Fundamentals of detrital zircon fission-track analysis for provenance and exhumation studies with examples from the European Alps

Matthias Bernet*

Mark T. Brandon

Department of Geology & Geophysics, Yale University, New Haven, Connecticut, 06520-8109, USA

John I. Garver

Brandi R. Molitor*

Geology Department, Olin Building, Union College, Schenectady, New York, 12308–3107. USA

ABSTRACT

Fundamental aspects of detrital zircon fission-track analysis in provenance and exhumation studies include etching of fission tracks in zircon, decomposition of grain-age distributions, detection of major bedrock age components, and reproducibility of results. In this study, we present new detrital zircon fission-track data of sediment samples from eight Italian rivers draining the European Alps and previously published data from the Rhône delta in southeastern France. These samples are used to demonstrate that variable etching rates in detrital zircon, which have been shown elsewhere to necessitate a multi-etch procedure during sample preparation, are not a significant problem for zircons from the Alps. Etching response in zircon is a function of radiation damage, principally caused by α -decay. Spontaneous fissiontrack density can be used as a proxy for total radiation damage. We use spontaneous track density, fission-track cooling age, and uranium content to define a "window of countability" for detrital zircon. We also show that detrital zircon fission-track results are reproducible by comparing results from modern sediments from the same river drainage. The results also compare well with the known distribution of bedrock cooling ages in each drainage area. On a regional scale, our data illustrate that a few samples can provide an overview of the fission-track age pattern of a whole orogen, which is useful for exhumation and provenance studies.

Keywords: zircon, fission-track, provenance, exhumation, European Alps.

INTRODUCTION

Fission-track analysis of detrital zircon has become an important tool for the study of sediment provenance and long-term exhumation of orogenic mountain belts (Hurford et al., 1984; Zeitler et al., 1986; Cerveny et al., 1988; Hurford and Carter, 1991, Brandon and Vance, 1992; Garver and Brandon, 1994a, 1994b; Lonergan and Johnson, 1998; Carter, 1999; Garver et al., 1999; Spiegel et al., 2000; Bernet et al., 2001; Garver and

Kamp, 2002). In this paper, we examine the ability of fissiontrack grain-age distributions of detrital zircon from modern river sediments to resolve bedrock cooling ages in an orogenic source region, and we consider the procedures used in this kind of study. A detrital sample commonly contains a variety of zircons with different cooling ages and uranium contents. In active orogenic settings, zircons cool in the crust by tectonic and erosional exhumation or by conductive cooling following volcanism or shallow plutonism. Therefore, in geologic settings with little or no

^{*}Present addresses: Bernet – LGCA, Université Joseph Fourier, 38041 Grenoble Cedex 9. France, matthias.bernet@aya.yale.edu; Molitor---Western Washington University. Bellingham, Washington 98225, USA.

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N = 654 FT grain ages

detrital zircons from Mesozoic and

Cenozoic strata of Washington State

The points are based on 654 fission-track (FT) grain ages for detrital zircons from Mesozoic and Cenozoic strata of Washington State (Brandon and Vance, 1992; Garver and Brandon, 1994a, 1994b; and Stewart and Brandon, 2004). The data reflect the fact that it is difficult to count grains with spontaneous track densities greater than $\sim 3 \times 10^7$ tracks/cm². This effect is largely due to overlapping tracks.

igneous activity, zircon fission-track ages can be used as a proxy for long-term exhumation (Cerveny et al., 1988; Garver et al., 1999). Many orogens have deeply exhumed metamorphic internal zones where the fission-track ages for zircon provide information about postmetamorphic cooling and exhumation. The main agents of erosional exhumation are rivers that flank the orogenic system. These rivers tend to sample the landscape of their drainage areas by yield, which means that faster eroding areas potentially deliver more material to nearby basins. In addition to sediment yield, which is a function of erosion rate, the effective zircon yield also varies with lithology (Poldervaart, 1955, 1956; Deer et al., 1992).

The purpose of this study is to examine whether it is possible to detect all major cooling age components in a drainage area with one detrital sample, despite the variable etching response of detrital zircon (Naeser et al., 1987; Kasuya and Naeser, 1988). Furthermore, we are also interested to see if detrital zircon fission-track results are reproducible. The European Alps are ideal for this study because the thermochronology of the bedrock has been thoroughly investigated over the past 40 years. Using the large data set of bedrock cooling ages in the European Alps available from the literature (Hunziker et al., 1992; Bernet et al., 2001, and references therein), we are able to address the questions above. For the Alps, the temperature for effective closure of the zircon fission-track system is \sim 240 °C (Hurford, 1986).

ETCHING OF ZIRCON FISSION TRACKS



An important analytical challenge in sample preparation of zircon is the variable etching rates of tracks caused by intergrain

> Figure 2. Overview map of the Rhône River drainage with its major tributaries and the Rhône delta in southeastern France. The Rhône delta samples. labeled A. B. and C. were collected east of. west of, and close to the main channel of the modern Rhône River. Also shown are locations of samples collected from eight Italian rivers draining the southern flank of the European Alps.

1000

800

Sample	Grains	Age range	P1	P2	P3	P4	P5	P6
		(Ma)						
Orco	29	24-44	-	-	31.8 ± 3.3 100%	-	-	-
Dora Baltea	60	9-225	-	18.0 ± 2.0 61%	34.0 ± 5.6 25%	-	101.1 ± 22.5 14%	_
Sesia	60	25–112	<i></i>	-	34.2 ± 2.7 91%	62.9 ± 15.1 9%		-
Ticino	95	6-306	8.6 ± 1.4 15%	15.6 ± 1.8 57%	25.6 ± 4.8 20%	-		140.1 ± 19.0 8%
Adda	125	12-152	-	17.9 ± 1.8 39%	30.6 ± 3.6 29%	-	92.4 ± 7.6 32%	-
Adige	118	7–147	-	13.6 ± 1.9 21%	24.2 ± 3.5 36%	45.5 ± 5.7 30%	101.5 ± 15.5 13%	-
Brenta	100	20-186	-	-	29.3 ± 3.2 13%	-	78.6 ± 9.0 39%	123.1 ± 13.2 48%
Piave	100	12-219	-	-	36.2 ± 5.6 10%	2 <u>—</u> 1	94.1 ± 9.5 63%	141.6 ± 21.1 27%

TABLE 1: DETRITAL ZIRCON FISSION-TRACK DATA FROM MODERN RIVER SEDIMENTS (ITALY)

Note: Grains = total number of grains counted; results from different mounts per sample were combined; binomial peak-fit ages (P1–P6) were determined with BINOMFIT (Brandon 1996, 2002), and are given in Ma ± 2 SE. Also given is the percentage of grains in a specific peak. Zircons were irradiated with thermal neutrons at Oregon State University with a normal fluence of 2 x 10¹⁵ n/cm², along with zircon age standards (Buluk Tuff and Fish Canyon Tuff) and a reference glass dosimeter CN-5. Samples were counted at 1250x dry (100x objective, 1.25 tube factor, 10 oculars) using a zeta (CN-5) of 332.8 ± 6.8 (± 1 SE; MB).

and intragrain variation in radiation damage. This is of particular concern in detrital suites, which may be made up of grains with a variety of cooling ages and uranium concentrations (Fig 1). Revelation of fission tracks in zircon is accomplished by etching grains in a chemical etchant (most laboratories use a NaOH/ KOH eutectic melt; Gleadow et al., 1976). Etching for short times reveals tracks in grains with high radiation damage (generally older grains), and etching for a long time reveals tracks in grains with low radiation damage (generally younger grains). To ensure that none of the major age groups is excluded from detection, the "multi-mount technique" is used (Naeser et al., 1987). This method utilizes at least two mounts per sample, one for a long etch and one for a short etch. Therefore, two mounts were processed for each Italian river sample in this study (Fig. 2; Table 1). Short etch times were selected between 7 and 15 hours, while long etch times ranged between 15 and 24 hours for these samples, which were all etched at 228 °C in a laboratory oven. Each mount generally contained between 500 and 1000 zircons, depending on the amount of available sample material. The range of uranium content against cooling age for all zircons counted in this study from Italian river samples is shown in Figure 3. Sample preparation and etching of Rhône delta samples is described in Bernet et al. (2004).

The ideal situation would be to have a short etch with no over-etched grains and a long etch with no under-etched grains, so that the differently etched mounts would overlap in their fission-track grain-age distributions. In most cases, the selected etch times were able to produce short-etch and long-etch mounts that conform to this objective. A bias introduced by etching could cause problems if etchability was correlated with specific sources. Comparison of fission-track grain-age distributions from paired mounts can be used to check for this problem. For our study here, we found that each mount contains the full spectrum of grain ages. There is a bias, however, in the relative sizes of peaks in these distributions. For example, the size of a young peak might be smaller in the fission-track grain-age distribution for a short-etch mount than that for the distribution from the corresponding long-etch mount.

Alpha Damage and Etching Response

Etch time influences which grains will be countable because it affects how fission tracks are revealed. The etching response of zircon is related to radiation damage in the grain (Gleadow, 1978; Kasuya and Naeser, 1988; Garver et al., 2000a). The result of the radiation damage is that grains with high radiation damage etch easily, and tracks are quickly revealed in a few hours. Grains with low radiation damage are less chemically reactive and need longer etch times, up to 40–100 h.

The main source of radiation damage is the production of α -particles associated with the decay series for ²³⁸U, ²³⁵U, and ²³²Th (e.g., Palenik et al., 2003). The number of α -decay events per gram, D_a, is given by

$$D_{\alpha} = [\mathbf{U}] \frac{N}{m_{\nu}} \Big\{ 8c^{238} \Big(\exp[\lambda_{\alpha}^{238}\tau] - 1 \Big) + 7c^{235} \Big(\exp[\lambda_{\alpha}^{235}\tau] - 1 \Big) \Big\} + [\mathbf{Th}] \frac{N}{m_{\nu}} \Big\{ 6c^{232} \Big(\exp[\lambda_{\alpha}^{232}\tau] - 1 \Big) \Big\}$$
(1)





Figure 3. Uranium content-fission-track age relation of all zircons dated from the Italian river samples in this study.

where τ is the amount of time for α -production, [U] and [Th] are the fractional concentrations by mass of U and Th in zircon, m_{11} and $m_{\rm Th}$ are atomic masses for U and Th (238.0289 and 232.038 g/mol, respectively), N is Avogadro's number (6.022 $\times 10^{23}$ mol-1), c^{238} , c^{235} , and c^{232} are the fractional abundances of 238 U, 235 U, and ²³²Th (0.992743, 0.7200, and 1.000, respectively), and the λ_{1} variables are the rate constants for alpha decay for 238U, 235U, and ²³²Th, with $\lambda_{\alpha}^{238} = 1.55125 \times 10^{-10} \text{ yr}^{-1}$, $\lambda_{\alpha}^{235} = 9.8485 \times 10^{-10} \text{ yr}^{-1}$, and $\lambda_{\alpha}^{232} = 0.49475 \times 10^{-10} \text{ yr}^{-1}$. The integer values in equation (1) represent the total number of α -particles ejected during the decay series for each of the U and Th isotopes. This equation shows that the contribution of Th to α -production is small, mainly due to the much slower decay of 232 Th. The α -production rate for Th is only 5% relative to that for an equivalent mass of U. A compilation of zircon analyses (discussed below) indicates that the ratio Th/U in natural zircons ranges from 0 to 2.9, with a mean of 0.5 (Fig. 4; see Garver and Kamp, 2002, for details). Thus, U content is the primary factor for assessing radiation damage in zircon.

Fission decay will also produce radiation damage, but ²³⁸U is the only isotope of the U and Th series that has a significant fission decay rate, $\lambda_f^{238} = 8.45 \times 10^{-17} \text{ yr}^{-1}$. The number of fission decay events per gram of zircon is given by

$$D_{f} = [\mathbf{U}] \frac{Nc^{238} \lambda_{f}^{238}}{m_{U} \lambda_{\alpha}^{238}} \Big(\exp[\lambda_{\alpha}^{238} \tau] - 1 \Big).$$
(2)

Fission decay produces highly energetic particles and visible damage zones, but α -decay occurs much more frequently, as indicated by

$$\frac{D_{"}}{D_{f}} \approx 8 \frac{\lambda_{\alpha}^{238}}{\lambda_{f}^{238}} + 7 \frac{c^{235} \lambda_{\alpha}^{238}}{c^{238} \lambda_{f}^{238}} = 7.3857 \times 10^{7}.$$
 (3)



Figure 4. A comparison of U content in detrital zircons as measured by sensitive high-resolution ion microprobe (SHRIMP) dating of detrital zircons (compilation from Garver and Kamp, 2002), and the fission-track (FT) method (compilation of detrital zircons dated in the Washington State from Mesozoic and Cenozoic strata [see Figure 1 for references]). The SHRIMP determinations show the true range for U contents, whereas the more limited range indicated by the FT method is due to difficulties in counting zircons with high track densities.

This equation ignores the contribution of Th decay and uses an approximation $\lambda \tau \approx \exp[\lambda \tau] - 1$, which is precise to within 5% for $\tau < 100$ m.y.

The spontaneous track density ρ_s is used in fission-track dating as a measure of ²³⁸U fission events in zircon. Spontaneous track density is related to D_t according to

$$\frac{\rho_s}{D_f} = L\rho_z = 4.929 \times 10^{-3} \text{g cm}^{-2}, \tag{4}$$

with ρ_s given in tracks per cm². The other variables are *L*, the etchable length of unannealed spontaneous fission tracks (10.6 µm for spontaneous tracks in Fish Canyon Tuff zircon, Brix et al., 2002), and ρ_z , the density of zircon (4.65 g cm⁻³). Combining equations (3) and (4) gives the following relationship of α -decay to spontaneous track density

$$\frac{D_{\alpha}}{\rho_s} \approx 1.4984 \times 10^{10} \,\mathrm{cm}^2 \mathrm{g}.$$
 (5)

This relationship suggests that p might be used as a proxy for D . A critical assumption is that fission tracks and α -damage should have a similar sensitivity to thermal annealing. Tagami et al. (1996) argue that α -damage is annealed at temperatures just below those needed to start annealing of fission tracks. However, their evidence is indirect, in that it was based on changes in the etching behavior of zircon, and not on a direct measure of adamage. Garver and Kamp (2002) show evidence that color in zircon, which is a manifestation of radiation damage, requires temperatures up to 400 °C to be fully annealed on geologic time scales, whereas fission tracks are fully annealed at temperatures of 250-300 °C. These observations suggest that α -damage and fission tracks are annealed at similar temperatures, but the partial retention zone for a-damage spans a broader temperature range than that for fission tracks. Thus, we propose that ρ_{i} is in fact a good proxy for D...

Comparison of the different estimates of the relationship between ρ_{i} and D_{i} are shown in Figure 5. The dashed line shows



Figure 5. Relationship of alpha decay to spontaneous fission-track density in zircon. The dashed line shows the approximate relationship given by equation (5). The filled circles were calculated for 336 zircons using U and Th measurements determined by sensitive highresolution ion microprobe (SHRIMP) analyses from several unrelated studies (Ireland, 1992; Zhao et al., 1992; Schäfer et al., 1995; Gray and Zeitler, 1997). The calculation is based on the full equations (1, 2, 4), and a representative cooling age of 100 Ma. The solid lines show the trend for these data for cooling ages of 10 and 500 Ma.

the approximate equation (5). The points are based on U and Th measurements for SHRIMP-dated zircon grains compiled from the literature (336 zircons from Ireland, 1992; Zhao et al., 1992; Schäfer et al., 1995; Gray and Zeitler, 1997). The estimated ρ_s and D_{α} values were determined using the full equations (1, 2, 4), and a representative cooling age of 100 Ma. The solid lines show the trend for these data for different cooling ages, 10 and 500 Ma. The conclusion is that cooling age and Th/U ratio have only a minor influence on the relationship between ρ_s and D_{α} .

With increasing radiation damage, the crystalline structure of zircon is gradually transformed into an increasingly disordered structure (Palenik et al., 2003). The metamict state starts at ~3.5 × 10¹⁵ α -decay events/mg, which corresponds to $\rho_s \approx 2 \times 10^8$ cm⁻². Compilation of our fission-track zircon ages indicates that zircons have to have $\rho_s < 3 \times 10^7$ cm⁻² to be dated using standard techniques (Fig. 1). Thus, the etching bias is associated with radiation damage well below the metamict state. Nonetheless, the influence of radiation damage on etching rates is dramatic. A zircon with an old cooling age or high-U content can be etched in a few hours, whereas zircons with young cooling ages and low-U

Window of countability for Alpine detrital zircons



Figure 6. The "window of countability" of fission tracks in zircon is shown as a function of spontaneous track density, uranium content, and cooling age. The zircon fission-track grain-age data for the Rhône delta and Italian River samples were used to define the size of the window of countability. The grains represent etching conditions ranging from 6 to 60 h, but most were etched between 15 and 24 h. The horizontal contours show the cumulative probability distribution (1%–99%) for spontaneous track density in zircons from our study etched for 15 h. The vertical contours shows the cumulative probability distribution for U content in natural zircons, based on sensitive high-resolution ion microprobe (SHRIMP) analyses from several unrelated studies (Ireland, 1992; Zhao et al., 1992; Schäfer et al., 1995; Gray and Zeitler, 1997).

content are less chemically reactive and need longer etch times, up to 40-100 h.

The zircon fission-track grain-age from our Rhône delta and Italian river samples are used to illustrate the "window of countability" (Fig. 6). The graph includes determinations from all etches, both long and short. However, the contours showing the distribution of spontaneous track densities are based on average etching conditions of a 15 h etch time at 228 °C. The zircons in this diagram appear to have a sufficient range of properties to highlight the area covered by the etchability window. Other studies may not find the same range, but this would reflect a more limited range of properties for the zircons being studied.

Grain-Age Distributions and Spontaneous Track Density

As noted above, the etching bias will be a significant problem in cases where etchability correlates strongly with zircon fission-track age. For example, Naeser et al. (1987) and Cerveny et al. (1988) showed a strong bias for very young zircon fission-track ages from sediments shed from the Himalayas. In areas with cooling ages older than ca. 5 Ma, this aspect of the etching bias seems to be less of a problem (Garver et al., 2000a). To illustrate, we considered an example from one of our Rhône delta samples (Fig. 2, sample B, Stes. Maries-de-la-Mer, Table 2). For this sample, five mounts were prepared and etched for 6, 10, 15, 30, and 60 hours each. About 20 grains were dated per mount. The results are illustrated in Figure 7 by comparing the two mounts that have the largest difference in etch time (6 and 60 h) and the two mounts with the smallest difference in etch time (10 to 15 h). Figure 7A and 7B show p for the grains in terms of cumulative probability, ranging from 0 to 100%. The Kolmogorov-Smirnov (KS) test (Press et al., 1992, p. 614-617) is used to assess the statistical significance of the difference between the distributions. A low probability on the test, such as P(KS) < 5%, would indicate that the differences between the two distributions are significant. If P(KS) >> 5%, then the differences could be due to random chance alone. The test indicates a significant difference between the 60 h and 6 h etches, with P(KS) < 1%, but no significant difference between the 10 and 15 h etches, where P(KS) = 96%.

The next step is to examine probability density plots for the fission-track grain-age distributions for each of the four mounts (Fig. 7C–D). The density plots were constructed from grain age data using the method of Brandon (1996). The density plots indicate that all four etches sampled the same range of grain ages, even though there are only 20 grains dated per mount. To apply the KS test, we recast the grain-age data as cumulative probability plots (Fig. 7E–F). The KS probabilities of 99% and 86% for these comparisons indicate that there is no significant difference between the fission-track grain-age distributions, despite the differences in etching times. Furthermore, comparing the range of detrital zircon fission-track ages with known bedrock zircon fission-track ages in the source area of the Rhône River system (Bernet et al., 2004), we find that all major bedrock-age components are covered.

The conclusion is that for the Alps, we can easily reveal countable fission tracks in all major grain-age components of the total grain-age distribution. There are three reasons for this outcome. (1) The Alps source regions are not dominated by very young grain ages (<5 Ma), so that the etching process is able to reveal tracks in almost all zircons. (2) Each zircon source appears to supply zircons with a wide range of uranium concentrations, which can be properly etched with different etch times. (3) Zircons from the Alps show no strong spatial correlation in the bedrock between the U and Th content and zircons cooling ages. This result was expected given that many of the metamorphic rocks in the source region are derived from sedimentary protoliths, so that zircon properties are already randomized in the source region.

The conclusion from this exercise is that for Alpine zircons etching does not have a large influence on the sampled grain-age distribution. A single mount, etched anywhere between 6 and 60 h, would have covered the expected range of grain ages. Nevertheless, it is strongly recommended that at least two mounts with different etch times are used for detrital samples. This is

Sample	Grains	Age range (Ma)	P1	P2	P3	P4
A—Fos-sur-Mer	71	6–434	14.2 ± 1.9 18%	29.3 ± 4.0 17%	66.1 ± 8.5 26%	162.6 ± 20.1 39%
B-Stes. Maries-de-la-Mer	97	5–353	11.4 ± 2.0 29%	21.4 ± 3.5 30%	65.4 ± 8.1 21%	162.1 ± 21.7 20%
C—Plage de Piémanson	80	5–301	10.1 ± 1.3 28%	23.1 ± 3.0 25%	54.0 ± 7.5 24 %	112.3 ± 14.9 23%
Rhône delta (combined)	248	5-434	11.7 ± 1.3 26%	24.1 ± 2.7 24%	64.5 ± 6.8 26%	153.9 ± 16.5 24%

TABLE 2: DETRITAL ZIRCON FISSION-TRACK DATA FROM RHÔNE DELTA SAMPLE (SOUTHEAST FRANCE)

Note: Grains = total number of grains counted; results from different mounts per sample were combined; best-fit peak ages (P1–P4) were determined with BINOMFIT (Brandon 1996, 2002) and are given in Ma ± 2 SE. Also given is percentage of grains in a specific peak. Zircons were irradiated with thermal neutrons at Oregon State University with a normal fluence of 2 x 10¹⁵ n/cm², along with zircon age standards (Buluk Tuff and Fish Canyon Tuff) and a reference glass dosimeter CN-5, and counted at 1250x dry (100x objective, 1.25 tube factor, 10 oculars) using a zeta (CN-5) of 359.9 ± 12.7 (± 1 SE; BRM).



Figure 7. Spontaneous track density and fission-track grain-age distribution comparison for different etch times. Examination of the influence that the etching bias might have on a typical detrital zircon fission-track grain-age distribution. The plots are based on fission-track grain-age distributions from sample B collected from modern sediments of the Rhône delta. The four distributions were treated with different etch times, 6, 10, 15, and 60 h. Cumulative probability plots of spontaneous track density distributions show (A) significant differences between the grains dated from the shortest and longest etches (6 and 60 h), but (B) little difference between the two closest etches (10 and 15 h). The fission-track grain-age distributions for these four mounts are presented as probability plots in (C) and (D), and cumulative probability plots in (E) and (F). The main conclusion is that etching bias observed in (A) appears to have little if any bias on the fission-track grain-age distributions of these samples.



Figure 8. Bedrock zircon fission-track contour maps of (A) Sesia River drainage, Western Alps: (B) Dora Baltea River drainage; Western Alps: and (C) Ticino River drainage, Central and Southern Alps. Contours were constructed from published zircon fission-track data (Hunziker et al., 1992; Bernet et al., 2001, and references therein).

especially true for detrital zircon suites that are dominated by very young (<5 Ma) or very old (>300 Ma) cooling ages, to fully understand the grain-age/U content range and to select appropriate etch times (Garver et al., 2000a, 2000b).

COUNTING CRITERIA

In most bedrock zircon fission-track studies, 10–20 clear, well-etched grains are typically dated to determine a cooling age of a single-source sample (i.e., a granitic intrusion). More zircon grains need to be dated for detrital samples because they usually contain a mixture of zircons with a variety of different cooling ages. Only a random selection of countable zircons

should be analyzed from a randomly mixed suite of zircons in the Teflon mount. Grains should only be selected by their countability and not by their shape, color, or other attributes. Grains with well-exposed and polished surfaces parallel to the zircon C-axis, containing well-etched fission tracks, with some tracks parallel to the C-axis, were considered countable in the present study. Under-etched or over-etched grains were not counted. In addition, metamict grains and grains with very high spontaneous track densities (>~3 × 10⁷ tracks/cm²) or strong zonation, as well as grains with uneven surfaces, cracks or very small counting areas of less than 270 μ m², were not counted. If possible, enough grains per sample should be dated to ensure that the major components—those that make up more than 20% of the distribution—are represented (see discussion on detection limits in Stewart and Brandon, 2004). In practice, this means that, if a sufficient number of grains were available, 60–100 countable grains per sample were dated in this study; otherwise, all possible grains were counted (Tables 1 and 2).

GRAIN-AGE PEAKS

All observed fission-track grain-age distributions were decomposed into their main grain-age components or peaks. We followed the approach of Galbraith and Green (1990) in using their binomial peak-fit method (Brandon, 1996, 2002). Zircon fission-track peak ages are a proxy for long-term exhumation rates, where cooling occurs by erosion or normal faulting, and not following magmatic events (Cerveny et al., 1988; Garver et al., 1999). The Alps are basically free of recent volcanism. The last major magmatic activity occurred ca. 30 Ma (von Blanckenburg, 1992; Dunkl et al., 2000).

Peak ages tend to remain fairly constant within their error range, whereas peak sizes are much more variable from sample to sample and through time. Peak age reflects the amount of time needed to exhume the zircons from the depth of the zircon fission-track closure temperature. Thus, peak age provides an estimate of long-term exhumation rates. Peak size, however, is influenced not only by the long-term erosion rate, but also by short-term variations in erosion rates (e.g., storms, rock slides, etc.) and also by spatial variations in zircon concentrations in the source region. Peak size can also be influenced by the etching bias discussed above. In general, we find that peak ages tend to be a more robust feature of a fission-track grain-age distribution, whereas peak sizes can vary considerably within replicated distributions.

DETECTION OF BEDROCK COOLING AGES

One detrital sample can provide a remarkably representative picture of the bedrock fission-track cooling-age distribution in a drainage area, as first shown by Zeitler et al. (1986) and Cerveny et al. (1988). This initial picture can be further refined by comparison of the detrital zircon fission-track grain-age distributions with the distribution of bedrock zircon fission-track ages in river drainage areas in the European Alps. Because there have been over four decades of bedrock fission-track analysis in the European Alps, a large data set of low-temperature cooling ages is now available (e.g., Hunziker et al., 1992). We selected the Sesia River, Dora Baltea River, and Ticino River in the Western, Central, and Southern Alps (Fig. 2, Table 1) to make the proposed comparison because these drainages have different sizes, they drain areas with diverse exhumation rates, and the bedrock cooling-age data set has the highest density in these areas. We constructed contour maps for zircon fission-track ages (Fig. 8) from published fission-track data (Hurford and Hunziker, 1985; Flisch, 1986; Hurford, 1986; Giger and Hurford, 1989; Michalski and Soom, 1990; Hurford et al., 1991; Hunziker et al., 1992;



Figure 9. Probability density plots of fission-track grain-age distributions of eight Italian modern river samples in west to east order (Fig. 2), reflecting the regional trend of faster exhumation in the Western and Central Alps and slower exhumation in the Eastern and Southern Alps.

Seward and Mancktelow, 1994; Bertotti et al., 1999; Fügenschuh et al., 1999; Bernet et al., 2001). Here, we review the results for all three river samples.

A. The Sesia River, the smallest of these three drainages, drains part of the Monte Rosa massif, the northern part of the Sesia-Lanzo Zone, and the Ivrea Zone (Fig. 8A). Bedrock zircon fission-track ages in this drainage range from 25 to 130 Ma with the majority of ages ca. 35 Ma for the Sesia-Lanzo Zone, but the area also has a number of zircon fission-track ages between 45 and 60 Ma (Hurford and Hunziker, 1985; Hurford and Hunziker, 1989; Hurford et al., 1991; Hunziker et al., 1992). This bedrock age pattern is reflected in the detrital sample, which has two major peaks at 34.2 ± 2.7 Ma and 62.9 ± 15.1 Ma.

B. The Dora Baltea River, which reaches the southern flank of the Mont Blanc massif, drains parts of the Dent Blanche nappe, the Aosta Valley, and Sesia-Lanzo Zone. Published bedrock zircon fission-track ages range between 12 and 190 Ma. The fission-track contour map (Fig. 8B) shows that there are three major cooling-age components in this drainage, <20, 20–40, and >100 Ma (Hurford et al., 1991; Hunziker et al., 1992). These age components are detected in the detrital sample with peak ages of $18.0 \pm 2.0, 34.0 \pm 5.6$, and 101.1 ± 22.5 Ma.

C. The Ticino River drains the Central Alps (Lepontine dome) and parts of the Southern Alps, including the northern end of the Ivrea Zone. This drainage allows the best comparison between bedrock ages and detrital age components, because it has the highest density of bedrock zircon fission-track ages of the examples presented in this paper (Hurford, 1986; Giger and Hurford, 1989; Michalski and Soom, 1990; Hunziker et al., 1992). The bedrock zircon fission-track ages show the largest range, between 8 and >200 Ma, and the ages can be divided into groups of <10, 10–20, 20–50, and >100 Ma on the fission-track contour map (Fig. 8C). In the detrital sample, components were detected as peaks at 8.6 ± 1.4 , 15.6 ± 1.8 , 25.6 ± 4.8 , and 140.1 ± 19.0 Ma, which is representative of the bedrock pattern despite its apparent complexity.



Probability density

Figure 10. A: Cumulative probability plot of fission-track grain-age distributions of the Rhône delta. southeast France, samples Fossur-Mer (A), Stes. Maries-de-la-Mer (B), Plage de Piémanson (C). B: Probability density plots of best-fit peaks of zircon fission-track grain-age distributions in the three Rhône delta samples (Fig. 2, Table 2). Comparison of peaks indicates similarity of detrital fission-track results for samples from the same depositional setting.

The above-mentioned results demonstrate the reliability of the detrital fission-track method to represent the zircon fissiontrack age distribution in a drainage area. To expand this approach from single, local drainages to a regional scale, we examined results from five more river drainages: the Orco, Adda, Adige, Brenta, and Piave rivers, all of which drain the southern flank of the European Alps. Probability density plots showing the observed fission-track grain-age distributions of all river samples are presented in Figure 9. Fission-track peak ages are given in Table 2. All of the fission-track grain-age distributions are consistent with the cooling history and exhumational evolution of the Alps. Essentially, the detrital fission-track data reflect: (*a*) fast exhumation of the Central Alps (~0.4–0.7 km/m.y.), where the metamorphic internal zone of the Alps is exposed; and (*b*) slower exhumation of the southern flank of the Alps and farther to the east (0.2 km/m.y. and less). In these areas, the proportion of young to old grains decreases.

CONSISTENCY OF DETRITAL ZIRCON FISSION-TRACK RESULTS

We noted above that a single detrital sample could be used to resolve the major fission-track age components in a drainage area, but an important assumption in many studies of detrital minerals is that a single sample from a small part of a depositional system is representative of all the sediment in that system. Few studies have addressed this issue. Here, we present three samples from the Rhône River delta in southeastern France (Fig. 2, Table 2; Bernet et al., 2004), which were selected to test for variability between samples collected from the same general part of a depositional system. The samples were collected to the east (Fig. 2, sample A, Fos-sur-Mer), the west (Fig. 2, sample B, Stes. Maries-de-la-Mer), and near the modern main channel of the Rhône delta (Fig. 2, sample C, Plage de Piémanson). All samples were collected from heavy mineral placer deposits along the shore face.

The fission-track grain-age distributions for these three samples have a similar range in ages, but the cumulative probability plots (Fig. 10A) show that sample A is significantly different from samples B and C, as indicated by the KS test. The character of each fission-track grain-age distribution is illustrated in more detail by the probability density plots (Fig. 10B) and by the bestfit peaks (Table 1). Each distribution contains a similar set of four peaks. The main thing that distinguishes sample A from the other two samples is that its young peaks are relatively small and its old peaks relative to large. This difference may merely reflect shortterm fluctuations in yield, as discussed above. Note, however, that the two young peaks in sample A are significantly older, by at least 1 m.y., than the ages of the two young peaks in the other two samples. This difference may reflect the fact that the sediments for sample A came from a different part of the drainage relative to those represented by samples B and C. Despite these differences, the fission-track grain-age distributions for samples A, B, and C are remarkably similar. As result, we conclude that zircons moving through the Rhône drainage are well mixed and thus are able to deliver a fairly complete representation of the distribution of zircon fission-track cooling ages in the drainage.

CONCLUSIONS

The data from the Rhône delta samples indicate that detrital zircon fission-track analysis gives similar results for different samples within a single depositional system. By dating 60–100 single grains of a detrital sample, using the multi-mount technique with different etch times, it is possible to detect all of the principle cooling-age components in a drainage area. The relative sizes of the peaks in the distribution are based on yield, which includes the erosion rate, lithology, and the size of area of the zircon sources at that time of erosion. In a number of drainage-spe-

cific case studies, the detrital fission-track results make sense on a local as well as on a regional scale, so we are encouraged that this technique can be applied to other sedimentary sequences.

In a broader context, the observations in our study on zircons of modern river sediment from an orogenic belt support the results of other workers from ancient stratigraphic sequences. One of the real promises of the fission-track grain-age technique is its ability to address the long-term exhumation of orogenic belts, because sediments in a stratigraphic sequence capture a representative picture of source exhumation through time (Cerveny et al., 1988; Brandon and Vance, 1992; Garver and Brandon, 1994b; Lonergan and Johnson, 1998; Carter and Moss, 1999; Garver et al., 1999; Carter and Bristow, 2000; Spiegel et al., 2000; Bernet et al., 2001). At this point, we can be reasonably assured that the basin strata captures representative samples of the orogenic belt through time, and there is a reasonably quantitative transfer of material. An outstanding issue for those studies that utilize stratigraphic sequences is the temporal variation of drainage basins and the effects of long-term sediment storage within or adjacent to the orogenic belt. Nonetheless, it seems clear that future studies will continue to advance our understanding of orogenic exhumation from cooling ages in the sedimentary record, and therefore it is important that the potential influences are understood and quantified.

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REFERENCES CITED

- Bernet, M., Zattin, M., Garver, J.I., Brandon, M.T., and Vance, J.A., 2001, Steady-state exhumation of the European Alps: Geology, v. 29, p. 35–38.
- Bernet, M., Brandon, M.T., Garver, J.I., and Molitor, B., 2004, Downstream changes of Alpine zircon fission-track ages in the Rhône and Rhine rivers: Journal of Sedimentary Research, v. 74, p. 82–94.
- Bertotti, G., Seward, D., Wijbrans, J., ter Voorde, M., and Hurford, A.J., 1999, Crustal thermal regime prior to, during, and after rifting: A geochronological and modeling study of the Mesozoic South Alpine rifted margin: Tectonics, v. 18, p. 185–200.
- von Blanckenburg, F. 1992. Combined high-precision chronometry and geochemical tracing using accessory minerals; applied to the Central-Alpine Bergell Intrusion (Central Europe): Chemical Geology, v. 100, p. 19–40.
- Brandon, M.T., 1992. Decomposition of fission-track grain-age distributions: American Journal of Science, v. 292, p. 535–564.
- Brandon, M.T., 1996, Probability density plot for fission track grain-age samples: Radiation Measurements, v. 26, p. 663–676.
- Brandon, M.T., 2002. Decomposition of mixed grain age distributions using BINOMFIT: On Track, v. 24, p. 13–18.

- Brandon, M.T., and Vance, J.A., 1992. New statistical methods for analysis of fission track grain-age distributions with applications to detrital zircon ages from the Olympic subduction complex, western Washington State: American Journal of Science, v. 292, p. 565–636.
- Brix, M.R., Stöckhert, B., Seidel, E., Theye, T., Thompson, S.T., and Küster, M., 2002. Thermobarometric data from a fossil zircon partial annealing zone in high pressure-low temperature rocks of eastern and central Crete, Greece: Tectonophysics, v. 349, p. 309–326.
- Carter, A., 1999, Present status and future avenues of source region discrimination and characterization using fission-track analysis: Sedimentary Geology, v. 124, p. 31–45.
- Carter, A., and Bristow. C.S., 2000. Detrital zircon geochronology: Enhancing the quality of sedimentary source information through improved methodology and combined U-Pb and fission-track techniques: Basin Research, v. 12, p. 47–57.
- Carter, A., and Moss, S.J., 1999. Combined detrital-zircon fission-track and U-Pb dating: A new approach to understanding hinterland evolution: Geology. v. 27, p. 235–238.
- Cerveny, P.F., Naeser, N.D., Zeitler, P.K., Naeser, C.W., and Johnson, N.M., 1988, History of uplift and relief of the Himalaya during the past 18 million years: Evidence from fission-track ages of detrital zircons from sandstones of the Siwalik Group, *in* Kleinspehn, K., and Paola, C., eds., New perspectives in basin analysis: New York, Springer-Verlag, p. 43–61.
- Deer, W.A., Howie, R.A., and Zussman, J., 1992, An introduction to the rockforming minerals (2nd ed.): Essex, England, Longman Scientific and Technical, 696 p.
- Dunkl, I., Spiegel, C., Kuhlemann, J., and Frisch, W., 2000. Impact of the volcanism on age-provenance studies—The Periadriatic event in the Alpine Molasse, *in* Noble, W.P., O'Sullivan, P.B., and Brown, R.W., eds., The 9th International Conference of Fission-track Dating and Thermochronology. Lorne Australia, February 6–11, 2000: Geological Society of Australia Abstracts, v. 58, p. 75–76.
- Flisch, M., 1986, Die Hebungsgeschichte der oberostalpinen Silvretta-Decke seit der mittleren Kreide: Bulletin Vereinigung Schweizer Petroleum-Geologen und Ingenieure, v. 53. p. 23–49.
- Fügenschuh, B., Loprieno, A., Ceriani, S., and Schmid, S.M., 1999, Structural analysis of the Subbriabçonnais and Valais units in the area of Moûtiers (Savoy, Western Alps): Paleogeographic and tectonic consequences: International Journal of Earth Sciences, v. 88, p. 201–218.
- Galbraith, R.F., and Green, P.F., 1990, Estimating the component ages in a finite mixture: Nuclear Tracks and Radiation Measurements. v. 17, p. 197–206.
- Garver, J.I., and Brandon, M.T., 1994a. Fission-track ages of detrital zircon from Cretaceous strata, southern British Columbia: Implications for the Baja BC hypothesis: Tectonics, v. 13, p. 401–420.
- Garver, J.I., and Brandon, M.T., 1994b, Erosional denudation of the British Columbia Coast Ranges as determined from fission-track ages of detrital zircon from the Tofino Basin, Olympic Peninsula, Washington: Geological Society of America Bulletin, v. 106, p. 1398–1412.
- Garver, J.I., and Kamp, P.J.J., 2002. Integration of zircon color and zircon fission track zonation patterns in Orogenic belts: Application of the Southern Alps, New Zealand: Tectonophysics, v. 349, p. 203–219.
- Garver, J.I., Brandon, M.T., Roden-Tice, M.K., and Kamp, P.J.J., 1999, Exhumation history of orogenic highlands determined by detrital fission track thermochronology. *in* Ring, U., Brandon, M.T., Willett, S.D., and Lister, G.S., eds., Exhumation processes: Normal faulting, ductile flow, and erosion: Geological Society [London] Special Publication 154, p. 283–304.
- Garver, J.I., Brandon, M.T., Bernet, M., Brewer, I., Soloviev, A.V., Kamp, P.J.J., and Meyer, N., 2000a. Practical considerations for using detrital zircon fission track thermochronology for provenance, exhumation studies, and dating sediments, *in* Noble, W.P., O'Sullivan, P.B., and Brown, R.W., eds., The 9th International Conference of Fission-track Dating and Thermochronology, Lorne Australia, February 6–11, 2000: Geological Society of Australia Abstracts, v. 58, p. 109–111.
- Garver, J.I., Soloviev, A.V., Bullen, M.E., and Brandon, M.T., 2000b. Towards a more complete record of magmatism and exhumation in continental arcs using detrital fission-track thermochronometry: Physics and Chemistry of the Earth, Part A, v. 25, no. 6–7, p. 565–570.
- Gleadow, A.J.W., 1978. Comparison of fission-track dating methods: effects of anisotropic etching and accumulated alpha-damage. *in* Zartman, R.E., ed., Short papers of the 4th International Conference on Geochronology. Cosmochronology and Isotope Geology, Snowmass, Colorado, August 1978: U.S. Geological Survey, Open File Report 78-701.

- Gleadow, A.J.W., Hurford, A.J., and Quaife, R.D., 1976. Fission-track dating of zircon—Improved etching techniques: Earth and Planetary Science Letters, v. 33, p. 273–276.
- Giger, M., and Hurford, A.J., 1989, Tertiary intrusives of the Central Alps: Their Tertiary uplift, erosion, redeposition and burial in the south-alpine foreland: Eclogae Geologicae Helvetiae, v. 82, no. 3, p. 857–866.
- Gray, M.B., and Zeitler, P.K., 1997, Comparison of clastic wedge provenance in the Appalachian foreland using U/Pb ages of detrital zircons: Tectonics. v. 16, 151–160.
- Hunziker, J.C., Desmond, J., and Hurford, A.J., 1992, Thirty-two years of geochronological work in the Central and Western Alps: A review on seven maps: Memoire De Geologie (Lausanne), v. 13, p. 1–59.
- Hurford, A.J., 1986, Cooling and uplift patterns in the Lepontine Alps. South Central Switzerland and an age of vertical movement on the Insubric fault line: Contributions to Mineralogy and Petrology, v. 92. p. 413–427.
- Hurford, A.J., and Carter, A., 1991, The role of fission track dating in discrimination of provenance, *in* Morton, A.C., Todd, S.P., and Haughton, P.D.W., eds., Developments in sedimentary provenance studies: Geological Society [London] Special Publication 57, p. 67–78.
- Hurford, A.J., and Hunziker, J.C., 1985. Alpine cooling history of the Monte Mucrone Eclogites (Sesia-Lanzo Zone): fission-track evidence: Schweizerische Mineralogische und Petrographische Mitteilungen. v. 65. p. 325–334.
- Hurford, A.J., and Hunziker, J.C., 1989, A revised thermal history for the Gran Paradiso massif: Schweizerische Mineralogische und Petrographische Mitteilungen, v. 69, p. 319–329.
- Hurford, A.J., Fitch, F.J., and Clarke, A., 1984. Resolution of the age structure of the detrital zircon populations of two Lower Cretaceous sandstones from the Weald of England by fission track dating: Geological Magazine, v. 121, p. 269–277.
- Hurford, A.J., Hunziker, J.C., and Stöckhert, B., 1991. Constraints on the late thermotectonic evolution of the Western Alps: Evidence for episodic rapid uplift: Tectonics, v. 10, p. 758–769.
- Ireland, T.R., 1992. Crustal evolution of New Zealand: Evidence for age distributions of detrital zircons in Western Province paragneiss and Torlesse greywacke: Geochimica et Cosmochimica Acta, v. 56, p. 911–922.
- Kasuya, M., and Naeser, C.W., 1988, The effect of α-damage on fission-track annealing in zircon: Nuclear Tracks and Radiation Measurements, v. 14, p. 477–480.
- Lonergan, L., and Johnson, C., 1998. Reconstructing orogenic exhumation histories using synorogenic zircons and apatites: An example from the Betic Cordillera, SE Spain: Basin Research, v. 10, p. 353–364.
- Michalski, I., and Soom, M., 1990, The Alpine thermo-tectonic evolution of the Aar and Gotthard massifs, Central Switzerland: Fission-track ages on zircon and apatite and K-Ar mica ages: Schweizerische Mineralogische und Petrographische Mitteilungen, v. 70, p. 373–387.

- Naeser, N.D., Zeitler, P.K., Naeser, C.W., and Cerveny, P.F., 1987. Provenance studies by fission track dating of zircon—Etching and counting procedures: Nuclear Tracks and Radiation Measurements, v. 13, p. 121–126.
- Palenik, C.S., Nasdala, L., and Ewing, R.C., 2003, Radiation damage in zircon: American Mineralogist, v. 88, p. 770–781.
- Poldervaart, A., 1955, Zircon in rocks 1, Sedimentary rocks: American Journal of Science, v. 235, p. 433–461.
- Poldervaart, A., 1956. Zircon in rocks 2. Igneous rocks: American Journal of Science. v. 234, p. 521–554.
- Press, W.H., Teukolsky, S.A., Vetterling, W.T., and Flannery. B.P. 1992. Numerical recipes in FORTRAN, 2nd ed.: New York. Cambridge University Press, 963 p.
- Schäfer, H.-J., Gebauer, D., Naegler, T., and Eguiluz, 1995. Conventional and ion-microprobe U-Pb dating of detrital zircon of the Tentudia Group (Serie Negra, SW Spain): Implications for zircon systematics. stratigraphy, tectonics, and the Precambrian/Cambrian boundary: Contributions to Mineralogy and Petrology, v. 113, p. 289–299.
- Seward, D., and Mancktelow, N.S., 1994. Neogene kinematics of the central and western Alps: Evidence from fission-track dating: Geology. v. 22. p. 803–806.
- Spiegel, C., Kuhlemann, J., Dunkl, I., Frisch, W., von Eynatten, H., and Balogh, K., 2000. The erosion history of the Central Alps: Evidence from zircon fission-track data of the foreland basin sediments: Terra Nova. v. 12, p. 163–170.
- Stewart, R.J., and Brandon, M.T., 2004. Detrital-zircon fission-track ages for the "Hoh Formation": Implications for late Cenozoic evolution of the Cascadia subduction wedge: Geological Society of America Bulletin. v. 116, p. 60–75.
- Tagami, T., Carter, A., and Hurford, A.J., 1996. Natural long term annealing of the zircon fission-track system in Vienna Basin deep borehole samples: Constraints upon the partial annealing zone and closure temperature: Chemical Geology, v. 130, p. 147–157.
- Zeitler, P.K., Johnson, M.N., Briggs, N.D., and Naeser, C.W., 1986. Uplift history of the NW Himalaya as recorded by fission-track ages of detrital Siwalik zircons, *in* Jiqing, H., ed., Proceedings of the Symposium on Mesozoic and Cenozoic Geology: Beijing, Geological Publishing House. p. 481–494.
- Zhao, J.X., McCulloch, M.T., and Bennett, V.C., 1992, Sm-Nd and U-Pb zircon isotopic constraints on the provenance of sediments from the Amadeus basin. Central Australia: evidence for REE fractionation: Geochimica et Cosmochimica Acta, v. 56. p. 921–940.

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