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New geochemical results indicate a non-alpine provenance for the Alpine Spectrum (epidote, garnet, hornblende) in quaternary Upper Rhine sediment

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ABSTRACT

The heavy mineral assemblage of late Pleistocene to Modern Rhine river sediment in the Upper Rhine Graben is dominated by garnet, green amphibole and epidote. This so-called Alpine Spectrum has been taken to indicate an exclusive derivation from the Alps and has hitherto been investigated with optical methods only. We present the first single-grain geochemical data set of garnet and amphibole from the upper Rhine river and some of its main tributaries. We use the new data to test the alleged Alpine provenance in the Rhine.

Our results show that the provenance of garnet, mostly pyrope-rich almandine is grain-size dependent. Particularly the pyrope content indicates that the 0.063–0.25 mm size fraction has an Alpine provenance whereas 0.25–0.5 mm grains derived from the Black Forest and the Vosges, which flank the Upper Rhine Graben. They mainly comprise of Paleozoic metamorphic and igneous rocks supplying a suite of heavy minerals similar to the Alpine Spectrum to the Rhine river. Amphibole, mostly Mg-hornblende, has an increasing TiO_2 content from the south to the north of the graben, which also indicates an input from the graben shoulders. The new data indicate that the so-called Alpine Spectrum of heavy minerals in Rhine river sediment of the upper Rhine derived to significant degrees from non-Alpine sources. Furthermore, our results indicate a relatively uniform provenance for the Rhine River system during the past 1 Ma.

1. Introduction

The temporal and spatial evolution of the sedimentary fill of peri-orogenic basins reflects the exhumation history of mountain belts. The volume and composition of sediment exported from an orogen is the product of the interplay of tectonic activity and climate (Gulick et al., 2015). The sediment offers insights into the source-rock assemblages and may allow for establishing a sediment transfer budget. Detrital single grain analysis provides the geochemical composition and conditions of formation of the source rocks and can therefore help to trace changes in provenance and changes in the relative influence of different sources. In the best case, this can help to quantify the amount of material that was eroded from certain sources (Weltje and von Eynatten, 2004; von Eynatten and Dunkl, 2012).

The Upper Rhine Graben is located in Central Europe along the border between Germany, France, and Switzerland (Fig. 1a) and is a sink for detritus from the Alps. It is part of the European Cenozoic Rift System that runs from the Mediterranean Sea in the south to the North Sea in the north (Ziegler and Dezes, 2007). Since its opening during the late Eocene the graben was repeatedly connected to the northern Alpine drainage system (Ziegler and Fraefel, 2009). The last reorganisation of the northwestern alpine drainage network at around 2.9 Ma established today's conditions and led to a significant change in the Upper Rhine Graben sand-sized sediments, in the heavy mineral composition (Hagedorn, 2004; Hagedorn and Boenigk, 2008) as well as in thermochronological data (Tatzel et al., 2015). The heavy mineral assemblage of sediment older than 2.9 Ma is dominated by stable minerals like zircon, tourmaline and rutile, whereas in younger strata garnet, green amphibole and epidote dominate. This association is called the Alpine Spectrum (van Andel, 1950). Zircon fission-track data show a similar shift from Permo-Mesozoic ages in sediment older than 2.9 Ma to an age spectrum dominated by Cenozoic cooling ages in younger sediment (Tatzel et al., 2015). However, the age distribution of detrital zircon fission-track ages is changing in the course of the Rhine along the Upper Rhine Graben. Bernet et al. (2004) showed that the alpine rivers and the alpine Rhine contain relatively young zircon grains between 10 and 30 Ma with a unimodal age distribution. At the northern margin of

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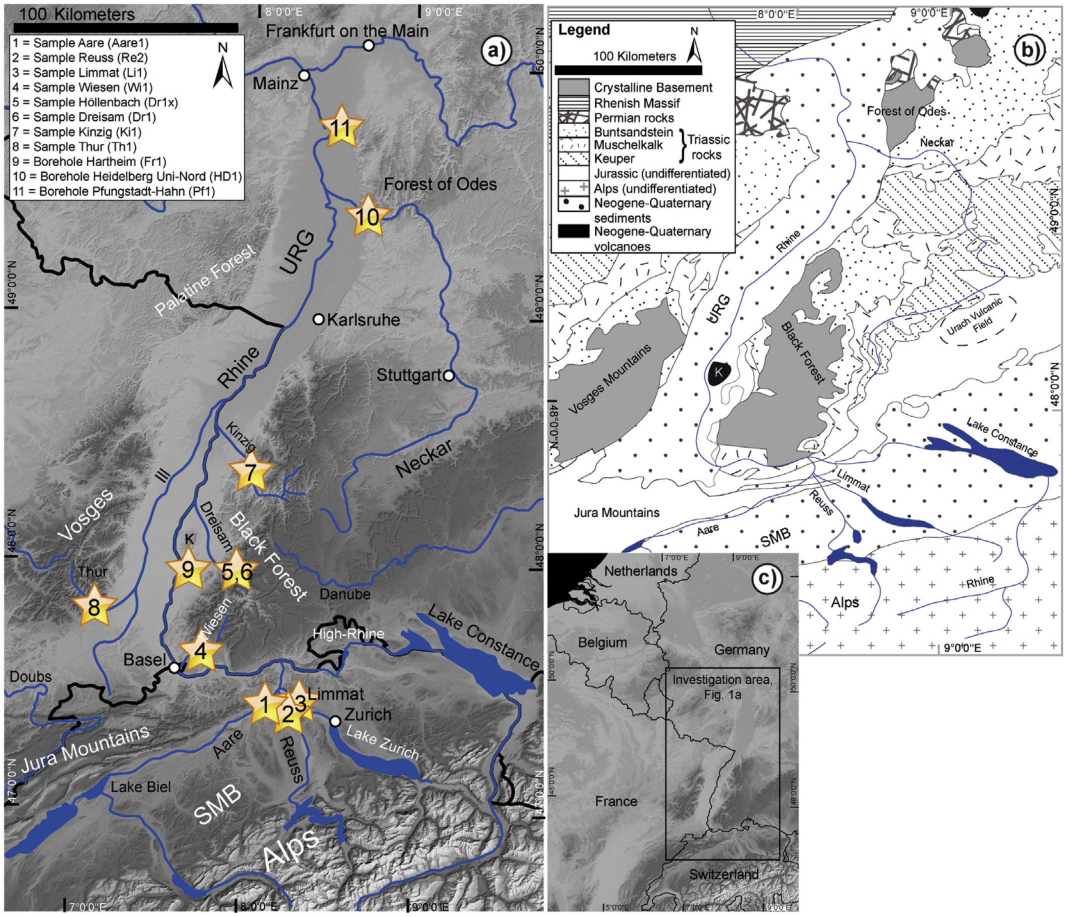

Fig. 1. a) Geographical map with sample locations, thick black lines = state border, blue lines = rivers (DEM baselayer provided by European Environment Agency), b) geological map of the investigation area, the crystalline basement mainly consists of gneiss and granitoid, (c) overview map of central Europe with the political boarders, SMB = Swiss Molasse Basin, URG = Upper Rhine Graben, K = Kaiserstuhl. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the graben, the ages are bimodal with main populations at 10–30 Ma and 100–250 Ma. The older ages are in accordance with zircon fission-track ages from the Black Forest and the Vosges Mountains (Fig. 1a; Timar-Geng et al., 2006).

The metamorphic flanks of the Upper Rhine Graben, the Black Forest and the Vosges Mountains, deliver a similar heavy-mineral assemblage as the Alps (van Andel, 1950; Hagedorn, 2004). In addition, a significant change took place in U-Pb age distributions of detrital zircon from the southern to the northern edge of the graben from a dominance of Caledonian ages (490–390 Ma) in the south to Variscan ages (330–300 Ma) in the north (Krippner and Bahlburg, 2013). This indicates an increasing transport from the graben shoulders because the Black Forest (Schaltegger, 2000; Hann et al., 2003; Marschall et al., 2003), and the Vosges Mountains (Gayk and Kleinschrodt, 2000; Skrzypek et al., 2012a), as well as the Forest of Odes (Fig. 1a; Altenberger and Besch, 1993; Siebel et al., 2012) are dominated by Variscan-age rocks. Other heavy minerals, including those of the Alpine Spectrum, may have been delivered from the flanks of the graben, too.

The geochemical composition of garnet and amphibole, both part of the alleged Alpine Spectrum, often are used in provenance research (Mange-Rajetzky and Oberhänsli, 1982; Morton, 1985; Schäfer, 1997; Andó et al., 2014; Krippner et al., 2014). The discrimination potential of garnet has been recently shown in modern alpine river sediments (Krippner et al., 2015; Stutenbecker et al., 2017). Discrimination of provenance is less straightforward for amphibole than for garnet though. The TiO_2 content and colour of amphibole are directly connected to the conditions of formation, and the TiO_2 content increases with increasing metamorphic grade whereas the SiO_2 content decreases (Leake, 1965; Andó et al., 2014). An additional advantage of using amphiboles arises from its potential to be $^{40}Ar/^{39}Ar$ dated.

We sampled recent sand-sized sediment of tributaries of the Rhine and middle Pleistocene drill core sand-sized sediment from the Rhine and Neckar rivers (Fig. 1) in order to test the provenance of the Alpine heavy-mineral spectrum. We concentrated on garnet and amphibole, the main constituents of the Alpine Spectrum, and performed single-grain major-element geochemical analysis with an electron microprobe. In addition, $^{40}Ar/^{39}Ar$ dating of selected amphibole grains were undertaken.

2. Geological setting

The Rhine is the largest drainage and river system of central Europe. From its springs in the Central Alps to its mouth in the Netherlands it has a length of 1239 km and drains 185,000 km^2. Lake Constance (Fig. 1) forms a local sediment sink. The tributary with the highest water discharge is the Aare river (Fig. 1), which drains the Swiss Molasse Basin. After the confluence with the Aare the Rhine enters the Upper Rhine Graben close to Basel (Fig. 1a, Preusser, 2008).

The geological and geomorphological evolution of the Upper Rhine Graben has been shaped during the past c. 3 Ma mainly by changing rates of uplift and denudation of the Alps (Schlunegger and Mosar, 2011; Reiter et al., 2013; Tatzel et al., 2015). Two main sediment sinks in the area – the Northern Alpine Forland Basin and the Upper Rhine Graben – store most of detritus derived from the Alps and from other mountain ranges bordering the Upper Rhine Graben, notably the Vosges Mountains, the Black Forest, the Jura Mountains, the Forest of Odes and

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