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► **To cite this version:**

Matthias Bernet, Mark T. Brandon, John I. Garver, Brandi Molitor. Downstream changes of Alpine zircon fission-track ages in the Rhône and Rhine rivers.. *Journal of Sedimentary Research*, 2004, 74, pp.82-94. hal-00097147

HAL Id: hal-00097147

<https://hal.science/hal-00097147>

Submitted on 21 Sep 2006

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Alpine zircon fission-track ages in the Rhône and Rhine rivers

DOWNSTREAM CHANGES OF ALPINE ZIRCON FISSION-TRACK AGES IN THE RHÔNE AND RHINE RIVERS

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Keywords: detrital, zircon, fission-track, Rhône River, Rhine River

Manuscript received:

ABSTRACT

Zircons from 13 sediment samples from the Rhône and Rhine drainages were dated by the fission-track method to study downstream changes in detrital fission-track grain-age distributions in large river systems draining the European Alps. The orogen-parallel Rhône River shows a zircon fission-track grain-age distribution similar to that in its Alpine source areas. This signal is well preserved because there are Alpine sources along its full length and input from non-Alpine sources is small. In contrast, only the headwaters of the north-flowing Rhine River are located in the Alps. As a consequence, Alpine-derived zircons, which are distinguished by young fission-track ages, become progressively diluted downstream by older ages from zircon sources external to the Alps. Nevertheless, the Alpine component is persistent and can easily be detected in sediments more than 1000 km downstream at the Rhine delta. These results demonstrate that fission-track dating of detrital zircons can provide useful information about orogenic processes, even where sediments have been transported hundreds of kilometers from the orogenic source, crossing ephemeral lakes and subsiding basins. Deposits along the lower reaches of the river appear to have a short-residence time (< 1 Myr), and thus many of these deposits serve to smooth out variations in the supply of sediment from fast-eroding heterogeneous sources in the Alpine headwaters of these drainages. For the Rhône and the Rhine rivers, we show that fast erosion in the Alps accounts for most of the sediment load. This finding supports a widespread observation that the sediment in most large continental drainages is usually derived from a small part of the drainage, where uplift, relief, and erosion rates are greatest.

INTRODUCTION

Heavy-mineral assemblages have long been used in provenance studies of siliciclastic sediments because they contain a wealth of information about the source region, as well as transport processes (Pettijohn et al. 1987). In this respect, zircon is one of the most useful heavy minerals because it is common in many source rocks, resistant to weathering and abrasion, and datable by various methods. We focus here on dating of detrital zircon grains using the fission-track (FT) method. Other methods, such as $^{40}\text{Ar}/^{39}\text{Ar}$ dating of mica and potassium feldspar, have also proven useful for detrital thermochronologic studies (e.g., Copeland and Harrison 1990; Heller et al. 1992). Hurford et al. (1984) conducted the first provenance study using detrital zircon FT ages, and useful reviews on the provenance aspect of detrital FT analysis can be found in Hurford and Carter (1991), Carter (1999), and Garver et al. (1999). As a provenance method, detrital FT dating provides information about the thermal and exhumational history of the source region (Baldwin et al. 1986; Cervený et al. 1988; Brandon and Vance 1992; Garver and Brandon 1994a, 1994b; Lornegan and Johnson 1998; Garver et al. 1999; Spiegel et al. 2000; Bernet et al. 2001; Stewart and Brandon 2003).

In this paper, we use modern orogenic sediments carried by two large rivers, the Rhône and Rhine rivers, from sources in the Alps to see whether transport and storage in these continental-scale drainages have a detectable influence on detrital zircon fission-track grain-age (FTGA) distributions. This issue is important given that detrital thermochronology studies of orogenic regions tend to focus on marine sediments (e.g., shallow marine deposits, turbidites, etc.), which commonly are deposited at a distance from the orogenic source. The advantage of using marine sediments for such studies is that they are well mixed and thus are able to provide

information about the orogen at its largest scale. Furthermore, marine fossils provide an independent method for determining depositional ages, in some cases to a precision of ± 1 Myr. Long transport represents a problem in that sediments from the orogenic source are subject to storage, and reworking within the river system, and dilution by sediments from tributary drainages.

We present here FT data for detrital zircons from 13 samples of modern river sediments collected along the Rhône and Rhine rivers (Fig. 1). Each sample is represented by a FTGA distribution composed of 60 to 100 grain ages. An advantage of these rivers is that the bedrock within their drainages has been studied widely by thermochronometric methods. In particular, the distribution of zircon FT ages in bedrock exposures of the Alps is well known (see Hunziker et al. 1992 and Bernet et al. 2001 for data and map compilations). Thus, we can compare detrital zircon FTGA distributions from river samples directly to modern bedrock cooling ages in the source region (Fig. 2).

The Rhône and Rhine rivers have contrasting drainage patterns. The Rhône River follows the curved front of the Swiss and French Alps, and ultimately empties into the Mediterranean Sea. Thus, it receives Alpine sediment along its entire course. The Rhine River flows northward, perpendicular to the Alps, crossing the Rhine Graben and lowlands of Germany and the Netherlands on its course to the North Sea. Both rivers are interrupted by major lakes along their courses: Lake Geneva for the Rhône River and Lake Constance for the Rhine River. These lakes provide an opportunity to see how sediment storage might influence FTGA distributions.

RESEARCH OBJECTIVE

Ideally, we would like the sample distribution of zircon FTGA to be an unbiased representation of the distribution of zircon cooling ages exposed over the area of the erosional source from which the zircons were derived. This expectation is compromised by several factors. First is the fact that the dated detrital zircons provide information about the age distribution relative to yield of sediment, whereas the distribution of zircon FT ages in the source region is usually viewed relative to the area of the source region. Thus, one must make a clear distinction between a distribution of cooling ages by area and by yield. Detrital zircon FTGA distributions are always determined relative to yield, so that the fastest and slowest eroding parts of the source area will be overrepresented and underrepresented, respectively, relative to the area that they occupied in the drainage.

A second factor is that zircon concentrations in different lithologies can vary considerably. For instance, carbonates and mafic igneous rocks typically have little to no zircon, whereas quartz-bearing rocks typically contain abundant zircon. This issue can usually be assessed by inspection of geologic maps of the source region. Also, local variations tend to average out if the active parts of the source region are large relative to the average length scale for lithologic variation.

A third factor concerns the FT method, in that grains have to be large enough (greater than about 70 μm) to be dated. Also, grains with very old (> 500 Ma) or very young cooling (less than about 5 Ma) ages can be difficult to date, depending on the uranium content, track density, and radiation damage (Garver et al. 2000). In general, we have found that these limitations are not a significant problem for most studies. Furthermore, it is easy during dating to identify those samples where the FT method might introduce a selection bias.

The influences discussed so far distort the relationship between the area distribution of bedrock cooling ages in the source region and the yield distribution of detrital cooling ages determined for a sediment sample. However, these influences do not cause any shift in ages in the zircon FTGA distribution. Some explanation is needed to appreciate this point. Experience indicates that FTGA distributions of detrital zircons tend to show clusters of ages. Individual clusters can commonly be correlated to specific geologic regions in the source area, each with a characteristic FT cooling age. Brandon and Vance (1992) referred to such regions as FT source terranes. The clustering in the distribution can be formally analyzed using the binomial peak-fitting method, which breaks the FTGA distribution into a set of peaks or components, with each component defined by an estimated age and size (Galbraith and Green 1990; Brandon 1992, 1996; Stewart and Brandon 2003). A FT source terrane might be a rapidly exhumed metamorphic core complex, a short-lived volcanic province, or the eroded core of an orogenic belt (e.g., Brandon and Vance 1992; Bernet et al. 2001).

The conclusion so far is that the factors discussed above might influence the relative sizes of peaks in distribution, but not the ages of the peaks. As a result, our work in the Alps (Bernet et al. 2001) has emphasized peak ages rather than peak size, because peak ages are a more robust feature of the zircon FTGA distributions. In particular, we have focused on the lag time of the peaks, defined as the difference between FT peak age and deposition. Lag time provides useful information about exhumation rates, assuming that the FT ages record exhumation-related cooling, due to normal faulting or erosion (Garver and Brandon 1994a; Garver et al. 1999; Bernet et al. 2001). This interpretation, however, assumes that the transport time after exhumation, which is the time involved in moving the zircons from the site of

erosion to the site of deposition, is a negligible fraction of the measured lag time. This issue has been addressed by comparing depositional ages to detrital zircon FT ages for volcanic zircons derived from contemporaneous sources (Brandon and Vance 1992; Stewart and Brandon 2003), and the transport times were insignificant in those cases. Nonetheless, there has been no examination yet about how large continental-scale river systems might modify a detrital FTGA distribution. The main concerns are that the distribution might become distorted because of recycling of sediment from ephemeral deposits in floodplains, hillslopes, and lakes, and also by dilution from erosional sources distant from the main orogenic source region.

SAMPLE PROCESSING

The 13 detrital zircon FTGA distributions presented here (Tables 1 and 2; also see data repository for single grain ages) were determined for samples from modern sand and gravel bars in the rivers and along the beach for delta samples. For the Rhône drainage, we focused on the downstream contribution of tributary sources. We sampled the Rhône River in its middle part below Lake Geneva and also four large tributary drainages by collecting sediment just upstream of their confluences with the main channel of the Rhône River. Delta sediments were collected in three locations around the mouth of the Rhône River. For the Rhine drainage, we have four samples that are used to characterize the Alpine part of the drainage, and two other samples, one collected from the middle part of the river and one collected from delta sediments near the mouth of the river.

All samples were collected from heavy-mineral placer deposits and were panned in the field to provide an initial concentrate of the heavy-mineral fraction. In the laboratory, zircons were separated from the bulk samples using standard heavy-

liquid and magnetic separation techniques (Naeser 1976). Aliquots of separated zircons were mounted in 4 cm² Teflon[®] sheets and then polished with 9 μm and 1 μm diamond paste to expose internal zircon surfaces. Subsequently, the mounted zircons were etched in a NaOH/KOH eutectic melt at 228°C in Teflon[®] dishes, in a thermostatically controlled laboratory oven. Following Naeser et al. (1987) and Garver et al. (2000), we used the "multi-mount technique" with 2 to 5 mounts per sample to account for the fact that detrital zircons typically have a range of cooling ages and uranium contents. For most samples, two mounts were prepared. One mount was etched for 24 hours (long etch) and the other for 10 hours (short etch). Each mount contained 500-1000 zircons, depending on available sample material. All mounts were irradiated at the Oregon State University reactor, along with CN5 glass standards and Fish Canyon Tuff and Buluk Tuff zircon standards (Hurford 1990).

For each mount, grains with flat surfaces were marked for counting using reflected light at 125x magnification. The mounting process ensured that grains were randomly distributed within the mounts with respect to their cooling ages and uranium content, which helps to avoid sampling bias when counting. Marked grains were counted at 1250x (dry) using an Olympus BH-60 microscope. Our goal was typically about 60 to 100 dated grains per sample, with a roughly equal proportion of dated grains from each mount.

We saw little evidence of correlation between the countability of zircons and their source. For instance, the long-etch and short-etch mounts gave similar grain-age distributions. Thus, the FTGA distributions presented here were constructed by combining grain ages from all mounts for each sample. Note that our study contrasts with that of Cervený et al. (1988) in that the FTGA distributions for their detrital

zircons were strongly influenced by the amount of time that the zircon mount was etched. The reason is that their samples, which were derived from fast-exhuming sources in the Himalaya, contained a large fraction of very young FT ages with low radiation damage. As noted above, very young, low U zircons are difficult to date because the low radiation damage means that they etch very slowly, so that their fission tracks etched properly only after prolonged etch times (Naeser et al. 1987; Kasuya and Naeser 1988).

THE RHÔNE DRAINAGE

The Rhône River flows from the Central Alps in Switzerland through the Rhône graben until it reaches the Rhône delta in the Gulf of Lyon in the Mediterranean Sea (Fig.1). The total length of the Rhône River is ~ 810 km, and the total drainage area is ~ 99,000 km² (Allen 1997). Sediment yield at the Rhône delta is currently ~ 60 x 10⁹ kg/yr (Allen 1997). An additional ~ 2.25 x 10⁹ kg/yr is currently being trapped upstream in ephemeral lakes and behind dams (see below). Thus, the total modern yield on an open river would be ~ 62 x 10⁹ kg/yr, which is equivalent to an average erosion rate for the entire drainage of 0.23 mm/yr, using a typical bedrock density of 2700 kg/m³. For comparison, long-term erosion rates in the Alpine part of the drainage are about 0.4 mm/yr (Bernet et al. 2001).

Approximately 60% of the Rhône drainage lies in the Alps, and 40% is in the Rhône and Bresse grabens, parts of the Massif Central, and the Jura Mountains. The main tributaries of the Rhône are: (1) the Saône River, which flows from the north, draining parts of the Bresse graben and the Jura Mountains; (2) the Isère River, the Drôme River and the Durance River, which drain the Western Alps; and (3) the Ardèche River, which drains the Massif Central (Fig. 1). Van Andel (1955)

showed that near the Rhône delta, ~ 80% of the modern sediment load comes from Alpine sources, and ~ 20% is derived from the Massif Central and Jura Mountains.

The Drôme and Saône rivers drain areas underlain mainly by carbonate bedrock, which commonly have a poor zircon yield (Poldervaart 1955). Even after intense sampling, the Drôme River provided only a trace of zircon, which was not enough to be dated. For this reason, the Saône River was not sampled. Abundant datable zircon was found in all other sampled tributaries, the Isère, Durance, and Ardèche rivers (Fig.1).

The Rhône and Bresse grabens started forming in the Oligocene, and have accumulated sediment fairly steadily to the present, with an average rate of 0.68×10^9 kg/yr and a Quaternary rate of 0.55×10^9 kg/yr (Hay et al. 1992; Kuhlemann 2000). The grabens represent the largest sedimentary basin within the Rhône drainage, but this sink accounts for < 1% of the modern sediment flux moving through the drainage.

THE RHINE DRAINAGE

The Rhine River originates in the Central Alps of Switzerland and flows northward through the Rhine graben and Renish massif until it reaches the North Sea coast near Rotterdam (Fig.1). The Rhine has a total length of ~ 1360 km and a drainage area of ~ 225,000 km² (Allen 1997). The modern sediment yield at the Rhine delta is ~ 3×10^9 kg/yr (Asselman 1997). Another 3×10^9 kg/yr is currently being trapped in Lake Constance (Spreafico 1999). Therefore, the total modern yield without storage is ~ 6×10^9 kg/yr, which is equivalent to an average erosion rate for the entire drainage of 0.01 mm/yr.

The Alpine part of the Rhine is occupied by two tributaries to the west, the Reuss and Aare rivers, and the main trunk of the Rhine River to the east. In each case, the mountainous stretch of the river flows through a glacially scoured lake. The largest, Lake Constance, is located on the main trunk of the Rhine. The other lakes are much smaller, Brienzer Lake and Thuner Lake on the Aare River, and Vierwaldstätter Lake on the Reuss River. The Inn River, a tributary of the Danube River, occupies the next drainage to the east of the Rhine River. We call attention to the Inn River here because one of our samples was collected there, to provide more information about spatial variations within the Alpine part of the Rhine drainage.

The Alpine part of the Rhine, defined as the area above the confluences of the Aare and Reuss rivers and above Lake Constance, makes up only 4% of the drainage, but it has the highest topography and fastest erosion. Sediment trapped in Lake Constance represents mainly the yield from the Alpine part of the drainage. Thus, we can conclude that half of the long-term yield of the Rhine comes from only 4% of the drainage area. This conclusion does not account for the sediment trapped in the smaller Alpine lakes noted above. The long-term area-averaged erosion rates are 0.12 mm/yr or more for the Alpine part and 0.005 mm/yr for the lowland part of the drainage, which is smaller by a factor of 24.

The Rhine graben represents the largest sedimentary basin within the Rhine drainage. It was initiated during the Oligocene, but it appears to have had a more punctuated history, as indicated by sediment accumulation rates (Hay et al. 1992; Kuhlemann 2000). Quaternary accumulation rates are 0.16×10^9 kg/yr, which is equal to $\sim 3\%$ of the modern sediment flux moving through the Rhine River. In comparison, accumulation rates were fastest during the Early Miocene, reaching 3.8×10^9 kg/yr, which is equivalent to $\sim 60\%$ of the modern yield.

SEDIMENT STORAGE

Our objective in this paper is to understand how well zircon FTGA distributions image the area distribution of zircon FT ages exposed in the source region. Storage and reworking of sediments is thus an important issue. There are two potential problems that we need to consider. The first is that storage can modulate the relative contribution of zircons from different parts of the drainage. The second is that reworking of stored sediment introduces zircons that have “aged” while in storage. Sites for storage, such as lakes, alluvial banks, floodplains, and sedimentary basins, tend to be more common in the lower reaches of a drainage. We can view this storage as increasing the residence time of the sediment during transport through the fluvial system. We discuss below glacial lakes, which are scoured and filled on a glacial-interglacial time scale; the sediments in those settings have a 50 to 100 kyr residence time.

A common feature of many rivers is cycles of aggradations and incision, alternating on time scales of 1 kyr to 100 kyr (e.g., Bull 1991), which means that some of the sediment stored adjacent to a river is reworked on that time scale. Meandering of the river channel will also cause reworking of older floodplain deposits.

In fact, tectonic subsidence, and significant accumulation in basin rocks is the only way by which sediments can avoid recycling in terrestrial environments. The Rhine, Bresse, and Rhône grabens are examples of long-term storage caused by tectonic subsidence. Tectonic uplift can cause inversion of basin sequences, resulting in the release of old sediments back into the drainage system. An example

of this situation is the uplift and erosion of foreland basin strata along the northern and western foothills of the Alps (Kuhlemann and Kempf 2002).

Thus, we make a distinction between temporary storage and long-term storage. Storage is considered temporary if sediment is reworked within a short time frame, here set to be about 1 Myr. This choice is based on the fact that we cannot detect storage that is shorter than 1 Myr using our zircon FT measurements. Other studies might focus on shorter-term variations in storage. For instance, cosmogenic studies of drainage-scale erosion rates are sensitive to variations in storage on the 10 kyr time scale (Schaller et al. 2001, Schaller et al. 2002).

Sediment Storage in Lakes

Lake Geneva on the Rhône River and Lake Constance on the Rhine River provide good natural examples to evaluate the influence of storage. Both lakes are Pleistocene in age and have formed by repeated scouring and filling during glacial and interglacial events (Trümpy 1980; Jerz 1993; Ehlers 1996). The largest scouring events are tied to the major glacial advances in the Alps, occurring during the Mindel (~ 640-300 ka), Riss (~ 265-100 ka), and Würm (~ 70-12 ka) cold stages (Trümpy 1980; Jerz 1993; Ehlers 1996).

Lake Geneva has a current volume of 88.9 km^3 and an upstream drainage area of 5200 km^2 . At present, the sediment accumulation rate in the lake is $\sim 2 \times 10^9 \text{ kg/yr}$ (Spreafico 1999). Dams, built in the 1960s upstream of the lake, currently accumulate $\sim 0.25 \times 10^9 \text{ kg/yr}$ of sediment that would otherwise end up in the lake (Loizeau et al. 1997). Given the present total accumulation rate of $\sim 2.25 \times 10^9 \text{ kg/yr}$ and a typical sediment density of 1350 kg/m^3 , the lake would be filled in $\sim 53 \text{ kyr}$.

Lake Constance has a volume of 48.5 km^3 and an upstream drainage area of 6100 km^2 (Vetsch and Faeh 2001). At present, the lake accumulates sediment at an average rate of $3 \times 10^9 \text{ kg/yr}$ (Spreafico 1999). During years with large floods, that rate can increase to $10 \times 10^9 \text{ kg/yr}$. (Spreafico 1999). At the current accumulation rate, the lake would be filled in $\sim 22 \text{ kyr}$.

These lakes are short-lived and thus represent ephemeral sediment storages. Current sediment storage started during deglaciation and will continue until the lakes are filled by sediment or scoured again by a glacial advance. Thus, the duration of the interglacial period, which has been $< 30 \text{ kyr}$ in the Alps, indicates an even shorter lifespan for these lakes.

The youngest cooling ages in the Rhône and Rhine drainages lie in the Central Alps, upstream of Lake Geneva, Lake Constance, Brienz Lake, and Vierwaldstätter Lake. At present, nothing coarser than silt makes it through the lakes (Spreafico 1999). The principle of hydraulic equivalence indicates that within that silt size fraction the zircon grains are half the diameter of the quartz and feldspar grains. The FT method requires zircons that are greater than about $70 \mu\text{m}$ (at least fine-grained sand size), but zircons of this size are currently completely trapped. A key question is how detrital zircon FTGA distributions in the modern river sediments downstream of these lakes have been affected by this trapping, which has been going on since deglaciation, starting at $\sim 12 \text{ ka}$.

Sediment Storage in River Channels and Floodplains

Floodplains and channel deposits are an additional source of sediment along the river. The distribution of Holocene deposits along the Rhône and Rhine rivers provides a schematic illustration of the size of this storage region (Fig. 3). We show

below that Alpine zircons are present in modern sediments carried by the Rhine, all the way to the mouth of the river, despite the fact that Lake Constance is presently storing all datable Alpine zircons. The Alpine zircons must be coming from reworking of stored Alpine sediment, downstream of Lake Constance. Some simple calculations are given here to provide some context for this problem.

Below Lake Constance, the area of Holocene deposits adjacent to the Rhine is $\sim 10,860 \text{ km}^2$, on the basis of an average width of 9.7 km and a river length of 1120 km. The modern volumetric flux of sediment at the mouth of the Rhine River is $2.2 \text{ km}^3/\text{kyr}$. If all of that flux were derived by reworking of the Holocene deposits, it would be equivalent to an average erosion rate of 0.2 m/kyr. This rate indicates that the Holocene deposits are of sufficient size, relative to the sediment load carried by the river, to play a role in buffering the delivery of Alpine-derived sediment in the Rhine River.

The Rhône River is also blocked by a glacial lake, Lake Geneva, but the influence of this blockage would not so obvious in the zircon FTGA distributions given that there are other sources of Alpine sediment downstream from Lake Geneva. Flood-plain deposits probably also play a role in buffering sediment supply along the Rhône River. Below Lake Geneva, the area of Holocene deposits adjacent to the Rhône River is $\sim 2750 \text{ km}^2$, based on an average width of 5.0 km and a river length of 550 km. The modern volumetric flux of sediment at the mouth of the Rhône River is $44 \text{ km}^3/\text{kyr}$. The Holocene deposits would have to be eroded at a rate of 16.2 m/kyr to deliver that much sediment. From this, we conclude that the Rhône has a much smaller buffer capacity, given its higher sediment flux and smaller floodplain area.

RESULTS

The observed FTGA distributions are graphically illustrated using probability density plots (Figs. 4, 5), estimated by the Gaussian kernel method (Brandon 1996). In all cases, the spread in grain ages within each distribution is greater than expected for the analytical uncertainty of the ages alone. In other words, all distributions are mixtures of different grain-age components. The density plots show that these different components commonly appear as discrete well-defined peaks. These peaks or components were estimated (Tables 1, 2) using the binomial peak-fitting method (Galbraith and Green 1990; Brandon 1992, 1996; Stewart and Brandon, 2003). These results are compared with the geology of the drainages and the distribution of bedrock zircon FT ages.

The geology of the Rhône and Rhine drainages indicates that the zircon FT ages are related to exhumational cooling, associated with erosion or normal faulting, and not to post-magmatic cooling. The closure temperature for FT in zircon is about 240°C, assuming typical orogenic cooling rates of $\sim 15^\circ\text{C}/\text{Myr}$. (Brandon et al. 1998; Bernet et al. 2002). Steady exhumation at a rate of $\sim 1 \text{ mm}/\text{yr}$ would give a closure depth of 6.5 km and a surface cooling age of 6.5 Ma (Garver et al. 1999).

There has been no active volcanism in the Alps since the Oligocene, and only minor Cenozoic mafic magmatism in surrounding regions. Within the Rhône and Rhine drainages, Cenozoic magmatism is limited to 15-3.5 Ma basalts in the Massif Central of southeastern France (Pomerol 1980; Michon and Merle 2001), 15 Ma basalts of the Kaiserstuhl, and the Vogelsberg in the Rhine Graben, and upper Cenozoic basalts in the East-Eifel and the Neuwieder Becken in the Rhenish Massif (Wimmenauer 1985; Henningsen and Katzung 1992; Schöenberg and Neugebauer 1997). Given the mafic basalt composition, zircon should be rare, if present at all.

Another check for young volcanic sources is euhedral zircon, but our samples contain <1% of such grains.

Zircon yield in a detrital sample is influenced by the distribution of zircon-bearing rocks in the drainage. Zircon abundance is variable and depends on rock type (Poldervaart 1955, 1956). Carbonate sedimentary rocks and mafic igneous rocks have particularly low zircon concentrations. As noted, some parts of the Rhône and Rhine drainages are dominated by carbonates, and as a result contain little to no datable zircon (e.g., Drôme and Saône rivers). The Alps and European lowlands have a mix of lithologies at the local scale, so that the zircon-concentration distribution in the bedrock is probably fairly uniform at the scale of the Rhône and Rhine drainages.

Samples from the Rhône Drainage

The youngest ages in our zircon FTGA distributions are from the middle Rhône (peak P1 for sample A in Table 1), which is the most upstream sample collected from the Rhône drainage. The ~ 9 Ma peak is small, making up 6% of the distribution, but it is well resolved. This peak can only have been derived upstream of Lake Geneva, in the Central Alps (Fig. 2A), where normal faulting and erosion have exposed the most rapidly exhumed parts of the Alps (Hurford 1986; Hunziker et al. 1992; Seward and Mancktelow 1994; Bernet et al. 2001). This component may be small because of zircon trapping in Lake Geneva, or because of the small area of the source region with young zircon FT ages (Fig. 2A).

Alpine cooling ages, defined by zircons with ages less than ~ 30 to 35 Ma, dominate samples derived from the Central and Western Alps (A and B in Table 1). About 30 to 50% of the zircons have ages of ~ 15 Ma (P2 in Table 1). This cooling

age is common for rocks above the Simplon normal fault in the Central Alps, and reflects long-term steady-state exhumation at rates of ~ 0.4 mm/yr (Bernet et al. 2001). Similar zircon FT cooling ages are reported for the Western Alps for the Mont Blanc and Pelvoux-Belledonne massifs (Seward and Mancktelow 1994; Fügenschuh et al. 1999).

The Rhône FTGA samples show a distinct cluster of older Alpine cooling ages at ~ 25 to 35 Ma (P3 in Table 1). Zircons with these FT ages apparently were derived from more slowly exhumed metamorphic rocks from the flanks of the Alps or from recycling of orogenic sediments from the Alpine foredeep.

The Ardèche River drains the Massif Central and thus provides information about the cooling history of that source area. Sample D from there shows a small component of old Alpine ages (9% for P3, with a peak age of 24 Ma). Otherwise the sample is dominated by old ages, with peaks at ~ 103 Ma (74%) and 180 Ma (17%).

The Durance River drains the southern part of the Western Alps. Bedrock zircon FT ages are relatively old in this region (generally > 50 Ma; Fig. 2A), indicating slow exhumation in this part of the Alps. The detrital-zircon FTGA distribution for the Durance (E in Table 1) gives the same result, with most of the ages concentrated in peaks at 70 Ma (38%) and 107 Ma (50%).

The Rhône delta is represented by three samples collected from widely separated locations (F, G, and H in Fig. 1), spanning a distance of 50 km across the delta front. These samples help illustrate the degree of variation within the full population of detrital zircons in the delta sediments (Table 1, Fig. 6). The source of variation was evaluated using the KS test (Press et al. 1992), which assesses the probability that differences between two sample distributions might be due to chance alone. The KS statistic is defined by the maximum difference in probability between

the cumulative probability curves (Fig. 7). The differences between samples G and H are small relative to the variation expected by random sampling. The KS test indicates a 74% probability that the observed variation might be due to random chance alone. Sample F, however, is significantly different than the other two samples; the KS test indicates a probability of < 1% that this variation might be due to random chance alone. Note, however that all three samples have similar peaks, especially the prominent Cenozoic peaks at 12, 24, and 64 Ma. In fact, the main difference for sample F is that there is a higher abundance of grain ages in the older peaks. This difference in abundance of old grains could be related to short-term variation in sediment delivery from different parts of the source region.

The average zircon FTGA distribution for the Rhône delta can be represented by merging the grain ages of all three of our delta samples (Delta composite in Table1; Fig. 8). The peak-fitting results for this composite sample indicate that about half of the zircons in the delta samples come from sources with Alpine cooling ages.

The upstream samples are used to make a quantitative estimate of the relative contribution of those sources to the zircon population at the Rhône delta. For this analysis, we use a standard description for a mixture

$$\rho_m(\tau) = \sum \pi_j \rho_j(\tau). \quad (1)$$

In this case, we are using the probability density distributions $\rho(\tau)$ to represent the FTGA samples, where density ρ is given as a function of FT age τ . Equation 1 specifies that the density distribution of the mixture, $\rho_m(\tau)$, is a linear combination of the density distributions of the components of the mixture, $\rho_j(\tau)$, where the subscript indicates the j th component of the mixture.

We use a constrained weighted least-squares method to find best-fit estimates for the unknown proportions π_j . The probability density values for the FTGA distribution of the combined Rhône delta samples (F, G, H) are used for the mixture variable $\rho_m(\tau)$ (i.e. the dependent variable), the probability density values for the four upstream samples (A, B, D, and E) are the components of the mixture, $\rho_j(\tau)$ (i.e., independent variables). The observed mixture densities $\rho_m(\tau)$ are Poisson distributed and thus have standard errors proportional to $\sqrt{\rho_m(\tau)}$, which could be used to calculate relative weights for the dependent variable. The best-fit solution is defined by those values of π_j that minimize the misfit between observed and predicted values of $\rho_m(\tau)$ (Fig. 8). The solution is constrained so that the mixing proportions π_j are all positive and sum to 100%. The results are shown in Table 3.

The mixture model emphasizes our previous conclusion, that most of the zircons at the Rhône delta are derived from Alpine sources. In fact, our estimate is nearly identical to that of Van Andel (1955) (reviewed above), in that 77% of the zircons are coming from Alpine sources (Middle Rhône and Isère) and 23% from non-alpine sources (Massif Central, as represented by the Ardèche). Our analysis indicates that almost no zircons are derived from the Western Alps (Durance). As noted above, the Western Alps retain fairly old cooling ages (Fig. 2A), which suggests that, relative to the Central Alps, exhumation has overall been slow in the southern Western Alps.

Samples from the Rhine Drainage

The Alpine source of the Rhine drainage is represented by four samples (Table 2, Fig. 1 and 2B). Sample I, from the upper part of the Inn River, comes from

an area in the Alps just to the east of the upper Rhine drainage. The upper Rhine drainage itself is represented by samples J, K, and L, collected at Buchs on the Rhine River, and from the upper part of the Reuss and Aare rivers, which flow into the Rhine. All samples have a well-defined peak at ~ 17 Ma (Fig. 5). This young component comes from fast-exhuming parts of the Central Alps, which, in this case, are the eastern part of the Aar massif and the Gotthard massif (Hunziker et al., 1992). Samples I to L provide an east-to-west traverse of the Central Alps. Note that the size of the 17 Ma peak increases from 15% to 90% from east to west within this traverse (Table 2). This result reflects the fact that the fraction of young reset rocks in the landscape increases from east to west in the Central Alps. Three of the four samples show a variably developed peak at ~ 28 Ma. This peak is largest (49%) in the Buchs sample (J in Table 2). The Aar and Gotthard massifs represent the upstream source for those zircons.

Two Alpine peaks (17 and 33 Ma) occur in sample M, located about 500 km downstream at Worms on the lower part of the Rhine River. This result indicates that the Rhine graben (RG in Fig. 1) does not significantly influence the transport of zircons.

Sample N, collected near Rotterdam on the Rhine delta, shows a small peak (17%) at 17 Ma, and no significant peak at 33 Ma. The Alpine component is apparently reduced by dilution from older local sources (~ 150 Ma cooling ages), mainly in the lower reach of the Rhine, below sample N.

The mixture model can be used again to calculate the relative contributions of zircons from upstream sources to samples in the lower part of the Rhine drainage. The components of the mixture, $\rho_j(\tau)$, are represented by density distributions for Alpine samples J, K, and L, and these are used to find best-fit mixtures for

downstream density distributions $\rho_m(\tau)$ for samples M and N. In this case, we fit for proportions π_j for three components, but we note that we do not have an independent sample to represent the FT zircon component for sources external to the Alps. We get around this problem by removing the constraint that the best-fit proportions π_j sum to 100%. As a result, the external sources are represented by the difference between the observed and calculated $\rho_m(\tau)$ (Fig. 9).

The results of our mixture calculation (Fig. 9, Table 4) indicate that the fraction of Alpine zircons decreases from $\sim 70\%$ at Worms (sample M) to $\sim 20\%$ at the Rhine delta (sample N). In both cases, the Alpine component appears to be sourced above Buchs on the Rhine River and in the Aare drainage as well. The mixture calculation indicates almost no contribution from the Reuss River. The reason is that the Reuss sample has a large 17 Ma peak (85%, Table 2), which makes it incompatible as a mixture component for the downstream samples.

We estimate that the non-Alpine source of zircon has an average FT age of ~ 150 Ma. Mesozoic cooling ages are typical for old sedimentary and crystalline rocks outside of the Alps. For instance, Köppen and Carter (2000) report zircon FT ages from Triassic sandstone units of southwest Germany, which range from 160 to 450 Ma with mean ages between 260 and 310 Ma. Detrital zircon FT ages from Oligocene sandstones in the Rhine graben have detrital zircon FT ages that cluster around 80 and 150 Ma (Kuhlemann et al. 1999). Kuhlemann et al. (1999) estimated that zircon FT ages from Hercynian basement in the Vosges and Black Forest uplifts are about 250-300 Ma. There are no bedrock zircon FT ages for the Rhenish Massif, but apatite FT ages of 130-240 Ma (Glasmacher et al. 1996) indicate that detrital zircons from this area would probably have Mesozoic cooling ages.

DISCUSSION

The results presented above indicate that FTGA distributions for detrital zircon samples can provide reliable information about the thermal evolution of orogenic source regions, and that that information is preserved during transport within continental-scale rivers over distances of 500 to 1000 km. Two factors are responsible for this observation. First, much of the sediment transported within the drainages ultimately comes from the mountainous part of the drainage, where uplift and erosion are the fastest. Second, storage and recovery of sediment within the drainage serves to average out variations in sediment yield due to short-term events, such as storms, landslides, and floods. Floodplains represent an important setting where source variations are averaged out, because of aggradation and incision cycles, and river meandering. Cyclic scouring and filling of glacial lakes in front of the Alps is another process that averages out variations in sediment supplied from the source.

Temporary zircon storage within the drainage might significantly increase lag time, which is the amount of time between closure of the FT age and deposition. This question can be addressed in two ways. Let's start with a conceptual approach.

Consider the drainage as a system with influx controlled by erosion and outflux by discharge of sediment at the river mouth. The volume of the system is defined by the volume of actively moving sediment within the drainage. For this system, flux steady state is defined when the influx and outflux become equal, at which point the volume of the system remains steady. The residence time—the average time for material to move through the system—is defined at steady state by the volume of the system divided by the flux through the system (e.g., Rodhe 1992). Thus, a system with a high flux and a low volume has a short residence time at

steady state. The time to steady state is dependent on the influx of sediment and the system volume at steady state. Thus, given a perturbation, a system returns to steady state more quickly if it has a higher influx or a smaller volume at steady state.

The system volume—that is the volume of sediment actively moving downstream with the system—is poorly known but is bounded between 50 to 15,000 km³ (assuming an average density of 1350 kg/m³ for the sediment). The lower limit represents the size of the glacial lakes on the Rhône and the Rhine rivers, whereas the upper limit represents the total volume of Cenozoic sediment along the Rhine, as estimated by Ziegler (1982). The Rhône-Bresse grabens hold about 12,300 km³ of Cenozoic sediment, and the Rhine graben, about 7,750 km³ (integrated from Hay et al. 1992). The modern flux of eroded sediment is 46×10^6 and 4×10^6 m³/yr for the Rhône River and the Rhine River, respectively. Using the range of sediment volumes above, these flux values indicate that the residence times for sediment transported in these rivers is in the range 1 kyr to 3.75 Myr.

The FT data provide another perspective on this issue. If the residence time for zircons in the river is long, then peak ages should get progressively older downstream. Peak ages reported in Tables 1 and 2 have large uncertainties, equal to about 10 to 15% of the age at the 95% confidence level. The uncertainties for FT ages increase proportional to age, so relative uncertainties are useful in that they are approximately constant, independent of age. Thus, our youngest peak ages have the best precision. The Rhône River samples show a slight decrease in peak age downstream, opposite of expected for a long residence time. The Rhône River, however, is complicated because it has young sources of zircon along a large part of its length. The Rhine River is a better test because it drains northward, away from the young Alpine sources. Linear regression of P2 ages as a function of upstream

distance (Fig. 10) indicate that the P2 peak age increases by 0.8 ± 1.2 Myr (68% confidence interval) over the 1000 km distance represented by our samples. Therefore, residence time is not significantly different from zero. Nonetheless, we use the best-estimate residence time to illustrate how this quantity relates to the effective transport volume for the river system. The residence time of 0.8 Myr times the flux of sediment through the system (4×10^6 m³/yr) would imply an effective volume of actively transporting sediment equal to 3200 km³. This transport volume can be visualized as equivalent to moving the upper 280 m of Holocene sediment adjacent to the Rhine (which covers an area of 11,400 km², Fig. 3). The average downstream velocity is slow, of course, equal to 1.25 km per thousand years (assuming that the 0.8 Myr residence time estimate is correct).

This analysis has implications for detrital FT thermochronology. In areas where cooling ages reflect exhumation (i.e., not magmatic or volcanic sources), lag times for detrital FT zircons can range from a few million years (e.g., Himalayan sources; Cervený et al. 1988) to 8 Myr and longer in the Alps (Bernet et al. 2001). For scale, a typical exhumation rate of 1 mm/yr would give zircon FT cooling ages of ~ 7 Ma, given a geothermal gradient of 30°C/km (Garver et al. 1999). In comparison, the residence time of zircons during fluvial transport is below the level of uncertainty of the FT ages themselves (i.e., < 1 to 2 Myr). Thus, we can anticipate that FT lag times for detrital zircons are controlled by the time needed to exhume the zircons from their closure depth and that the additional time for transport is commonly negligible.

Our focus in this discussion has been on the average time for transport of zircon through the drainage system. Clearly, there will be variance around the average. Thus, residence time might be short, but variance could be very large. At

present, it is not possible to provide any direct assessment of this question. We note, however, that a large variance would tend to smooth out the probability density distributions (i.e., diffusive smoothing). In other words, individual peaks would tend to broaden downstream. Our present data show no evidence of peak broadening, so variations in residence time does not seem to be a significant issue for FT studies of detrital zircons.

CONCLUSIONS

This study of the Rhône and Rhine drainages shows that detrital zircon FT ages from river and delta sediments do give a reliable and persistent record of the cooling history of the source region from which they were eroded. The fluvial system helps average out short-term variations in sediment transport. As a result, the zircon FTGA distribution provides a long-term average of the yield of zircons from the erosional source region. Furthermore, storage within the drainage does not appear to have a significant influence on the FTGA distributions. The reason is that average residence time for sediment moving through the drainage appears to be very short (< 1 Myr) relative to the uncertainties in the FT ages. The short residence time is a consequence of the fact that sediment discharge is large compared to the volume of actively moving sediments within the drainage. Thus, the lag time between FT closure and deposition provides a good estimate of the time needed to exhume the zircons from their FT closure depth. This result is important because it supports the use of FT lag times as an estimate of exhumation rates in studies of ancient orogenic settings (Garver et al. 1999; Bernet et al. 2001).

A final conclusion is that the Alps have a large influence on the composition of the zircon FTGA distribution in the Rhine and the Rhône, even after 500 to 1000 km

of transport. This result reflects the generally observed fact that the sediment yield in continental drainages is commonly dominated by mountainous parts of the drainage where uplift and erosion are fastest. Thus, we can be confident that the cooling information by FTGA distributions from old orogenic sediments mainly reflects the thermal and exhumational history of orogenic areas within the drainage.

ACKNOWLEDGMENTS

This study was supported by student grants from the Geological Society of America (Bernet) and a summer research fellowship from Yale University (Bernet); National Science Foundation grants (Garver: EAR-9614730; Brandon: OPP-9911925), and the Union College Faculty Research Fund (Garver). Neutron irradiation was subsidized by the Reactor Use Sharing Program (U.S. Department of Energy), granted to the Oregon State University Nuclear Reactor. Uwe Baaske is acknowledged for his help with sample collection and a great field season. We thank Paul O'Sullivan, Zell Peterman, and Mark Johnsson for critical and constructive reviews, which helped us to improve the manuscript. The detrital zircon fission-track data presented in this paper have been archived and all single grain ages are available in digital form at the World Data Center-A for Marine Geology and Geophysics, NOAA/NGDC, 325 Broadway, Boulder, Co, 80303; (phone: 303-497-6339; fax: 303-497-6513; E-mail: wdcamgg@ngdc.noaa.gov, or use the following web address URL:<http://www.ngdc.noaa.gov/mgg/sepm/archive/index.html>

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FIGURE CAPTIONS

Fig. 1 Simplified map of the Rhône River and Rhine River drainages in central Europe. Labels for lakes and geologic features are identified in the legend. Labels for sample locations are: Rhône drainage: A = middle Rhône River, B = Isère River, C = Drôme River, D = Ardèche River, E = Durance River, and three from the Rhône delta: F = Fos sur Mer, G = St Marie de la Mer, H = Plage de Piemonsan. Rhine drainage and associated areas: I = Inn River (Danube drainage, but included here for comparison), J = Buchs on the Rhine River, K = Reuss River, L = Aare River, M = Worms on the Rhine River, and N = Rotterdam on the Rhine delta.

Fig. 2 Contour maps of zircon FT cooling ages in the: A) Rhône River and B) Rhine River drainage areas. The maps are based on a compilation of bedrock zircon FT ages (Bernet et al. 2001). Note that contour intervals are different for each map and that data coverage in the lowlands is much less dense than that for the Alpine highlands.

Fig. 3 Width of Holocene deposits along the length of the: A) Rhône and B) Rhine rivers. These data were derived from geologic maps. The average width of Holocene deposits adjacent to the river channel was determined from 1:1,000,000 scale geologic maps (Geologische Karte der Bundesrepublik Deutschland 1993; Carte Géologique de la France 1996). Holocene deposits in Lake Geneva and Lake Constance are treated separately, and thus are not included here.

Fig. 4 Probability density plots (Brandon 1996) of FTGA distributions and best-fitted peaks (Galbraith and Green 1990) for the Rhône drainage. See samples A through H in Table 1 and Fig. 1 for further details.

Fig. 5 Probability density plots and best-fitted peaks of A) the Alpine part of the Rhine River and other Alpine tributaries, and B) downstream Rhine River samples. See samples I through N in Table 2 and Fig. 1 for further details.

Fig. 6 Probability density plots of FTGA distributions for three samples from the Rhône delta, used to show reproducibility of the distributions. See samples F, G, and H in Table 1 and Fig. 1 for further details.

Fig. 7 Cumulative probability plots for FTGA distributions for the three Rhône delta samples. Samples G and H have similar distributions, whereas sample F is significantly different. The statistical significance of these differences was tested using the KS statistic (Press et al. 1992), which is defined in terms of the maximum difference in cumulative probability between two distributions. The KS statistic for F vs. H is 29%, for F vs. G, 30%, and for G vs. H, 11%. The probability of observing these values for random resampling is 0.2%, 0.3%, and 74%, respectively.

Fig. 8 Summary of best-fit results for mixture model for the composite Rhône delta FTGA density distribution. The observed Rhône delta sample (black line) is represented by samples F, G, and H combined. The predicted density plot is based on results shown in Table 3, where upstream samples A, B, C, and E, are combined as components to give a best-fit mixture for the downstream delta samples.

Fig. 9 Summary of best-fit results for mixture models for the Rhine drainage. The downstream samples M from Worms and N from the Rhine delta at Rotterdam are modeled as mixtures with the upstream samples J, K, and L representing the components of the mixtures. The black lines show the observed density plots for samples M and N, and the gray lines show the predicted density plots given by the mixture calculation, as reported in Table 4. Note that we have allowed the solution to account for the fact that we have no proxy for sources external to the Alps. The difference between the observed and predicted density plots represents the estimate of the contribution due to those external sources.

Fig. 10 A weighted regression showing the FT age for the P2 peak from the Rhine drainage as a function of distance from the mouth of the Rhine. The regression shows a slight increase in age downstream, equal to 0.8 Myr over 1000 km. However, the one standard error uncertainty for that estimate, 1.2 Myr, means that the downstream increase is not significant at the 68% level.

Table 1. Best-fit age components for zircon FTGA samples from the Rhône drainage

Location* (upstream distance)	No. of Grains	Range (Ma)	P1	P2	P3	P4	P5	P6
<i>Upper Rhône and tributaries (in order moving downstream)</i>								
A) middle of Rhône River (340 km)	100	8 – 138	8.8 ± 1.9 6%	16.2 ± 1.5 52%	30.0 ± 3.7 20%	--	93.6 ± 11 22%	--
B) Isère River (230 km)	100	8 – 146	--	14.1 ± 2.0 29%	35.5 ± 4.5 38%	--	87.7 ± 9.2 33%	--
D) Ardèche R. (135 km)	100	14 – 301	--	--	23.9 ± 3.6 9%	--	103 ± 11 74%	181 ± 37 17%
E) Durance R. (105 km)	100	17 – 218	--	--	30.3 ± 4.2 12%	70.4 ± 13.2 38%	107 ± 16 50%	--
<i>Rhône delta</i>								
F) Fos sur Mer (~0 km)	71	6 - 434	--	14.2 ± 1.9 18%	29.3 ± 4.0 17%	66.1 ± 8.5 25.9%	--	163 ± 20 39%
G) St Marie de la Mer (~0 km)	77	7 – 293	--	12.1 ± 2.0 29%	21.4 ± 3.5 29%	64.6 ± 8.1 21%	--	158 ± 22 21%
H) Plage de Piemonsan (~0 km)	80	5 – 301	--	10.1 ± 1.3 28%	23.1 ± 3.0 25%	54.0 ± 7.5 24%	112 ± 15 23%	--
F-H) Delta composite	228	5 – 434	--	11.7 ± 1.3 26%	24.1 ± 2.7 24%	64.5 ± 6.8 26%	--	154 ± 16 24%

* Sample C from the Drôme River contained no zircon.

Note: Best-fit components are indicated by their mean age ± 2 SE, followed by their estimated relative size (%) in the FTGA distribution. All samples were counted at 1250x dry (100x objective, 1.25 tube factor, 10x oculars).

Rhône delta samples were counted by B. Molitor (zeta (CN-5) of 359.9 ± 12.71 (± 1 SE). The remaining samples were counted by M. Bernet using a zeta (CN-5) of 334.22 ± 3.40 (± 1 SE).

Table 2. Best-fit age components for zircon FTGA samples from the Rhine and related drainages

Location (upstream distance)	No. of Grains	Range (Ma)	P1	P2	P3	P4	P5	P6
<i>Inn drainage (representative of Alpine sources in the upper part of Rhine drainage)</i>								
I) Inn River (n.a.)	60	16 – 252	--	17.9 ± 3.6 15%	27.5 ± 8.4 6%	49.2 ± 4.5 70%	103 ± 24 9%	--
<i>Alpine part of Rhine drainage</i>								
J) Buchs, Rhine River (915 km)	60	12 – 160	--	16.6 ± 2.5 41%	26.4 ± 3.4 49%	--	102 ± 16 10%	--
K) Reuss River (1005 km)	60	10 – 118	--	17.1 ± 1.3 85%	28.8 ± 8.0 10%	--	98.7 ± 28 5%	--
L) Aare River (970 km)	60	9 – 244	--	16.1 ± 1.5 90%	--	--	123 ± 28 10%	--
<i>Lower part of Rhine drainage, including the Rhine delta</i>								
M) Worms, Rhine River (495 km)	60	9 – 188	--	17.2 ± 1.8 45%	33.1 ± 3.9 23%	--	131 ± 14 32%	--
N) Rotterdam, Rhine delta (20 km)	80	10 – 407	--	17.4 ± 2.5 17%	--	--	90.2 ± 18 26%	155 ± 23 57%

Note: See Table 1.

Table 3. Best-fit mixture for zircons in Rhône delta sediments

Component of Mixture	Proportion (%) ± 1 SE
A) middle reach, Rhône River	40 ± 5
B) Isère River	37 ± 6
D) Ardèche River	23 ± 5
E) Durance River	0 ± 6

Table 4. Best-fit mixture for zircons in samples from the lower Rhine River

Component of Mixture	Proportion (%) \pm 1 SE	
	M) Worms, Rhine River	N) Rotterdam, Rhine delta
J) Buchs, Rhine R.	50 \pm 5	10 \pm 5
K) Reuss River	0 \pm 7	0 \pm 7
L) Aare River	21 \pm 7	9 \pm 7
External sources	29 \pm 11	81 \pm 11

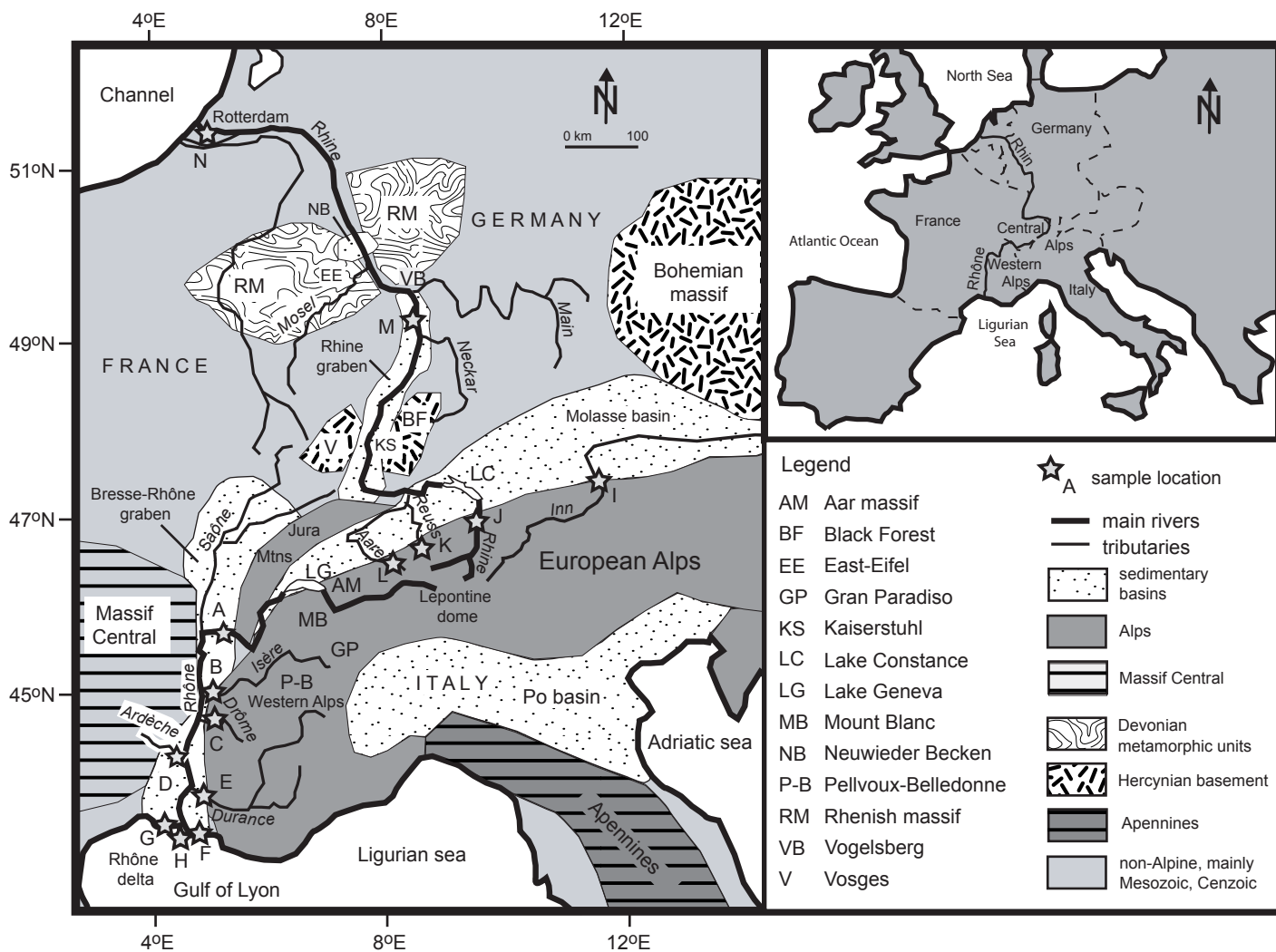


Fig. 1 Bernet et al

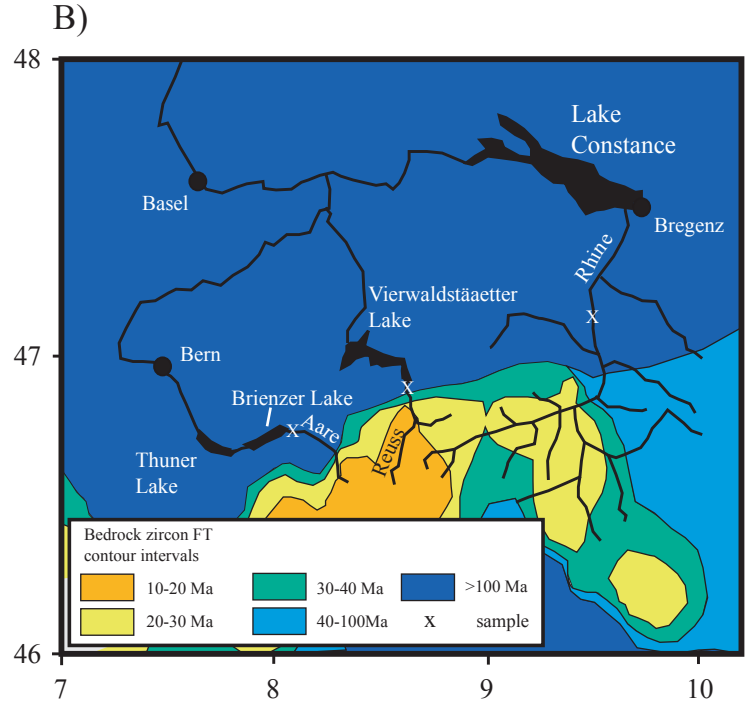
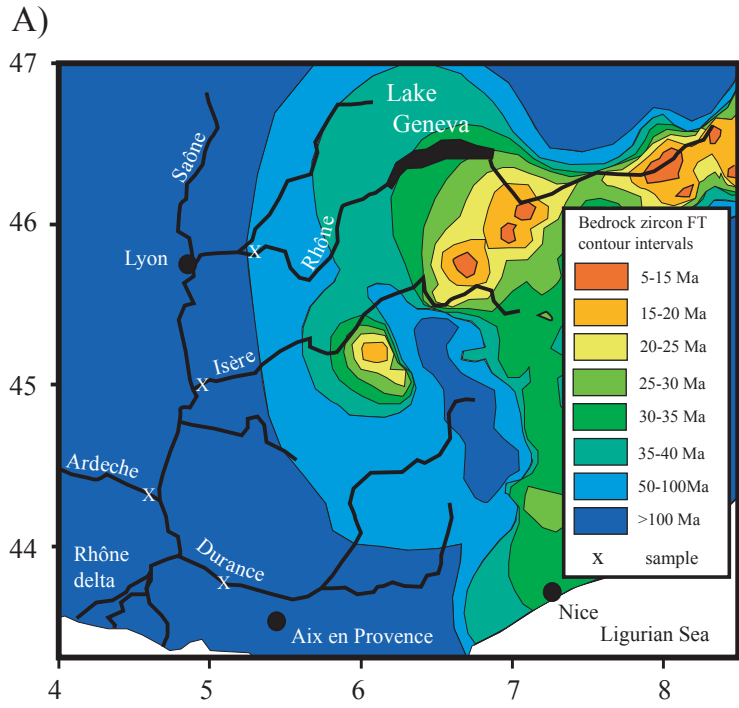
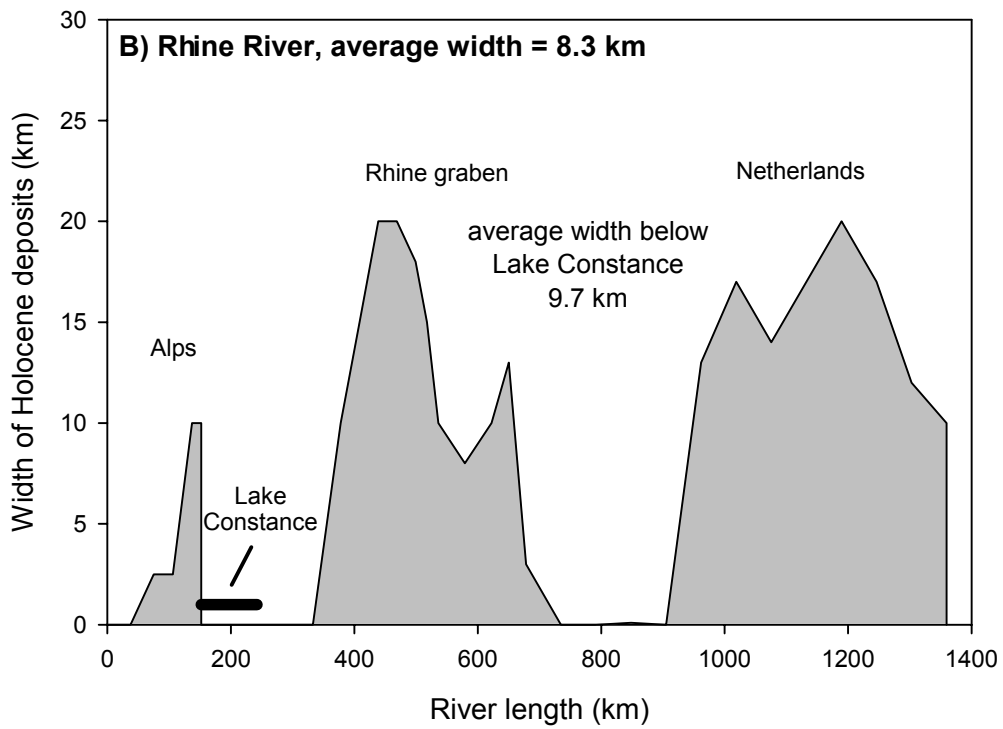
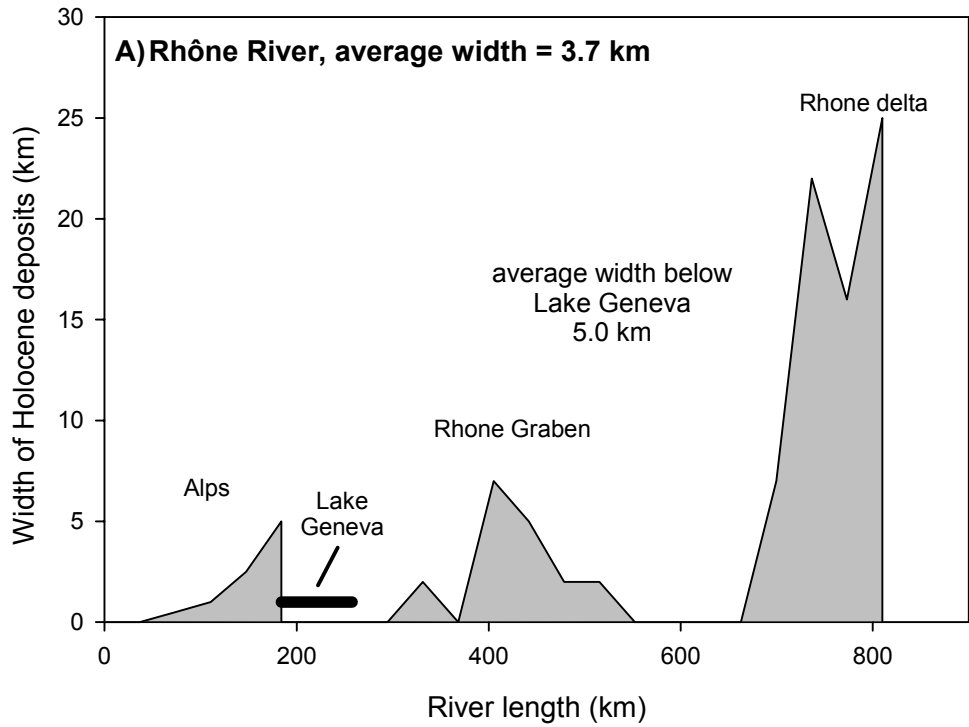


Fig. 2 Bernet et al.



Bernet et al. Fig 3

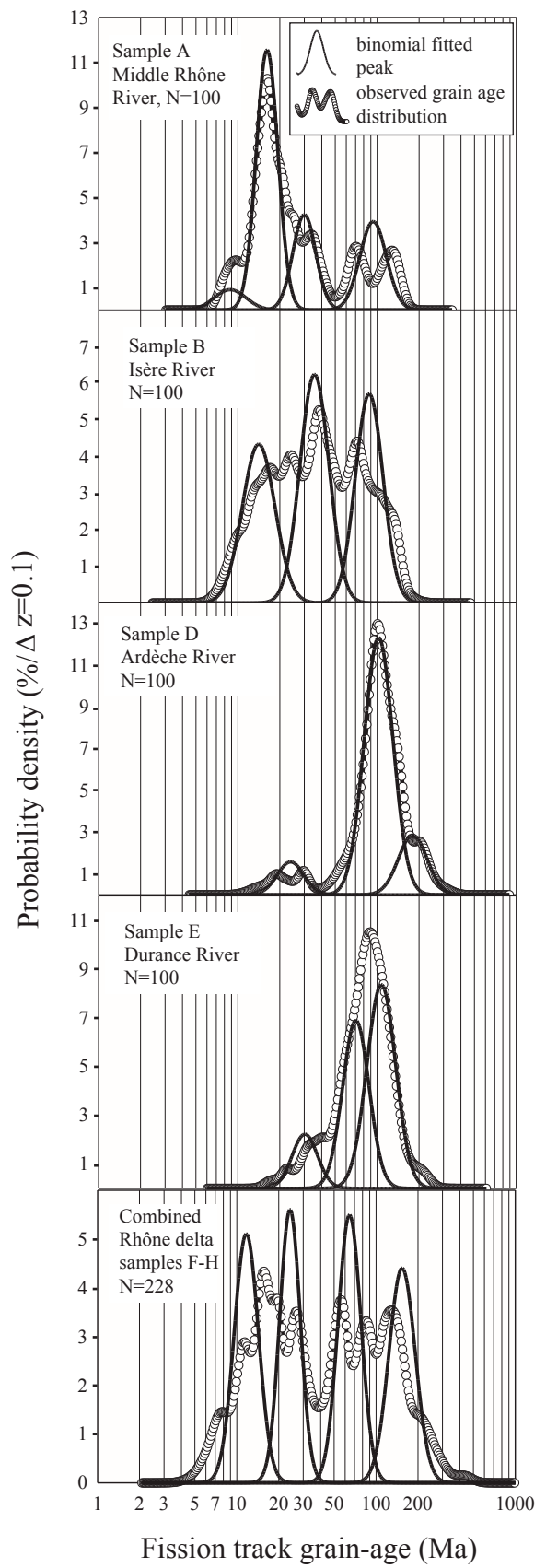


Fig.4 Bernet et al.

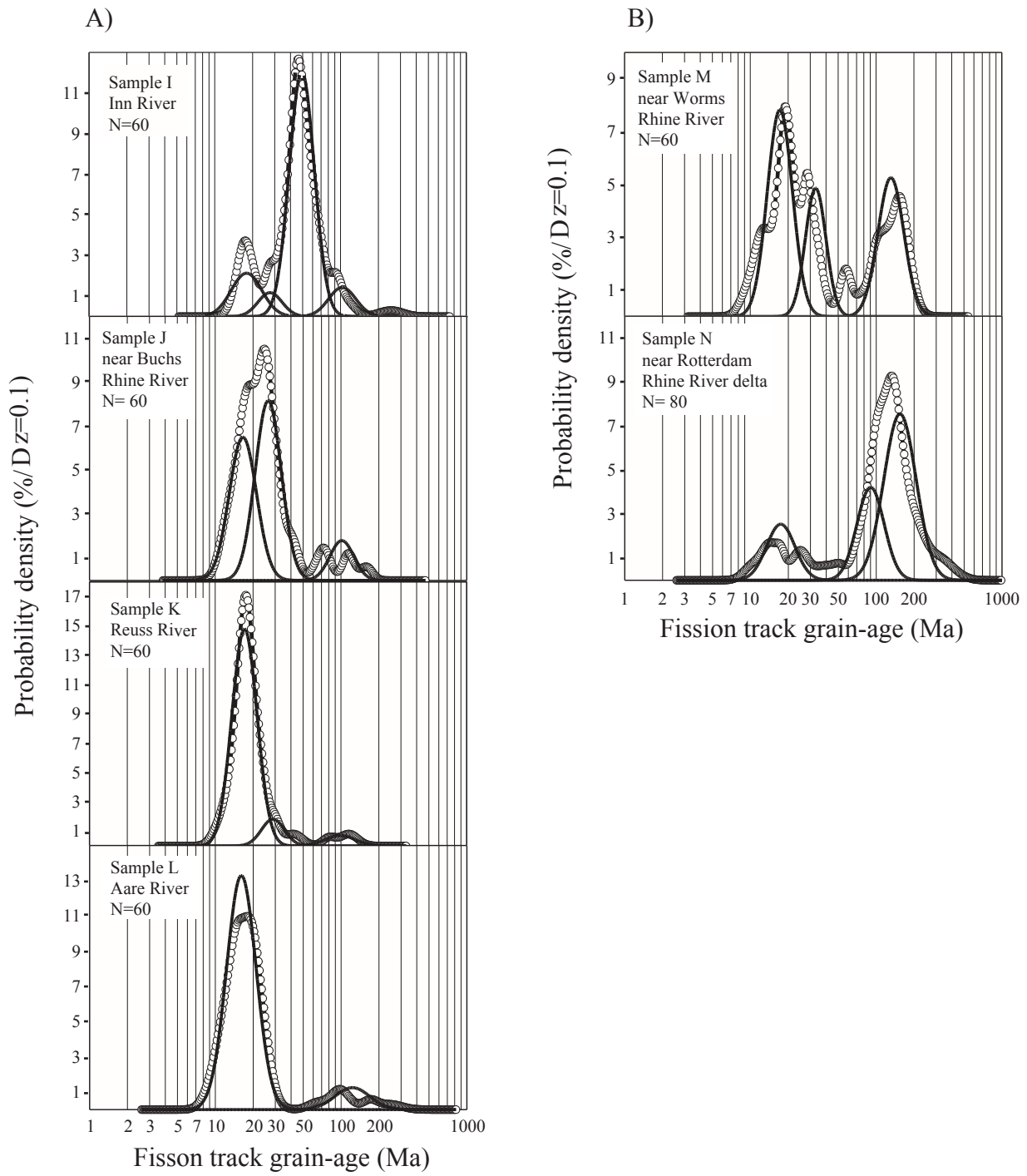


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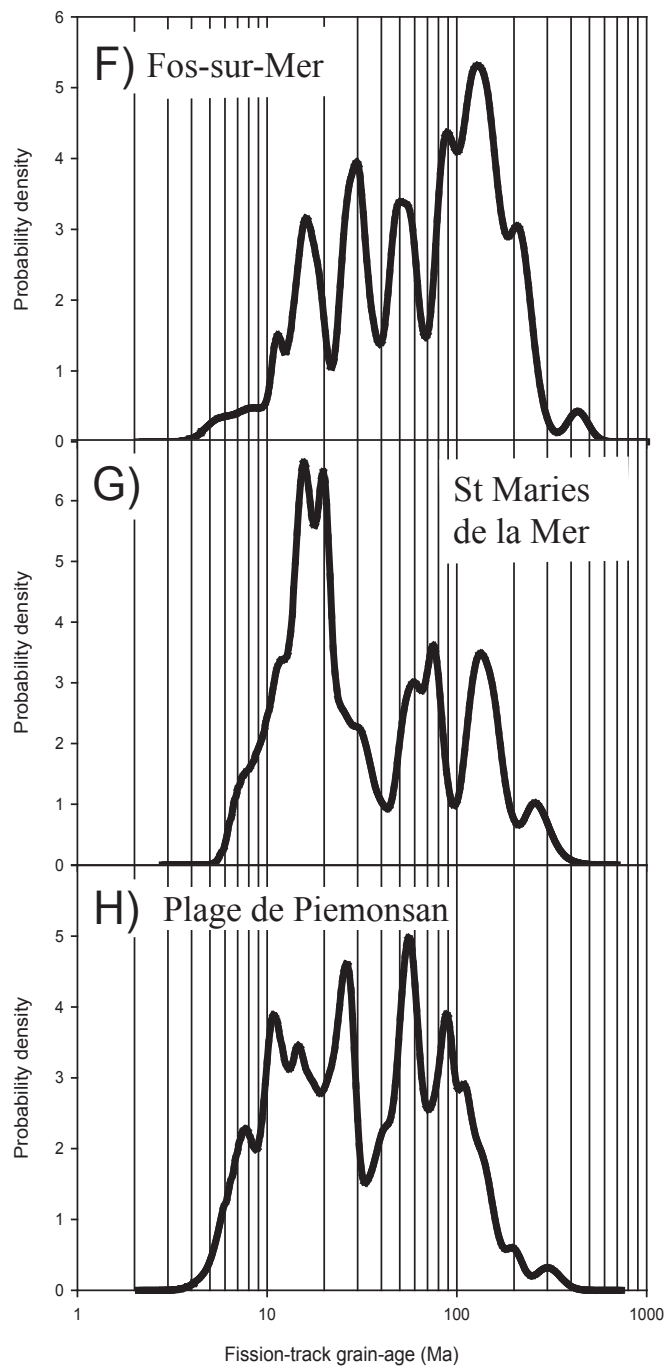


Fig. 6 Bernet et al.

Rhône delta

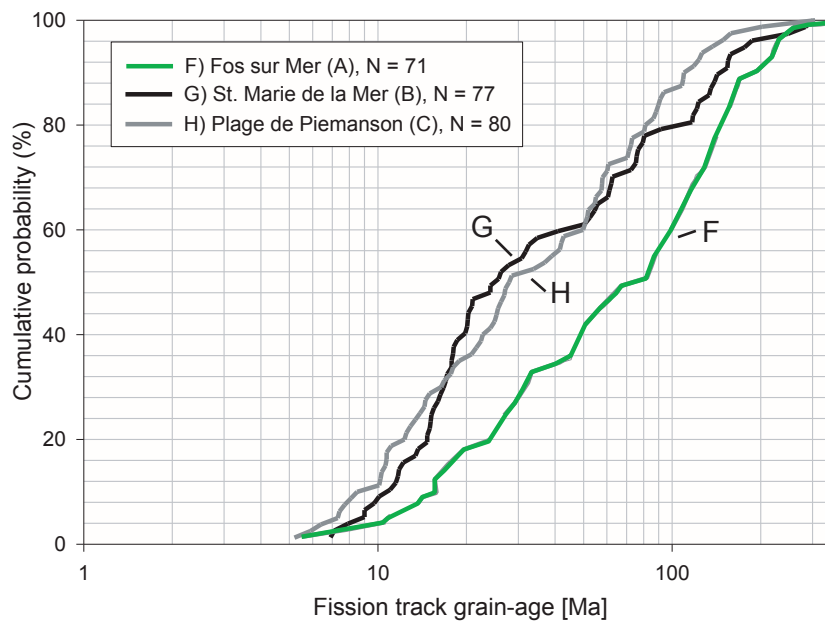


Fig. 7
Bernet et al.

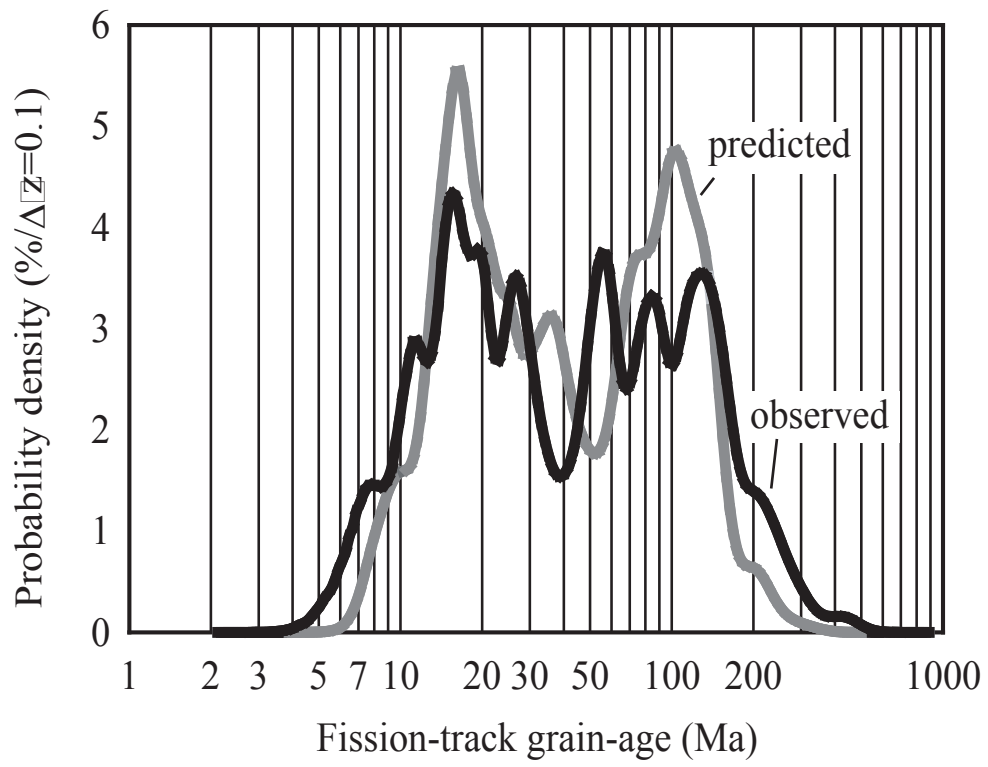
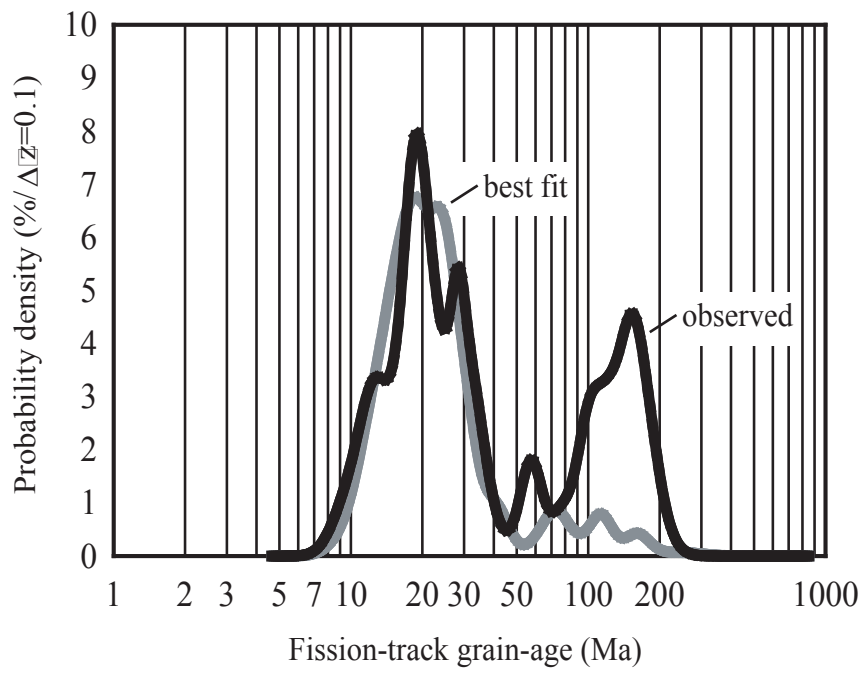


Fig. 8 Bernet et al.

Mixture Model for M) Rhine at Worms



Mixture Model for N) Rhine Delta at Rotterdam

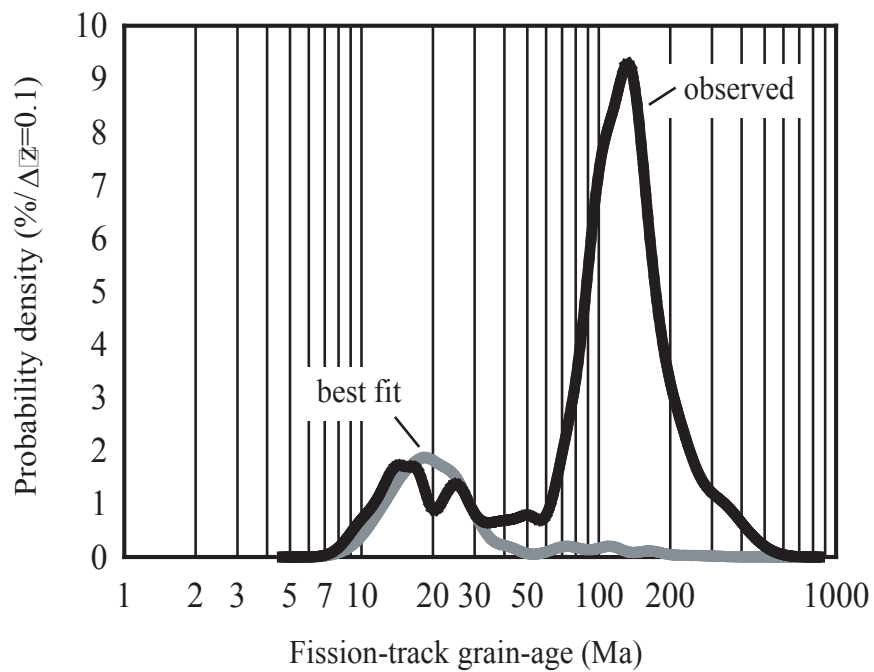


Fig. 9 Bernet et al.

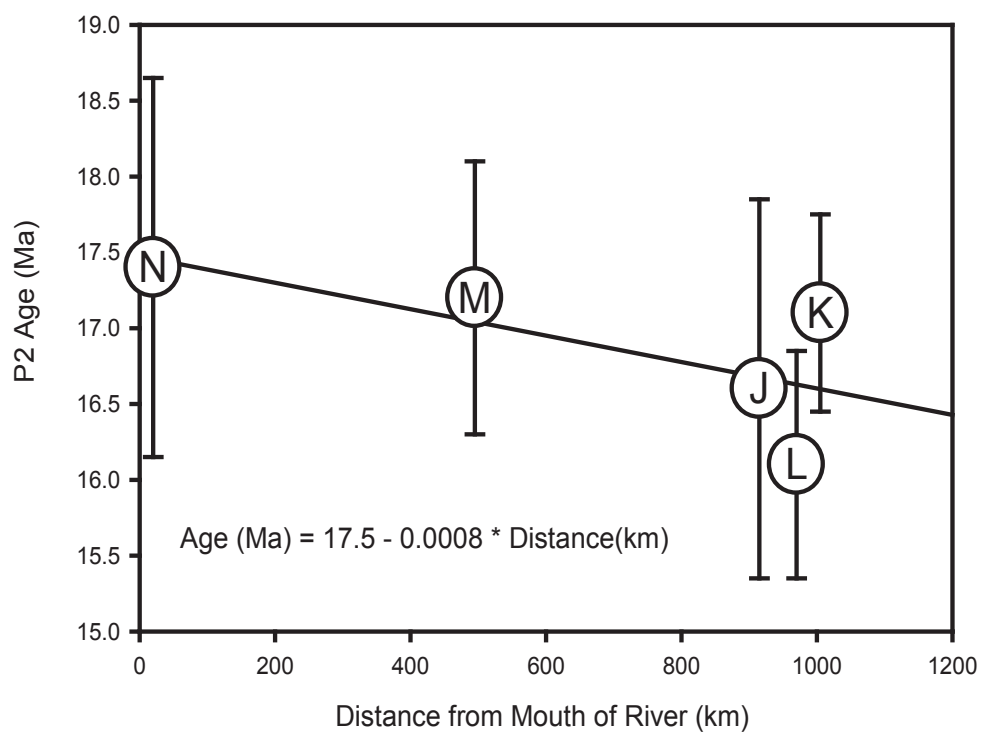


Fig. 10 Bernet et al.