

Provenance of Upper Devonian clastic (meta)sediments of the Böllstein Odenwald (Mid-German-Crystalline-Zone, Variscides)

Wolfgang Dörr¹ · Gernold Zulauf¹ · Axel Gerdes¹ · Filip Loeckle¹

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Abstract Detrital zircons separated from paragneisses of the northern Böllstein Odenwald (Mid-German Crystalline Zone, Variscides) yielded Cambrian to Upper Devonian U–Pb ages. The age of the youngest detrital zircon population (371 ± 3 Ma) and the Lower Carboniferous metamorphic overprint indicate an Upper Devonian depositional age of the paragneiss protolith. The age spectra of detrital zircons suggest the latter to be derived from (1) Ordovician igneous rocks of the Saxothuringian Zone, (2) a late Cadomian magmatic arc (Teplá-Barrandian Unit) and (3) a Silurian-Devonian magmatic arc. Cadomian igneous activity is also documented by Cambrian zircon cores in Lower to Middle Devonian detrital zircons. Meso- and Paleoproterozoic detrital zircons, which are typical for the Old Red Continent and the Rheohercynian Zone, are entirely lacking. The restricted Palaeozoic detrital zircon age spectrum is attributed to a Silurian-Devonian intra arc or trench setting. Both the lack of Mesoproterozoic detrital zircons and the striking similarity of the U–Pb ages of the detrital zircon obtained from the Böllstein Odenwald with U–Pb ages from crystalline rocks of the Saxothuringian basement, rules out that the Böllstein Odenwald is forming a tectonic window (Rheohercynian lower plate) inside

the Mid-German Crystalline Zone (Saxothuringian upper plate).

Keywords U–Pb ages · Detrital zircons · Devonian age · Böllstein Odenwald · Mid-German Crystalline Zone · Variscides

Introduction

The configuration of lithospheric plates in Central Europe is well understood back to the Pangaea supercontinent during Permian times. The pre-Permian reconstruction of the positions of continents hampers, as evidences, such as active/passive-margin matchups, fossil record, paleomagnetism etc., get fainter (e.g. Bradley 2011). For this reason Paleozoic plate reconstructions in Central Europe are badly constrained. In recent times, U–Pb age clusters of detrital zircon populations, determined using Laser Ablation combined with Inductively-Coupled-Plasma Mass-Spectrometry (LA-ICP-MS), have been used as a robust and fast tool to provide constraints on provenance areas of clastic rocks (Linnemann et al. 2012; Pereira et al. 2012; Fernández-Suárez et al. 2013; Heinrichs et al. 2012; Zulauf et al. 2014; Dörr et al. 2015). Such detrital zircon ages are particularly important for the paleogeographic reconstruction of different continents and/or terranes and help to identify the provenance of tectonic thrust sheets, which are common in collision zones. Geologists have to decide if these thrust sheets were part of the downgoing slab or did they belong to the upper plate?

During the Paleozoic era three main continents, Laurentia and Baltica to the north and Gondwana to the south were separated by large oceans (Stampfli and Borel 2002). Microcontinents or terranes, like Avalonia or Armorica,

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✉ Wolfgang Dörr
w.doerr@em.uni-frankfurt.de
Gernold Zulauf
g.zulauf@em.uni-frankfurt.de

¹ Institut für Geowissenschaften, Goethe-Universität, FE Geologie, Altenhöferallee 1, 60438 Frankfurt am Main, Germany

were split off from Gondwana and drifted to the north colliding with the northern continents. The Avalonian terrane is a Neoproterozoic terrane (O'Brien et al. 1996), which collided with Baltica and Laurentia after the Iapetus ocean was closed during the Late Ordovician. This early subduction/collision, referred to as Caledonian orogeny, resulted in the formation of Laurussia, the Old Red Continent (Torsvik and Rehnström 2003; Winchester et al. 2002, 2006; Torsvik and Cocks 2013; Cocks and Torsvik 2006; Cocks and Fortey 2009; von Raumer et al. 2002, 2003; Murphy et al. 2004b; Nance et al. 2008). The clastic sediments derived from the Old Red Continent (e.g. the Paleozoic sediments of the Rheohercynian Zone) are characterized by a dominance of Mesoproterozoic and Paleoproterozoic detrital zircons with local Ediacaran influence and minor input of Silurian zircons (Haverkamp 1991; Linnemann et al. 2012; Geisler et al. 2005; Zeh and Gerdes 2009; Eckelmann et al. 2013).

In the Late Paleozoic era, the Armorican terrane assemblage (ATE) collided with Laurussia after the closure of the Rheic ocean during the Variscan orogeny (Tait et al. 1997, 2000). The sediments derived from the ATE are characterized by dominant Ediacaran and Paleoproterozoic input with a Mesoproterozoic gap (1–1.6 Ga) in the age spectra of their detrital zircons (Linnemann et al. 2004, 2008; Drost 2008; Drost et al. 2010; Gerdes and Zeh 2006; Zeh and Gerdes 2009; Hajna et al. 2013, 2016).

Variscan subduction and collision led to a complex crustal architecture in central Europe, where the following Variscan zones are distinguished: Moravo-Silesian, Moldanubian/Teplá-Barrandian, Saxothuringian (including the Mid German Crystalline Zone, MGCZ), and Rheohercynian (Kossmat 1927). Some authors correlate different units of the MGCZ and the Rheohercynian Zone with specific Baltic, Armorican and Avalonian terranes (e.g. Geisler et al. 2005; Zeh and Gerdes 2009; Eckelmann et al. 2013).

Oncken (1997) attributed Barrovian-type metamorphic rocks of the MGCZ to the Rheohercynian Zone. In his model of tectonic underplating, rocks of the Rheohercynian downgoing slab were accreted to the base of the overriding Saxothuringian plate. Due to considerable extension of the Saxothuringian upper plate, the accreted Rheohercynian rocks could have been exhumed forming a tectonic window in the Saxothuringian domain (Oncken 1997). Franke and Dulce (2016) presented a similar model and proposed the MGCZ as “source regions of Baltica-derived metamorphosed Devonian clastic sediments”. Further to the north, Zeh and Gerdes (2009) investigated the detrital zircon ages of the Ruhla Crystalline Complex (Fig. 1), which is a part of the MGCZ. They found two contrasting U-Pb age spectra, one with a Peri-Gondwanan and the other with a Laurussian (Baltica) provenance. The striking difference in provenance suggests the Rheic Suture to be

situated inside the MGCZ (Zeh and Gerdes 2009). Based on lithology and metamorphic overprint, Zeh and Will (2010) defined different domains in the MGCZ. Domain III includes the Böllstein Odenwald and contains metasediment-metabasalt-orthogneiss associations, which underwent a Barrovian-type metamorphism and is interpreted as Rheohercynian crust. Taking into account the composition and origin of various amphibolites of the different domains of the MGCZ, Will et al. (2015) argue that Baltica/Avalonia-derived rocks are exposed in a tectonic window in the eastern (Böllstein) Odenwald. Will et al. (2015) further suggest that the Oetzberg fault, which separates the eastern (Böllstein) Odenwald from the western (Bergsträsser) Odenwald, represents the Rheic Suture.

In the present paper, we will present U–Pb zircon ages, obtained from detrital zircons of paragneisses exposed in the Böllstein Odenwald (Fig. 1; MGCZ) and compare them with U–Pb zircon ages derived from the Old Red Continent and the Saxothuringian Zone. The new age data will be used to constrain (1) the deposition age of the Böllstein paragneisses, and (2) the Devonian palaeogeography of the Saxothuringian and Rheohercynian realm. The new data will also be used to test the quality of the tectonic models listed above.

Regional geology

The crystalline Odenwald is the largest outcrop of the MGCZ (Brinkmann 1948), which is commonly assumed to represent the northern extremity of the Variscan Saxothuringian Zone as defined by Kossmat (1927, Fig. 1). The crystalline rocks of the Odenwald can be subdivided into the larger Bergsträsser Odenwald to the W and the smaller Böllstein Odenwald to the E (Fig. 2). Both are separated by the N–S trending Oetzberg Zone (Chatterjee 1960; Altenberger et al. 1990; Krohe 1992; Stein 1996, 2001). A subdivision of the crystalline Odenwald into four subunits, based on their different lithology and metamorphic history, is widely accepted (Fig. 2). These four subunits consist of metamorphic complexes and intrusive bodies, which differ in age and degree of tectonometamorphic overprint. Units I–III are forming the Bergsträsser Odenwald, whereas unit IV is the Böllstein Odenwald. Significant differences in lithology, tectonic structures (Chatterjee 1960; Krohe 1992; Altenberger and Besch 1993; Stein 1996, 2001) and the ages of magmatic and metamorphic evolution (Kreuzer and Harre 1975; Lippolt 1986; Todt et al. 1995; Reichmann et al. 2001) suggest an independent pre- to early Variscan development and later juxtaposition of the four metamorphic subunits.

The WNW-dipping Oetzberg zone with its normal fault characteristics is supposed to have been formed during

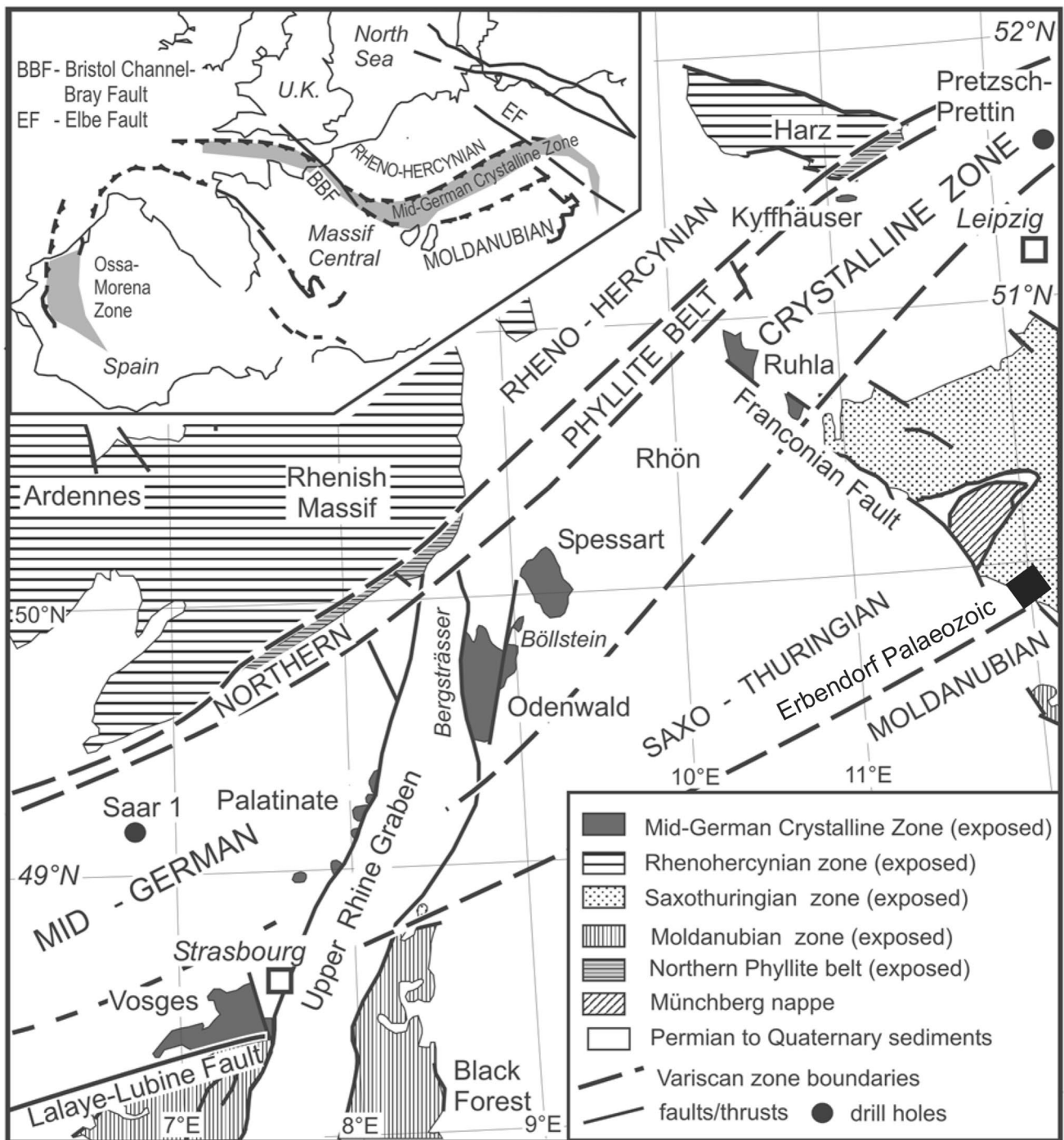


Fig. 1 Map showing distribution of main geotectonic units of Central Europe and the location of the Mid-German Crystalline Zone (after Zeh and Will 2010)

rapid exhumation of the Böllstein massif relative to the Bergsträsser units to the W (Willner et al. 1991; Krohe 1992). A minimum age for the shearing along the Otzberg fault is given by cross-cutting lamprophyric dykes, biotites of which yielded an ^{39}Ar – ^{40}Ar age of 327 ± 3 Ma (Hess and Schmidt 1989). This biotite cooling age is in line

with the K–Ar dating of hornblende from gabbro amphibolite (328 ± 15 Ma, locality Weichberg) and of muscovite from different pegmatites displaying ages from 325 ± 8 Ma (locality Steinkopf) and 325 ± 10 Ma (locality Wannberg; Lippolt 1986). Slightly younger K–Ar ages have been obtained from biotite of crystalline basement, which

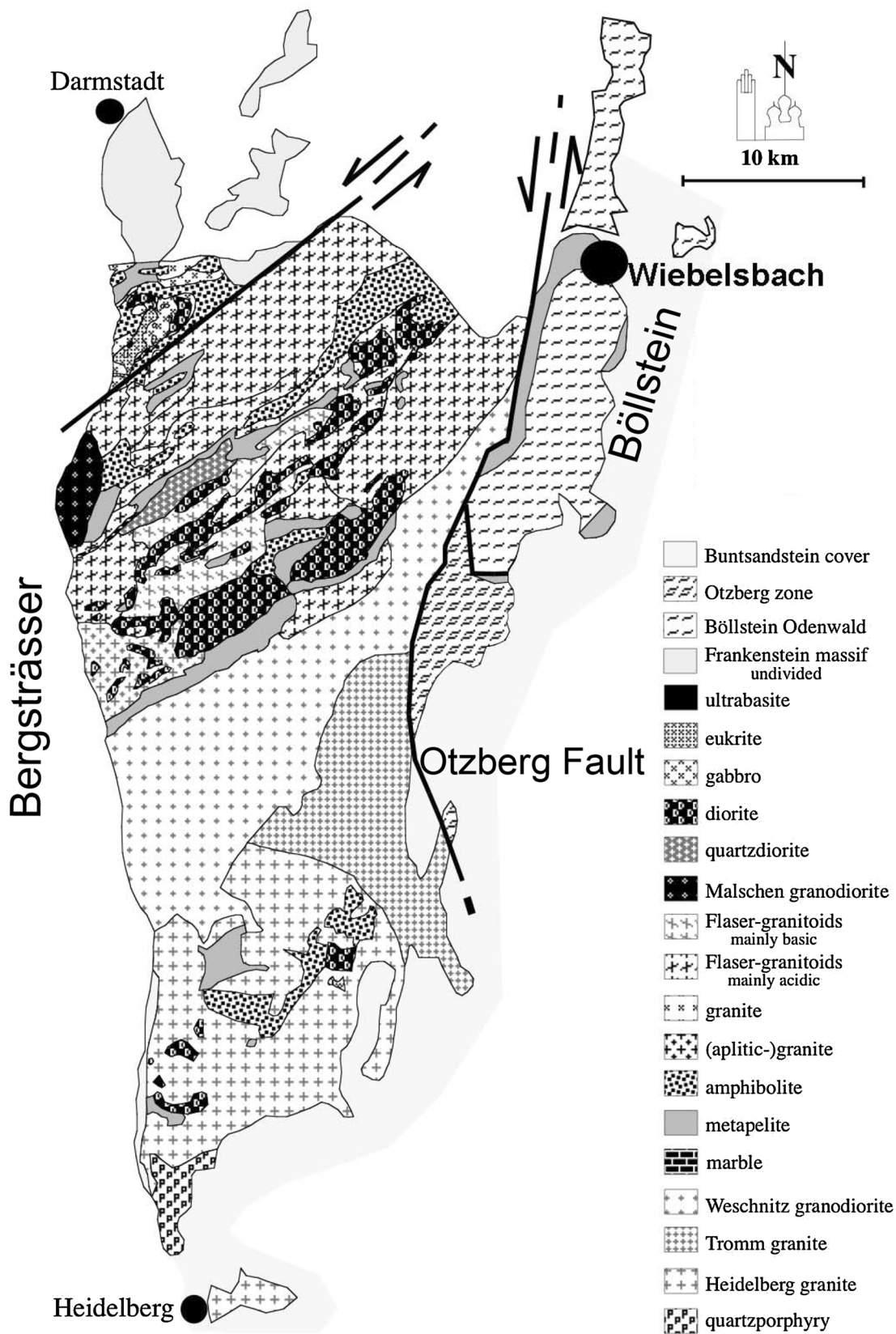


Fig. 2 Geological map of the Odenwald Crystalline Complex (after Stein 2001). Sample locality Wiebelsbach drilling is shown

yielded ages at 318 ± 8 and 319 ± 9 Ma (Lippolt 1986). Kreuzer and Harre (1975) and Lippolt (1986) interpreted the contemporaneous K–Ar mineral ages of gneiss, gabbro amphibolite, lamprophyre and pegmatite between 320 and 330 Ma as the time of regional cooling of the Böllstein massif below 500 °C (hornblende) or 300 °C (micas) after metamorphism, mylonitisation and exhumation.

Early workers attributed the antiform of the Böllstein Odenwald to a gneiss core surrounded by a concordant metasedimentary mantle and both penetrated by mafic to ultramafic intrusives (e.g. Ramdohr 1922; Korn 1929; von Bubnoff 1926). Recent workers do not support a dome-like structure of the Böllstein rocks, but favour a NNE striking anticline plunging slightly towards the NNE (Altenberger and Besch 1993; Stein et al. 2001). The metamorphic mantle, also termed schist envelope, consists of metasedimentary rocks developed mainly as schist, migmatic mylonitic gneiss, quartzite and intercalated calc-silicates (Chatterjee 1960; Altenberger et al. 1990; Altenberger and Besch 1993). The core comprises two types of mylonitic orthogneiss: (1) a largely coarse grained granodiorite-gneiss, consisting of plagioclase, quartz and biotite, and (2) a finer grained granite-gneiss, which contains more alkali-feldspar and muscovite and less biotite and plagioclase (Chatterjee 1960). Folds are more common in granodiorite-gneiss, but are lacking in granite-gneiss, suggesting a more complex deformation history for the former (Chatterjee 1960). The D2 fold axes of the core dip to the NE (δ ca. 30°) on the E flank and to the NW (δ ca. 330°) on the W flank and show large regional variations (Altenberger and Besch 1993). Both orthogneisses show evidence for migmatization (Chatterjee 1960; Altenberger and Besch 1993). Based on field relations, e.g. granitoid apophyses in metasediments in a quarry at the central Weichberg, it has been suggested that the felsic to intermediate orthogneiss protoliths intruded into the pre-existing schist envelope (Altenberger and Besch 1993).

Mafic rocks are present in the schist envelope as well as in the orthogneiss core. They can be grouped into metabasites of high-alumina basalt composition and metaultramafics. The metabasites include gabbros forming stock-like bodies, and diabbases occurring as sills. The metaultramafics include garnet-bearing spinel-chlorite-hornblende-felses and talc- and spinel-bearing chlorite-hornblende-felses, which are similar to the metaultramafics of the Spessart crystalline complex and the Bergsträsser Odenwald (Knauer et al. 1974).

Some of the mafic rocks show relic magmatic mineral assemblages and fabrics, additionally geochemical whole-rock analyses attest to an igneous origin (Knauer et al. 1974). HFSE and REE contents of the mafic metamorphic rocks reflect a primitive tholeiitic composition. The REE-distribution of amphibolites and metabasites shows

an affinity towards island-arc tholeiites (Altenberger et al. 1990). In comparison, the mafic parts from the orthogneiss core and the schist envelope show different enrichment factors, but the patterns of element distribution are alike, thus a common origin in a subduction-type setting is assumed (Altenberger et al. 1990). The eclogite-facies metabasic rocks of the orthogneiss core, exposed south of Wiebelsbach (Böllstein Odenwald), show peak temperatures of 700 ± 50 °C at minimum pressures of 16–17 kbar (Will and Schmädicke 2001). Scherer et al. (2002) determined Lu–Hf Grt–WR minimum ages from two samples at 357 ± 7 and at 353 ± 11 Ma with an initial ϵ_{Hf} of +11.3. This is indicative of long term depletion in their mantle source. Lenses of former eclogites are retrograded into garnet amphibolites during the prograde Carboniferous amphibolite facies metamorphism (8 kbar/580 °C; Will and Schmädicke 2001) together with their country rocks, the orthogneisses. The eclogite facies event must be older than Upper Devonian.

Geochemical investigations of the metasediments from the schist envelope hint to greywackes and subordinate arkoses as protoliths. Arkosic sediments point to a continental magmatic arc as possible source for the detritus (Altenberger and Besch 1993). The metasedimentary schist envelope represents the oldest unit of the Böllstein massif. The age of the metamorphism of the mantle sequence in the Böllstein Odenwald has been dated using U–Pb analyses of zircons, which define a lower intercept at 375 ± 2 Ma (Beerfurth paragneiss; Todt et al. 1995). Although these analyses have been obtained from zircon fractions, the spread of the data and the well defined lower intercept by two nearly concordant fractions, both smaller than 30 μm , point to robust data. This age is confirmed by the upper intercept age of zircons at 375 ± 5 Ma obtained from orthogneisses, which yielded a $^{207}\text{Pb}/^{206}\text{Pb}$ age of 379 Ma. This age marks the intrusion of the granitoids into the Beerfurth paragneiss of the mantle sequence shortly before the amphibolite facies metamorphism (Todt et al. 1995).

The orthogneisses of the central core are derived from granitic and granodioritic protoliths. Geochemical analyses revealed an S-type character, which point to a convergent setting, presumably a continent-island arc collisional environment along an active continental margin (Altenberger et al. 1990; Reischmann et al. 2001). Several inherited zircon ages of 882 and 1138 Ma and Nd model ages of 1.3–1.7 Ga suggest an involvement of Proterozoic material during melt generation (Reischmann et al. 2001). Single zircon Pb/Pb evaporation dating yielded an age of 405 ± 3 Ma for the granodiorite-gneiss. Using the ID-TIMS U–Pb technique on the same sample, a discordia line with an upper intercept at 410 ± 11 Ma and a lower intercept at the origin proves recent lead loss (Reischmann et al. 2001). The analyses with the highest concordance of 97% plot at 386 ± 4 Ma. The sequence of intrusions cannot be

determined solely by rock dating, but field evidence suggests that the protolith of the granodiorite-gneiss intruded into the schist envelope earlier than the protolith of the granite-gneiss was emplaced between the two units (Chatterjee 1960).

Based on the lithological, geochemical and structural differences between the Böllstein and Bergsträsser crystalline rocks, both domains are assumed to have experienced an independent magmatic evolution before being juxtaposed by late-Variscan crustal movement (e.g. Krohe 1992; Reischmann et al. 2001). However, a close relation of the Böllsteiner Odenwald and the Spessart to the N (Fig. 1) has long been suspected based on petrographic (e.g. Korn 1929; Chatterjee 1960) and geochemical similarities in both areas (Okrusch and Richter 1986; Altenberger et al. 1990; Dombrowski et al. 1995; Will et al. 2015). In the central part of the Spessart antiform two types of orthogneiss (Red gneiss and Haibacher Gneiss) intruded at $418\text{--}410 \pm 18$ Ma (Dombrowski et al. 1995; Weber 1995; Okrusch et al. 2011). Because of the similar intrusion age, these gneisses have been correlated with the similar intrusion ages of the granitoids of the Böllstein Odenwald (Lippolt 1986; Anthes and Reischmann 2001; Reischmann et al. 2001). The poorly exposed SW–NE striking Michelbach fault, which is thought to separate the northernmost Spessart basement from the central and southern parts, is suspected to represent the northern prolongation of the Oetzberg Zone (Weber and Juckenack 1990; Will et al. 2015).

Analytical techniques and sampling

Laser-ablation (LA)-ICP-MS method

Samples for isotope mass spectrometry were processed at the Institut für Geowissenschaften of Frankfurt University using standard mineral separation techniques. These include crushing and sieving, before concentration of heavy minerals by heavy liquids (bromoform, methylen-iodide) and magnetic-separation with a Frantz isodynamic separator. Hand-picked zircon grains were mounted in 25 mm-diameter circular epoxy mounts and polished to expose a section at their inner core. Prior to their analysis, the grains were examined using cathodoluminescence (CL) imaging to recognize their internal structure and to identify cracks and mineral inclusions. Zircon U–Pb isotope analysis was performed by LA-ICP-MS technique using a ThermoFinnigan Element II sector field ICPMS attached to a New Wave LUV213 laser ablation system ($\lambda = 213$ nm). Ablation was carried out in a He carrier gas in a low volume (2.5 cm^3) cell; laser beam parameters used were 30 μm diameter; 5 Hz repetition rate 75% power output. Isotope data were acquired in peak-jumping mode on eight masses;

^{202}Hg , ^{204}Pb , ^{206}Pb , ^{207}Pb , ^{208}Pb , ^{235}U and ^{238}U . Background and ablation data for each analysis were collected over 90 s, with background measurements (carrier gas, no ablation) being taken over the first 30 s prior to initiation of ablation. Data were collected at time-resolved mode allowing acquisition of the signal as a function of time (ablation depth), and subsequently recognition of isotopic heterogeneities within the ablated volume. Raw data were processed offline using an Excel® spreadsheet program (Frei and Gerdes 2008). Mass discrimination of the MS, and elemental fractionation during laser ablation were corrected by calibration against the GJ-1 zircon standard (Jackson et al. 2004), which was analyzed routinely during analytical sessions (three standard analyses at the beginning and end of every session of 33 unknowns, and two standard analyses every 10 unknowns). Prior to this correction, the change of elemental fractionation (e.g. Pb/U and Pb/Th ratios as function of ablation time and thus depth) was corrected for each set of isotope ratios by applying a linear regression through all measured ratios versus time, excluding some outliers (>2 s.e.), and taking the intercept $t=0$ as the correct ratio. Changes in isotopic ratios arising from laser drilling into domains of distinct Pb/U ratio (core/rim), mineral inclusions, and zones affected by Pb loss (metamictization/cracks), can usually be detected by careful monitoring of the time-resolved signal, such analyses are normally rejected. Common Pb correction was applied only when the interference- and background-corrected ^{204}Pb signal was significantly higher than the detection limit of about 20 cps. The latter is limited by the amount of Hg in the carrier gas and the accuracy to which the ^{202}Hg and thus the interfering ^{204}Hg can be monitored. Corrections made were based on common Pb composition given by the second stage growth curve of Stacey and Kramers (1975). Data presentation was made with Isoplot (Ludwig 2001). To monitor the reproducibility and accuracy of our analytical procedure, the standard zircon 91,500 (Wiedenbeck et al. 1995) has been reproduced with an age of 1063 ± 3 Ma.

Sampling

For provenance analyses we used samples of the schist envelope of the Böllsteiner Odenwald, which have been drilled by the Heghölzchen-Wiebelsbach drilling (R 3496800, H 5521300; Fig. 2). Details of the Heghölzchen-Wiebelsbach drilling are reported by Loeckle et al. (2016). The analysed samples HW06 (20.6 m depth) and HW20 (39.2 m depth) consist of an intercalation of mylonitic and weakly foliated gneisses. Mylonitic sections are marked by lenticular or sheet-like quartzofeldspathic domains, while weakly foliated and fine grained parts often display a compositional layering with only few quartzofeldspathic lenses. Estimates in thin section revealed 30–40 vol% plagioclase,

20–40 vol% biotite, 20–30 vol% quartz, up to 10 vol% K-feldspar, and 5–10 vol% garnet as the major constituents in all domains. The rock is thus classified as garnet-biotite gneiss (Loeckle et al. 2016). Accessory titanite and sillimanite are frequent in mylonitic sections and are always associated with biotite (see Fig. 7e in Loeckle et al. 2016). The overall structure and the type of accessory minerals suggest that the Heghölzchen-Wiebelsbach drilling samples are metasediments. Garnet of various sizes is present in all sections and usually displays pronounced strain caps and strain shadows. From gneisses exposed in surface outcrops of the Wiebelsbach area (Gauss Krüger coordinates: R 3496367, H 5521433, 207 m a.s.l.) a top-to-the-ESE sense of shear has been determined (Loeckle et al. 2016).

Results

The LA-ICP-MS U–Pb data obtained from detrital zircons of the biotite-garnet paragneiss samples are reported with 2σ uncertainties in Supplementary Tables 1 and 2. The U–Pb results are presented on density/probability plots in Figs. 3 and 4. Only analyses with a 90–110% concordance (88 grains from 147) are plotted as ^{238}U – ^{206}Pb ages. If not stated elsewhere, the age peaks are calculated as concordia age of a zircon population (Ludwig 2001). In most cases the oscillatory zoned parts between the core and rim of the detrital zircons are measured, because these parts reflect the undisturbed zircon growth, and thus an undisturbed U–Pb system of the zircons with no or minor lead loss. Two analyses (A466 and A467 of sample HW20) of an oscillatory zoned zircon without a core yielded $^{206}\text{Pb}/^{238}\text{U}$ ages of 476 ± 8 and 489 ± 10 Ma, which overlap in their uncertainties.

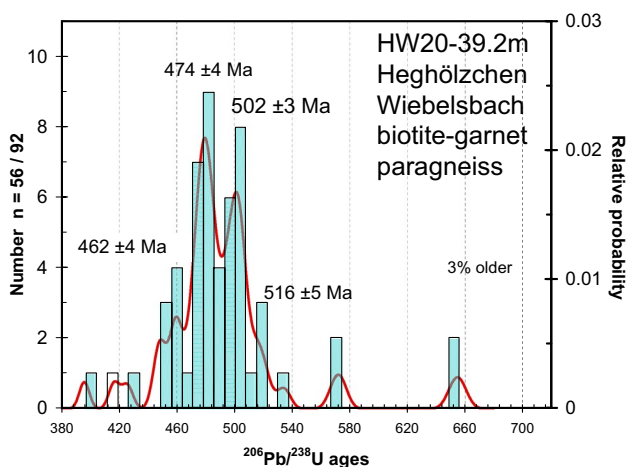


Fig. 3 Frequency/density plot of detrital zircons of metasediment (paragneiss) of the sample Wiebelsbach HW20 (Böllstein Odenwald, MGCZ). 90–110% concordance of the $^{206}\text{Pb}/^{238}\text{U}$ ages for zircons

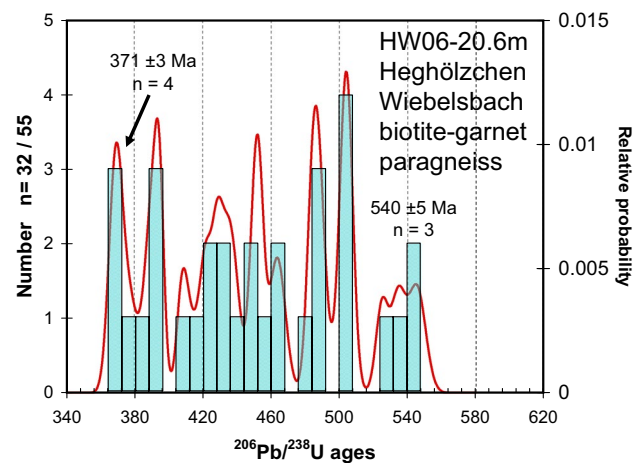


Fig. 4 Frequency/density plot of detrital zircons of metasediment (paragneiss) of the sample Wiebelsbach HW06 (Böllstein Odenwald, MGCZ). 90–110% concordance of the $^{206}\text{Pb}/^{238}\text{U}$ ages for zircons

Biotite-garnet paragneiss Wiebelsbach

92 zircons from the biotite-garnet paragneiss HW20 were analyzed. Only 56 analyses display a concordance of 91–104%. The grain size of the zircons is ranging from 80 to 220 μm . Most of them are colorless, only few well rounded zircons are pink. 30% of the zircons are angular to subhedral. Most of them (57%) are rounded and spherical. Only 9% are elongated and rounded. 3% are subhedral to euhedral. 56% of the zircons have Th/U ratios between 0.1 and 0.5, which are typical for magmatic zircons. 39% display ratios between 0.7 and 1.1. Only two Paleoproterozoic zircons (zircons A424 and A425) are below the Th/U ratio of 0.1, which is related to a metamorphic origin of zircons (Hartmann and Santos 2004).

The youngest analysis A444 (Supplementary Table 1), with an age of 396 ± 7 Ma and a Th/U ratio of only 0.13, has been obtained from the tip of a pyramid of an angular zircon, which contains a zoned core with an age of 493 ± 9 Ma and a Th/U ratio at 0.53, typical for a magmatic origin. Analysis A516 is 7% discordant, but the next older zircon (A429) is concordant at 426 ± 8 Ma. The age spectrum of the detrital zircons is dominated by Paleozoic ages. Only 7% Ediacaran, 3% Paleoproterozoic and 2% Archean zircons are present. The Ordovician zircons (49%) are forming the main group with a main age peak at 474 ± 4 Ma and a second peak at 462 ± 4 Ma (Fig. 3). There is a significant input of Cambrian zircons (33%) with an age peak at 502 ± 3 Ma and a less significant peak at 516 ± 5 Ma (Fig. 3). Two analyses (A481 and A482, Supplementary Table 1) of a sector zoned zircon (probably granulite facies) yielded nearly identical $^{206}\text{Pb}/^{238}\text{U}$ ages of 500 ± 9 and of 502 ± 9 Ma, which are compatible with the Cambrian main peak.

Sample HW06 contains 55 zircons (32 zircons with a 91–104% concordance) and a lot of apatite. The grain size of zircons ranges from 60 to 160 μm . Most of them are colorless, only few rounded zircons are yellow or brown. 24% of the zircons are angular to subhedral. Most of them (46%) are elongated and rounded. Only 26% are rounded and spherical. 2% are subhedral to euhedral. The Th/U ratios of the zircons are similar to those of the sample HW20. Most of them (44%) have ratios between 0.1 and 0.5, which are typical for magmatic zircons.

The age spectrum of the detrital zircons of sample HW06 is similar to that of sample HW20, but it does not contain Precambrian detrital zircons. There are four zircons (Supplementary Table 2) which proof later lead loss. The youngest zircon population is angular to long prismatic in shape and from core to rim oscillatory zoned. The U–Pb ages are slightly younger than in sample HW20 defining a concordia age of 371 ± 3 Ma (Fig. 4). The age spectrum of the detrital zircons is dominated by Paleozoic ages. The Cambrian zircons (25%) are forming the main group with an ages peak at ca. 500 Ma and a second peak at 540 ± 5 Ma (Fig. 4). There is again a striking input of Ordovician zircons (22%) with an age peak at ca. 480 Ma. Sample HW06 contains much more Middle to Upper Devonian (16%) zircons than sample HW20 (1%). One angular to subhedral zircon contains an Ediacaran to Early Cambrian core (analysis 168: 542 ± 13 Ma, Supplementary Table 2) and a Lower Devonian oscillatory zoned middle part (analysis 167: 415 ± 10 Ma, Supplementary Table 2).

Discussion

Depositional age of the paragneiss protolith

For the correlation of detrital zircon U–Pb age spectra, the stratigraphic age of the investigated rock is important. If the rock contains no fossils or the latter were destroyed by metamorphic overprint and/or deformation, than the youngest U–Pb zircon age is often used to define a maximum depositional age. The youngest possible deposition age can be constrained using the age of an unconformity, of cross cutting dykes, of metamorphic overprint and exhumation/cooling. Dickinson and Gehrels (2009) used the youngest U–Pb ages of individual detrital zircons to correlate the maximum depositional age of clastic rocks from the Colorado Plateau with the chronostratigraphy. In 95% of these cases the maximum depositional ages are compatible with the stratigraphic age of the clastic rocks (Geological Time Scale, Ogg 2004a, b). We used multiple zircon grain clusters to determine robust maximum depositional ages calculated with the function “concordia age” of the software Isoplot/Ex 2.49 (Ludwig 2001) with analyses of

98–102% concordance to obtain a reliable youngest zircon population.

As the two analyzed samples HW06 and HW20 are only 18 meters apart, and their U–Pb age spectra of detrital zircons are similar, the maximum deposition age of the paragneiss protolith of both samples will be discussed together. The youngest concordant analyses (A444, Supplementary Table 1) of a zircon rim from sample HW20 yielded an age of 396 ± 7 Ma containing a Cambrian core. The next older zircon (A429) is concordant at 426 ± 8 Ma. Thus, the youngest zircons could be Lower Devonian or Silurian in age. Sample HW06 contains more Devonian zircons. Measurements from oscillatory zoned cores of four zircons define the youngest population at 371 ± 3 Ma (Fig. 5a) of the schist envelope which is interpreted as maximum deposition age of the metasedimentary sequence. The stratigraphic range of the deposition of the paragneiss protolith is bracketed between the maximum deposition age at 371 ± 3 Ma and the age of the amphibolite facies metamorphism of the schist envelope as well as the intrusion of granitoids at 375 ± 5 Ma (Todt et al. 1995). The intrusion age is the same like the maximum deposition age when considering the uncertainties. Two interpretations are possible: (1) If the youngest zircon population is influenced by a strong lead loss, than a Middle Devonian deposition age is possible. (2) In cases of no lead loss, the deposition age of the metasediments could be Upper Devonian (Famennian).

Sedimentation of the paragneiss protolith, granite intrusion, and metamorphic overprint should have occurred between 374 Ma and ca. 365 Ma. The granitoids might have intruded at ca. 370 Ma into the Upper or Middle Devonian clastic sediments, which together were overprinted by an amphibolite facies metamorphism <370 Ma. The amphibolite facies metamorphism is not younger than Tournaisian (Lower Carboniferous), because cross-cutting undeformed lamprophyric dykes of the Oetzberg zone yielded a Viséan ^{39}Ar – ^{40}Ar biotite cooling age of 327 ± 3 Ma (Hess and Schmidt 1989).

In summary, the Upper Devonian maximum deposition age of the paragneiss protolith of the northern Böllstein Odenwald is much younger than expected. Silurian to Lower Devonian intrusion ages of granitoids of the southern Böllstein Odenwald (Lippolt 1986; Reischmann et al. 2001) proves a pre-Silurian, probably Ordovician or Cambrian, deposition age. Altenberger et al. (1990) and Reischmann et al. (2001) interpret these Lower Palaeozoic metasediments with tholeiites of the southern Böllstein Odenwald as a continent-island arc collisional setting. The Devonian sedimentation age of the northern schist envelope is in striking contrast with the data obtained from the southern Böllstein Odenwald. The intrusion of granitoids in the northern Böllstein complex must be much younger (probably Lower Carboniferous; but not older than 371 Ma) than

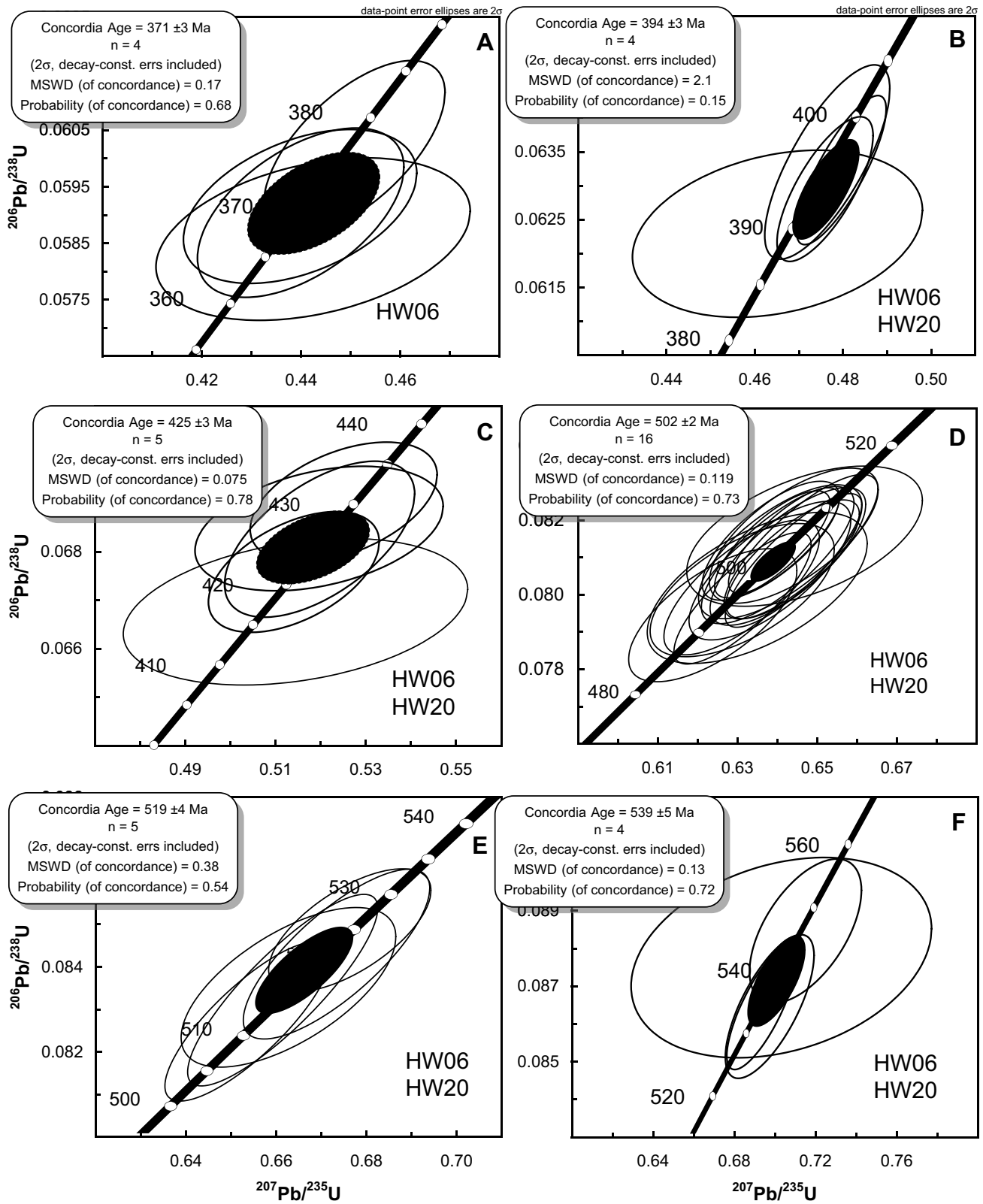


Fig. 5 U–Pb concordia plots for of detrital zircons of metasediments (paragneiss) of the samples Wiebelsbach (Böllstein Odenwald, MGCZ)

the intrusions in the southern Böllstein Odenwald. Based on the different sedimentation ages, the Böllstein Odenwald must be divided into to an older, southern part and a younger, Devonian to Carboniferous northern part. Upper Devonian deposition ages of clastic sediments are yet not known from the MGCZ, but they are typical for the synorogenic clastic sediments of the Saxothuringian Zone (Erbendorf Palaeozoic, Adam and Reuter 1981, Fig. 1). Upper Devonian synorogenic clastic sediments are also known from Saxothuringian related nappes of the Rhenohercynian Zone (Henningsen 1966; Stibane et al. 1984; Dörr 1986).

Provenance

The U–Pb age spectrum of detrital zircons separated from clastic sediments of the Böllstein Odenwald will be compared with published age spectra obtained from potential protoliths of the Rhenohercynian and Saxothuringian Zone. The composition of the age spectrum can help to reconstruct the source area. The age spectrum of detrital zircons from Upper Devonian paragneisses of the schist envelope of the northern Böllstein Odenwald is unique in the crystalline basement of the MGCZ and of the European Variscides in general (Murphy et al. 2004a; Geisler et al. 2005; Gerdes and Zeh 2006; Zeh and Gerdes 2009; Linnemann et al. 2004, 2008, 2012; Drost 2008; Drost et al. 2010; Eckelmann et al. 2013; Fernández-Suárez et al. 2013; Dörr et al. 2015; Zulauf et al. 2014; Hajna et al. 2013, 2016). The only available U–Pb age spectra of detrital zircons from the MGCZ (Ruhla Crystalline Complex, Fig. 1) are characterized by Precambrian ages. The Silurian (youngest zircon 435 ± 9 Ma) Rögis quartzite (Fig. 6, green line, North Ruhla, Zeh and Gerdes 2009) contains 78% Mesoproterozoic zircons together with only 1% Ediacaran input which is typical for Baltica-derived detritus of the Rhenohercynian Zone (Haverkamp 1991; Geisler et al. 2005; Eckelmann et al. 2013). The Lower Palaeozoic metapelite (South Ruhla, Brotterode group, sample KuK) also contains a high amount of Precambrian zircons (68%) similar to the Silurian Rögis quartzite, but with 30% Ediacaran input together with a Mesoproterozoic age gap indicative for Gondwana derived detritus of the Armorican terrane (Fig. 6, red line, Zeh and Gerdes 2009). Based on the different age spectra of the metapelite (without Mesoproterozoic zircons) and Rögis quartzite (with Mesoproterozoic zircons) Zeh and Gerdes (2009) assessed the Rhenic suture inside the Ruhla Crystalline Complex of the MGCZ.

The metasediments of the schist envelope of the northern Böllstein Odenwald contains only four Precambrian zircons (Fig. 6, black line), which could not be used for provenance analyses. The only similarity between the South Ruhla metapelite and the paragneiss of the northern Böllstein schist envelope is the high amount of Cambrian

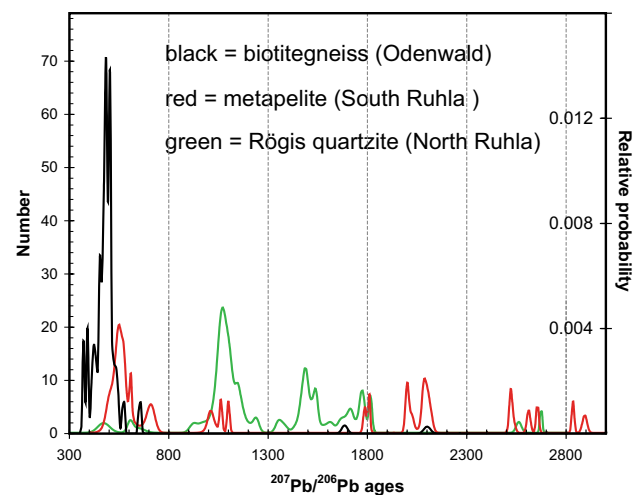


Fig. 6 Comparative relative probability plot of metasediments of the samples Wiebelsbach (*black line* Böllstein Odenwald, MGCZ) with a metapelite of South Ruhla Crystalline Complex (*red line* Zeh and Gerdes 2009) and with the Rögis quartzite of North Ruhla Crystalline Complex (*green line* Zeh and Gerdes 2009)

zircons (ca. 30%) in both samples (Fig. 6). Cambrian and Ordovician sedimentary rocks of the Saxothuringian Zone (Schwarzburg Anticline, Linnemann et al. 2007) also display an Armorican-type detrital zircon age cluster typical for West Gondwana provenance. Similar to the sample from the southern Ruhla Crystalline Complex they received also a high input of cratonic zircons (35%, 1.7–3.4 Ga). Cratonic zircons, however, are almost absent in the northern Böllstein metasediments (4%).

A comparison between the zircon age spectrum of Upper Devonian metasediments of the MGCZ (Böllstein Odenwald: black line, see Fig. 7) with that of the contemporaneous Middle to Upper Devonian sandstones of the Rhenohercynian Zone (Old Red Sandstone, Givetian, Brabant Massif: blue line in Fig. 7, Linnemann et al. 2012 and Famennian Sandstone of the Dill syncline: magenta line in Fig. 7; Eckelmann et al. 2013) in a relative probability plot displays significant differences. The Famennian Sandstone of the Dill syncline contains 41% Mesoproterozoic zircons, 41% Paleoproterozoic zircons and 3% Silurian zircons (Fig. 7: magenta line). This ratio of detrital zircons is typical for Baltica/Laurentia sources. The low amount of Lower Paleozoic zircons (7% Ordovician, no peak, 0% Cambrian) of the Famennian Sandstone is in contrast to the large number of Lower Paleozoic zircons (42% Ordovician, 30% Cambrian, Fig. 8) of the metasediments of the Odenwald, which contain only 3 Mesoproterozoic and Paleoproterozoic zircons. The Givetian conglomerate of the Brabant Massif also contains only few Lower Paleozoic zircons (1 grain = Ordovician, 3 grains = Cambrian) and again only few Silurian

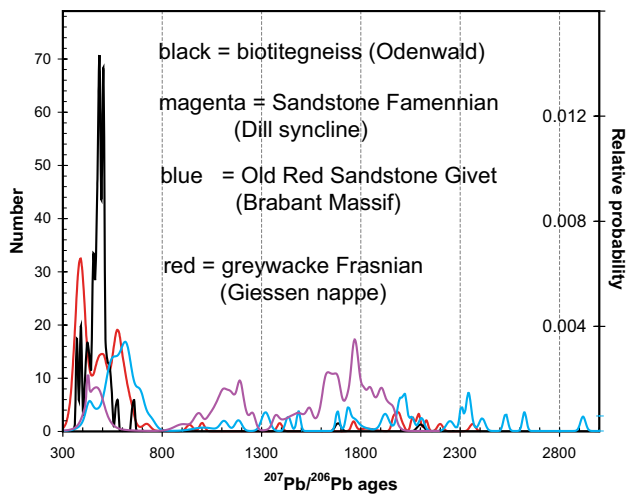


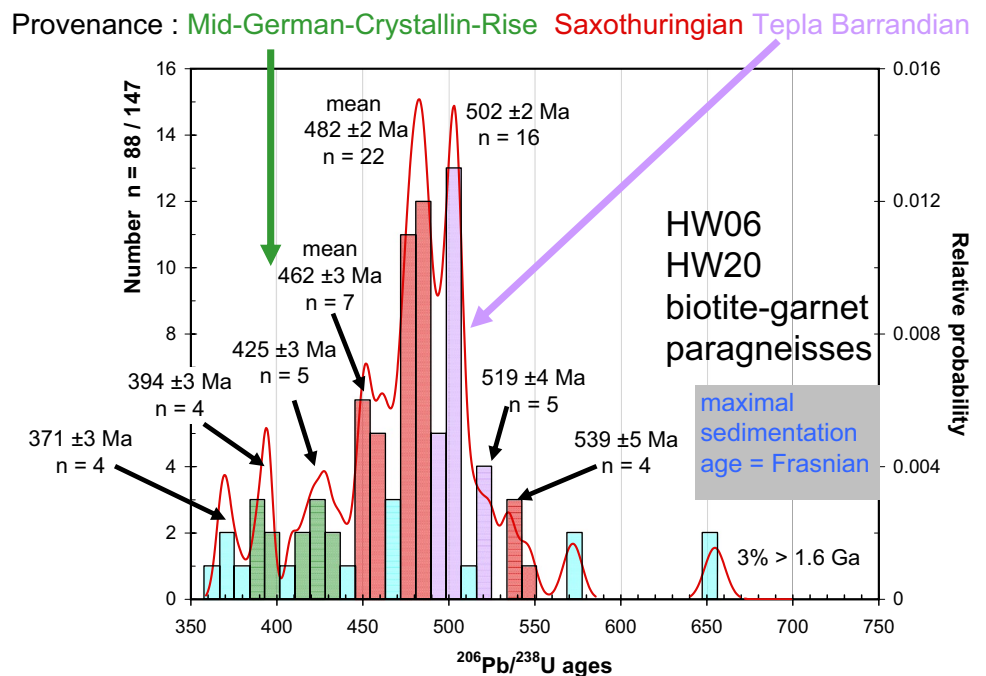
Fig. 7 Comparative relative probability plot of metasediments of the samples Wiebelsbach (black line Böllstein Odenwald, MGCZ) with Givetian Old Red Sandstone of the Brabant Massif (blue line Linnemann et al. 2012), Famennian sandstone of the Dill syncline (magenta line Rhenohercynian Zone, Eckelmann et al. 2013) and Frasnian greywacke from base of the Giessen nappe (red line Supplementary Table 3, sample description see Dörr 1986)

(4%) zircons. Despite of the Givetian deposition age, Devonian zircons are not available in the source area. The high amount of Proterozoic zircons (89%, with main Ediacaran age peaks at 537 ± 6 Ma, $n=6$, 600 ± 6 Ma, $n=7$ and 626 ± 7 Ma, $n=5$) of the Old Red Sandstone is not compatible with the spectrum of the contemporaneous metasediments of the Böllstein Odenwald (Fig. 7). The Devonian sediments of the Rhenohercynian Zone

received their zircons from a different source and thus cannot form the protolith of the paragneiss of the northern Böllstein Odenwald. Only a Frasnian greywacke (Dörr 1986) analyzed from the Giessen nappe (Fig. 7 red line, Supplementary Table 3, Armorican provenance) displays a detrital zircon age spectrum with Devonian zircons, which are similar to those of the Upper Devonian metasediments of the Böllstein Odenwald.

The U–Pb age spectrum of the detrital zircons of the northern Böllstein Odenwald is characterized by Palaeozoic zircon age peaks, which can be correlated with the magmatic rocks exposed in the entire Saxothuringian Zone and in the northern part of the Teplá-Barrandian Unit. The oldest detrital zircon population with a U–Pb age of 539 ± 5 Ma ($n=4$, Figs. 5f, 8) correlates with the U–Pb ages of zircons of granodiorite of the eastern Saxothuringian Zone (Kröner et al. 2001; Dörr et al. 2002; Zelazniwicz et al. 2004; Linnemann 2003; Linnemann et al. 2008, 2013). The Cambrian age peak of the detrital zircons of the northern Böllstein Odenwald at 519 ± 4 Ma ($n=5$, Figs. 5e, 8) correlates with U–Pb ages of zircons from gabbro and quartz diorite at 522 ± 2 Ma of the Teplá-Barrandian Unit (Zulauf et al. 1997; Dörr et al. 2002). The prominent Cambrian age peak at 502 ± 2 Ma ($n=16$, Figs. 5d, 8) fits with U–Pb ages of zircons from granitoids at 502 ± 2 Ma from the Saxothuringian Zone (Vesser Complex and the Polish West Sudetes; Kröner et al. 2001; Linnemann et al. 2007) and with U–Pb ages from gabbros and orthogneisses at 502 ± 2 Ma from the Teplá-Barrandian Unit and Mariánské Lázně Complex (Bowes and Aftalion 1991; Dörr et al. 1998; Timmermann et al. 2006).

Fig. 8 Provenance of meta-sediment of the samples Wiebelsbach: green color Mid-German-Crystalline Zone, red color Saxothuringian Zone, magenta Teplá-Barrandian Unit (Moldanubian Zone), blue color no correlation



The Lower Ordovician age of detrital zircons of the schist envelope (northern Böllstein Odenwald) at 482 ± 2 Ma ($n=22$, mean $^{206}\text{Pb}/^{238}\text{U}$ age, Fig. 8) reflects the main magmatic events of the Saxothuringian Zone (Linnemann et al. 2008 and references therein) ranging from 479 ± 2 to 486 ± 4 Ma (volcanic rocks) and from 485 ± 6 to 489 ± 6 Ma (granites). Lower Ordovician zircons can also be directly delivered from the MGCZ, because Anthes and Reischmann (2001) dated the syenite of the borehole Volkach at 488.8 ± 0.6 Ma (Pb–Pb zircon age). Similar ages have been obtained from volcanic-arc granite boulders of an Upper Devonian conglomerate of the northern Vosges (Dörr et al. 1992) pointing to Ordovician source rocks during the Devonian at the southern boundary of Saxothuringian Zone. The prominent Lower Ordovician age peak of the detrital zircons of the northern Böllstein Odenwald also fits the major magmatic episode in the French Massif Central, Armorican Massif and of the Allochthonous Complex in Spain (Dias da Silva et al. 2016).

The smaller Late Ordovician age peak of the detrital zircons at 462 ± 3 Ma ($n=7$, mean $^{206}\text{Pb}/^{238}\text{U}$ age, Fig. 8) of the Böllstein Odenwald is similar to the age of orthogneisses situated at the Saxothuringian/Moldanubian boundary (457 ± 2 Ma) and to the age of the Saxothuringian metavolcanic rocks (450 ± 4 Ma, Teufel 1988). Late Ordovician igneous rocks dated at 458 ± 4 Ma (volcanic rock), 445 ± 2 Ma (tuffite) and 444 ± 6 Ma (dacite) are also known from the Rhenohercynian Zone (Ardennes, Brabant Massif, U–Pb on zircon, Linnemann et al. 2012). These igneous rocks are intercalated with Ordovician sandstones which contain 69% Neoproterozoic and 5% Mesoproterozoic detrital zircons. The Ordovician sequence of thin volcanic rock layers, sandstones and slates are covered by a ca. 3 km thick sequence of Silurian slate and sandstone, which contain 48% Neoproterozoic, 18% Mesoproterozoic and only 4% Late Ordovician detrital zircons, typical for a recycled Avalonian spectrum (Linnemann et al. 2012). The igneous rocks of the Brabant Massif could be correlated with Late Ordovician sub-volcanic intrusions of North England (Lake district, Hughes et al. 1996).

The Silurian detrital zircons (425 ± 3 Ma, $n=5$, Figs. 6c, 8) of the Middle to Upper Devonian Böllstein paragneiss could be correlated with the Silurian granodiorite-gneisses the southern Böllstein Odenwald (418–435 Ma, Rb–Sr whole rock, Lippolt 1986) and the Spessart area (418 ± 18 Ma, Pb–Pb on zircon, Dombrowski et al. 1995). South of the Rhenohercynian Zone, in the Northern Phyllite Belt (Fig. 1), a Silurian volcano-sedimentary sequence is present, dated at 442 ± 22 Ma (tuffite), $433 + 9/-7$ Ma (metadacites) and $426 + 14/-15$ Ma (sericite-gneiss) (U–Pb on zircon, Sommermann et al. 1992). This sequence is covered by 4 km thick Lower Devonian (Reitz 1989) shelf sediments of

the Rhenohercynian Zone (Klügel 1997). Early Silurian igneous rocks intruding at 430 ± 3 Ma (quartz diorite) into Ordovician sediments are known from the Brabant Massif (Ardennes). These small igneous bodies are also covered by a ca. 3 km thick sequence of Silurian slates and sandstones (Linnemann et al. 2012).

The Lower Devonian zircons of the northern Böllstein Odenwald paragneiss with an U–Pb age peak at 394 ± 3 Ma (Figs. 5b, 8) could be derived from (1) the nearby orthogneisses of the southern Böllstein Odenwald (386–405 Ma, U–Pb and Pb–Pb on zircon, Reischmann et al. 2001), (2) the Spessart basement (410 ± 18 Ma, Pb–Pb on zircon, Dombrowski et al. 1995), and (3) the orthogneisses situated at the Saxothuringian/Moldanubian boundary (ca. 404 Ma, U–Pb on zircon, Teufel 1988). The Upper Devonian detrital zircons (Figs. 5a, 8) of the northern Böllstein paragneiss prove the existence of a younger magmatic arc as Silurian, partly probably coeval with the deposition of the metasediments. Similar U–Pb ages of detrital zircons are also known from the Upper Devonian autochthonous flysch of the Saxothuringian Zone with Armorican affinity (Erbendorf Palaeozoic, Schäfer et al. 1997). Devonian U–Pb zircon and monazite ages (380 to 387 ± 3 Ma) have also been reported from the Moldanubian basement (Teplá-Barrandian Unit and Mariánské Lázně Complex; Timmermann et al. 2004, 2006). This Devonian thermal event is widespread in crystalline basement of the Saxothuringian Zone. In the allochthonous units of the Saxothuringian Zone ^{39}Ar – ^{40}Ar and K–Ar ages of 390 Ma (3 T phengites in eclogite), 380 Ma (hornblende, also Rb–Sr mica/whole), and 372 Ma (muscovite in pegmatites, Kreuzer and Seidel 1989; Kreuzer et al. 1989; Söllner et al. 1981) are common. The presence of these HP phengites in Upper Devonian autochthonous flysch of the Saxothuringian Zone is interpreted by Schäfer (1997) that the allochthonous unites (former part of the Moldanubian Zone) were exhumed during Middle Devonian times and subsequently thrust over the rocks of the Saxothuringian Zone.

Middle Devonian igneous rocks related to extensional tectonics are known from the eastern Rhenohercynian Zone (Lahn-Dill syncline). They are intercalated with / or intruded into Devonian sediments which contain 40–60% Mesoproterozoic zircons (Haverkamp 1991; Eckelmann et al. 2013). Middle to Upper Devonian igneous rocks are also known from the western Rhenohercynian Zone (Ardennes). In the Stavelot Massif a granitoid intruded at ca. 381 ± 8 Ma (Kramm and Buhl 1985) into Cambro-Ordovician sediments which contain 7% Ediacaran and 40% Mesoproterozoic detrital zircons typical for Avalonia sources (Willner et al. 2013). A lower intercept age of $373 + 8/-9$ Ma (discordant multigrain fractions) is interpreted by Goffette et al. (1991) as Middle to Upper Devonian emplacement age of a microgranite dyke, which

also intruded into Cambrian to Ordovician sediments of the Rocroi Massif (Ardennes).

In summary, the sequence of the magmatic events of the Bohemian Massif (Saxothuringian and Moldanubian Zone) fits well with the U–Pb age peaks of the detrital zircons of the schist envelope of the northern Böllstein Odenwald. The main igneous activity in the western Rhenohercynian Zone (Ardennes, Brabant Massif) extends from the Late Ordovician up to the mid-Silurian (Linnemann et al. 2012). During the Upper Devonian deposition of the Böllstein (meta)sediments, these volcanic rocks were covered by Silurian to Lower Devonian sediments referred to as Caledonian Molasse. The small Devonian plutons and dykes of the Ardennes intruded into Lower Palaeozoic sediments and were also covered at this time. The Middle Devonian igneous rocks of the eastern Rhenohercynian Zone (Lahn-Dill syncline) were exposed at the surface during the Upper Devonian deposition of the Böllstein (meta)sediments. If these igneous rocks would have served as the source of the Böllstein (meta)sediments, a significant amount of Mesoproterozoic zircons, derived from the associated sandstones, should occur in the age spectrum of the detrital zircons of the Böllstein (meta)sediments. This, however, is not the case (Figs. 7, 8). Recycling of sedimentary rocks of the Rhenohercynian Zone can be ruled out as most of them contain 20–60% Mesoproterozoic detrital zircons, derived from Baltica (Haverkamp 1991; Geisler et al. 2005; Zeh and Gerdes 2009; Eckelmann et al. 2013; Willner et al. 2013). Only the Upper Devonian clastic rocks of the Brabant Massif contain a high amount of Ediacaran zircons apart from minor Mesoproterozoic zircons, typical for recycled zircons derived from Avalonia and Laurentia (Linnemann et al. 2012).

The provenance areas of Upper Devonian detrital zircons of the northern Böllstein Odenwald are the igneous rocks of the Saxothuringian Zone together with the MGCZ. The Silurian (8%) and Devonian (11%) age peaks of the detrital zircons of the Böllstein Odenwald point to a proximal source, such as the MGCZ. Magmatic rocks, with ages typical for the Bohemian Massif (ca. 540, 520, 500 Ma), which underwent Devonian metamorphism at ca. 380 Ma, are entirely missing in the Rhenohercynian Zone.

Conclusions

The following conclusions can be drawn from the present study:

1. Middle to Upper Devonian deposition ages of clastic sediments (now paragneisses) have not been published yet from the MGCZ. They are typical for the oldest synorogenic clastic sediments of the Saxothuringian Zone exposed in the Bohemian Massif (Fig. 1; Erben-dorf Palaeozoic, Adam and Reuter 1981; Schäfer et al. 1997) and in the Vosges (Wickert and Eisbacher 1988; Dörr et al. 1992).
2. The unique age spectrum of detrital zircons of the northern Böllstein paragneiss resembles the sequence of magmatic events of the Saxothuringian Zone, but is incompatible with the coeval age spectra of detrital zircons of Rhenohercynian clastic sediments, which display a strong Baltic input.
3. The provenance area of the Upper Devonian metasediments of the northern Böllstein Odenwald is characterized by Upper Cambrian detrital zircons, typical for the late Cadomian magmatic arc documented in the Bohemian Massif (Teplá-Barrandian Unit, Hajna et al. 2013, 2016; Armorican terrane) and by Early Ordovician zircons representing the main igneous activity of the Saxothuringian Zone.
4. The high amount of Silurian and Devonian zircons (19%) together with the restricted age spectrum of the Upper Devonian metasediments of the Böllstein Odenwald point to a deposition in an intra arc or trench setting, probably within a basin situated at the northern boundary of the Saxothuringian Zone.
5. The new results are not in line with published tectonic models assuming that parts of the MGCZ are forming Rhenohercynian crust (Oncken 1997; Will and Schmädicke 2003; Will et al. 2015; Franke and Dulce 2016).

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