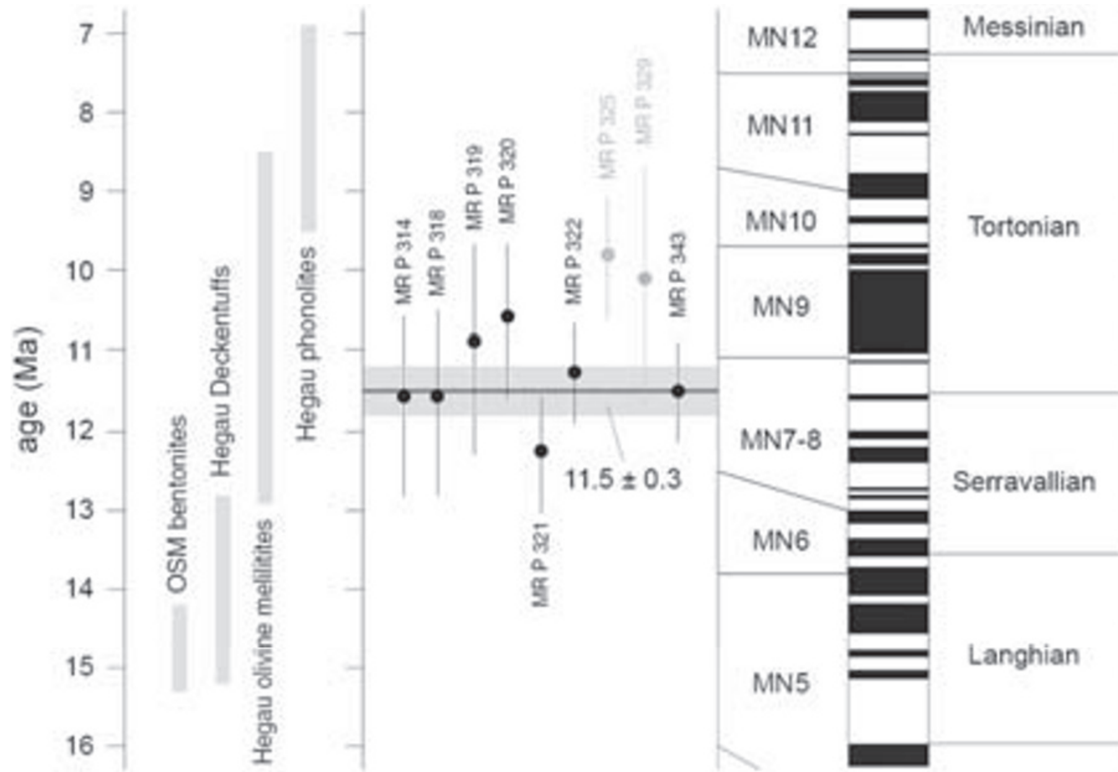

Fig. 4. Apatite fission track ages from volcanic ash layers of the Swiss Upper Freshwater Molasse and the Hegau and their relationship to Mammal stratigraphy (after Mein, 1999) and the polar magnetic reversals and seawater stratigraphy (both after Gradstein et al. 2004). Time ranges of the different magmatic phases were taken from Lippolt et al. (1963), Gubler et al. (1992) and Schreiner (1992). The two fission track ages not included into the combined age calculation are marked in grey.

mapped as the same ash layer B (Hofmann 1959), the ages of the 7 samples from Seerücken and Wellenberg (Fig. 1) are combined into one volcanic event. Their weighted mean gives an age of 11.5 ± 0.3 Ma, which corresponds to a lowermost Tortonian, perhaps latest Serravallian age (Gradstein et al. 2004). This mean age agrees with the previous relative age proposed on the basis of mammalian teeth stratigraphy (Bolliger 1998; Kälin 2003). It is distinctly older than the age found for the Höwenegg hornblende-bearing tuff, but overlaps within error with the Humlikon ash layer age. It is distinctly younger than the ages of the bentonites in the lower OSM. Therefore, from our data, it is suggested that OSM sedimentation in Switzerland has continued to the Serravallian-Tortonian boundary. At the Höwenegg, OSM sedimentation clearly lasted into the Tortonian.

Apatite compositional variation and division of ash levels

The clear distinction of two different compositional groups of apatites bears the potential to differentiate among observable ash layers and/or source areas on the basis of apatite composition. At two outcrops, two different layers were sampled in order to test for age and compositional differences in neighbouring ash layers (at Freudenfels and Wittobel): Electron microprobe analyses from apatites at Freudenfels (MR P 321 and 322, Fig. 2) have different apatite composition within the two distinct layers, that are separated by about 50 cm of marly siltstones, i.e. they clearly come from two distinct eruptions. At Wittobel, two sampled layers of different colour are equal in apatite composition and age (MR P 319 and 320, Fig. 2 and Table 1), suggesting that the upper red layer may only represent the oxidised top of the entire layer (representing a former palaeosoil?). The situation at Freudenfels, where both ash layers were mapped as belonging to B layer (Hofmann 1959) illustrates that the number of ash layers in the OSM is higher than previously mapped.

The composition of the ash layer apatites in the OSM is distinctly different from the normal F-rich apatites commonly found within most igneous and metamorphic rocks (Deer et al. 1992). No apatite compositional data have been published from the Hegau igneous rocks. However, since the Hegau volcanic activity is related to the Kaiserstuhl by their common position along the Bonndorf graben and its similar intraplate igneous character (Keller 1984), one may perhaps draw a comparison to those of the Kaiserstuhl. The Kaiserstuhl apatites from silicate magmatites have a large compositional range between F and OH end member (Sommerauer & Katz 1985; Kraml et al. 2006). Cl-rich apatites, as found in the volcanic ash layers of the OSM are absent: at the Kaiserstuhl, Cl end member contents of the apatites are below 20%. Thus, the OSM ash layer apatites clearly differ in their composition from their counterparts of the Kaiserstuhl and suggest that ash layers of unknown origin (e.g. in the Tabular Jura, Hofmann 1961) may perhaps be attributed to either a Kaiserstuhl or Hegau origin on the basis of apatite composition. Such distinction, however, has yet to be confirmed by more systematic investigations on the different magma generations of the Hegau. The yet restricted data set suggests the possibility of discriminating different ash layers (representing either eruption events or different venting systems) on the basis of apatite chemistry.

Despite the proven presence of detrital apatites of older age (Fig. 3) and supposedly shorter mean track lengths (Table 1), EMP data of MR P 325 and 329 show only apatite compositions in the range of purely volcanogenic samples (Fig. 2). The absence of apatites with different composition (in particular F-rich apatites, Deer et al. 1992) may be due to the relatively low frequency of such grains. The distinctly higher number of measurable confined tracks and better track visibility of the detrital grains with respect to their dislocation-rich volcanic counterparts, however, leads to a methodical overestimation of their fraction in single grain age and track length data.

OSM sedimentation rate and origin of the volcanic material

Stratigraphic considerations between bentonite and ash layers suggest that the OSM section between the uppermost bentonite layer (14.5 Ma) and the dated ash layers (11.5 Ma) amounts to appr. 350 m (Kälin 2003). This allows estimating an average sedimentation rate between the two volcanic events of ca. 100 m/myr. This rate is lower than the sedimentation rate of 250 m/myr derived for the time period between 15 and 13 Ma by Kempf & Matter 1999, but the difference may be explained by the very distal position of the investigated area in this study and with a late decrease in sedimentation rates after 13 Ma in the eastern Swiss Molasse basin and its transition zone to an extended river channel of micaceous sands ("Glimmersandrinne", e.g. Hofmann 1956). If, however, we assume the missing OSM section in the Hegau area (100–200m, Schreiner 1992) to have sedimented with the same rate (100 m/myr), sedimentation would have lasted until the time of eruption at the Höwenegg site at around 9.8 Ma presented in this study. Thus, the new age data support constant sedimentation rate of OSM sediments in the north-eastern Molasse Basin and Hegau area.

Within the volcanic evolution of the Hegau, the calculated age of 11.5 Ma corresponds to an intermediate time period, contemporaneous with data from the olivine melilitites, slightly younger than the "Deckentuffs", but distinctly older than the phonolites (Fig. 4). Based on field observations at the OSM ash layers and at the nearby Schienerberg (Fig. 1), Hofmann (1959, 1975) suggested an origin from this nearby volcanic vent across the lower Lake Constance, where he described the occurrence of at least three lower ash levels around a central vent. However, no ash layer was indicated to represent the equivalent of ash layer B. The presence of morphologically and chemically similar apatites at the Höwenegg documents that such apatites are common within the Hegau tuffs and ashes. An origin from the Schienerberg (with S to SW wind directions during eruption) remains the most likely explanation, but more specific arguments, based for example on age or compositional comparison, are yet missing.

The presence of apatites of similar morphology and composition near Humlikon (MR P 329) raises the question as to whether the volcanic material could have been transported over a distance of 25 km and, if yes, whether this occurrence may be linked to the ash layer B of Hofmann (1959). The bentonitic material in the layer at Humlikon (Hofmann 1967), which is not observed in the ash layers B at the Seerücken and Wellenberg hills (Hofmann 1959) may be the result of a transport-specific accumulation of vitric material that later was altered into clay minerals. For the origin of the Humlikon ash layer, an alternative scenario may be an eruption site north of the Randen area, where volcanic material has been described from within the OSM sediments (Fig. 1, Hofmann 1958). Furthermore, Hofmann (1967) suggested the bentonite to be contemporaneous with the upper bentonite in the Hegau area that was dated at 12.5 Ma (Lippolt et al. 1963). An origin from the Kaiserstuhl can be excluded, because magmatic activity in the Kaiserstuhl area ceased at around 15 Ma (Keller et al. 2002), and the youngest Kaiserstuhl tuffs contain F-OH apatites (Sommerauer & Katz 1985; Kraml et al 2006). Occurrences of volcanic material with apatite dominated heavy mineral fractions have been reported from OSM rocks in the Tabular Jura (Hofmann 1961; Stumm 1964), i.e. even further to the West and some of them closer to the Kaiserstuhl than to the Hegau. By a combination of apatite fission track dating and compositional data, it should be possible to trace the origin of these ashes back to their eruption site in the future. At present, the Humlikon ash layer is very likely to have originated from an eruption site in the Hegau.

The difference between the age of the ash layers B (Fig. 4) and the volcanic age peak of the hornblende bearing tuff from Höwenegg (Fig. 3) is significant on a 1σ error level. The Höwenegg FT age is in agreement with a hornblende age of 9.4 (± 0.5) Ma by Weiskirchner (1972, in Schreiner 1992), but significantly younger than another hornblende age of 12.4 (± 1.0) Ma (Lippolt et al. 1963) for the same tuff layer. The oldest of these three ages would require an unlikely long time period between tuff eruption and filling of the crater with lake sediments and Hipparion relicts.

Conclusions

The age of the ash layer B in the OSM sediments of 11.5 ±0.3 Ma is in agreement with existing data on the local Mammalian stratigraphy. The age is interpreted to indicate the event of an eruption at Schienerberg and the immediate sedimentation of its ejecta on OSM sediments, only a few metres below the present-day erosional gap. The age therefore represents a maximum age for the end of Molasse sedimentation in Switzerland. OSM sedimentation, however, may have continued at the same rate into the Tortonian, completely covering the vents of the Hegau volcanoes and major tuff layers that are exposed today. The observed compositional variation of the apatites infers that there are more distinguishable ash layers present than previously suggested, and they may have been erupted either from different vents or in a short time interval from the same venting site. The distinct compositional pattern of the individual ash layers forms an important prerequisite for future investigations on the source of the OSM ash layers and volcanic material within the Tabular Jura.

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