Boninitic blueschists record subduction initiation and subsequent accretion of an arc–forearc in the northeast Proto-Tethys Ocean

Dong Fu^{1*}, Bo Huang¹, Tim E. Johnson², Simon A. Wilde², Fred Jourdan², Ali Polat^{3,1}, Brian F. Windley^{4,1}, Zhaochu Hu¹, Timothy Kusky^{1*}

Supplemental Text and Figures

Supplemental text: Detailed description of analytical methods, description of phase equilibrium modelling, geological data and evaluation of element mobility.

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Figure S8 Summary of peak P – T conditions and P – T paths of HP metamorphic rocks in the North Qilian orogenic belt;

ANALYTICAL METHODS

1. Mineral chemistry

The mineral chemistry of major elements was conducted using a JEOL JXA-8230 electron microprobe at the Center for Global Tectonics (CGT), School of Earth Sciences, China University of Geosciences (CUG), Wuhan. Analytical conditions for each spot were 15 kV accelerating voltage, 20 nA beam current with 1–5 μm beam diameter. Each analysis includes 5 s background counting time and 10–20 s routine analysis. The analytical data were corrected online using a ZAF correction procedure. Natural and synthetic minerals, including albite (Na), orthoclase (K), diopside (Si, Mg, Ca), yttrium–aluminium garnet (Al), magnetite (Fe), apatite (P), and rutile (Ti), served as standards. Analytical uncertainties were less than 2 %.

2. Whole-rock geochemistry

Samples were crushed to a 200-mesh powder in an agate mill. The major oxides were analyzed using an X-ray fluorescence spectrometer (XRF) at the State Key Laboratory of Geological Progress and Mineral Researches (GPMR), CUG, Wuhan. Loss on ignition (LOI) was measured by heating a portion of the sample for two hours at 1000°C. Analytical uncertainties are better than 5%. For trace elements, sample powders were dissolved in a $\text{HNO}_3 + \text{HF}$ solution and measured using an Agilent 7700 system inductively coupled plasma-mass spectrometer (ICP-MS) at GPMR, CUG. Detailed sample digesting and analytical procedures are given in ([Liu et al., 2008](#)). International standards included AGV-2, BHVO-2, BCR-2, and RGM-2 and the analytical precision for trace elements was better than 5%.

3. Titanite U–Pb dating

The blueschist sample (17QL27-1) was crushed and titanite grains were separated using standard magnetic and heavy liquid techniques. Selected titanite grains with good crystal shapes were hand-picked, mounted in epoxy resin and polished to expose internal structures. Transmitted and reflected light images were taken to record internal structures, as well as backscattered electron (BSE) images that were acquired

using a JEOL JXA-8230 electron microprobe with a 15 kV accelerating voltage at the CGT, CUG, Wuhan. Titanite U–Th–Pb isotopic and trace element measurements were conducted at the John de Laeter Centre at Curtin University using a 193 nm excimer laser ablation system coupled to an Agilent 7700s quadrupole mass spectrometer. Detailed operating conditions are given in [Kylander-Clark et al. \(2013\)](#). The analytical spot diameter was 38 μm , with a laser fluence of 2 J cm^{-2} and a repetition rate of 5 Hz. The uncertainties of ages are given at the 2σ level. Primary reference standard OLT1 (1014.8 ± 2.0 ; [Kennedy et al., 2010](#)) and secondary standards BLR-1 (1047.1 ± 0.4 Ma; [Aleinikoff et al., 2007](#)) and MKED1 (1517.32 ± 0.32 Ma; [Spandler et al., 2016](#)) were used to monitor data accuracy and to correct for mass fractionation. Data reduction and plotting were performed using the software Iolite and Isoplot, respectively ([Paton et al., 2011](#); [Ludwig, 2003](#)). Trace element data (including P, Ti, V, Zr, Hf, Nb, Ta and REE) were collected concurrently with U–Pb data, using primary and secondary standards BLR, MKED and OLT.

4. Zircon U–Pb dating

Zircon grains from the mafic schist (18QL03) were separated for zircon U–Pb dating. Grains were separated by magnetic and heavy liquid techniques and then hand-picked, mounted in epoxy resin and polished to expose the grain interiors. Cathodoluminescence (CL) images were acquired at the GPMR, CUG, Wuhan using a Gatan Mono CL4+ instrument installed on a Quanta 450 FEG scanning electron microscope (SEM). Grains were analyzed for U–Pb isotopes using both a high sensitive high-resolution ion microprobe (SHRIMP) at the John de Laeter Centre, Curtin University and an Agilent 7500a inductively coupled plasma-mass spectrometry (ICP-MS), with a 193 nm ArF-excimer laser ablation, at the GPMR, CUG, Wuhan. Operating conditions and analytical procedures for the SHRIMP followed [Williams \(1998\)](#). The zircon standard M257 ($^{206}\text{Pb}/^{238}\text{Pb}=0.09100$, corresponding to an age of 561.3 Ma; [Nasdala et al., 2008](#)) was used for Pb/U ratio corrections. Data were reduced and plotted using the SQUID and Isoplot software, respectively ([Ludwig, 2001](#); [Ludwig, 2003](#)). The analytical data are presented with 1σ

uncertainties. LA-ICPMS analysis used a 32 μm spot diameter. Zircon 91500 was analyzed twice every 5 analyses as an external standard. National Institute of Standards and Technology SRM 610 and zircon standard GJ-1 were also measured twice every 20 analyses as external standards. Detailed analytical procedures follow (Liu et al., 2008). Off-line selection and integration of background and analyte signals, time-drift correction and quantitative calibration for trace element analyses and data reduction were performed using ICPMSDataCal (Liu et al., 2008). Concordia diagrams and weighted mean ages were calculated using Isoplot/Ex_ver3 (Ludwig, 2003).

5. $^{40}\text{Ar}/^{39}\text{Ar}$ dating

Glaucophane grains from blueschist sample (17QL27-1) were selected for $^{40}\text{Ar}/^{39}\text{Ar}$ dating. The sample was first crushed, then unaltered amphibole and phengite crystals with a diameter of 212-355 μm were hand-picked under a binocular microscope.

The preparation for $^{40}\text{Ar}/^{39}\text{Ar}$ analyses was undertaken at the Western Australian Argon Isotope Facility at Curtin University. The selected minerals were leached in diluted (5N) HF for one minute and then rinsed using distilled water in an ultrasonic bath. After rinsing, minerals were loaded into aluminium discs with Cd-shields to avoid undesirable nuclear interference reactions. Sample irradiation (40 hours) was conducted in the OSU TRIGA nuclear reactor (Oregon State University, USA) in the central position. The J-values calculated from the GA1550 standard grains in the surrounding pits are given in Table S4 and calculated using an age 99.738 ($\pm 0.10\%$) Ma and the decay constant of Renne et al. (2011). The correction factors for interfering isotopes were $(^{39}\text{Ar}/^{37}\text{Ar})_{\text{Ca}} = 6.95 \times 10^{-4}$ ($\pm 1.3\%$), $(^{36}\text{Ar}/^{37}\text{Ar})_{\text{Ca}} = 2.65 \times 10^{-4}$ ($\pm 0.84\%$) and $(^{40}\text{Ar}/^{39}\text{Ar})_{\text{K}} = 7.3 \times 10^{-4}$ ($\pm 12.4\%$). Mass discrimination was monitored regularly through the analysis using an automated air pipette relative to an air ratio of 298.56 ± 0.31 (Lee et al., 2006).

The $^{40}\text{Ar}/^{39}\text{Ar}$ analyses were performed at the Western Australian Argon Isotope Facility at Curtin University by step-heating the samples using a continuous 100 W

PhotonMachine© CO₂ (IR, 10.4 µm) laser fired on the crystals during 33 seconds. Each of the standard crystals was fused in a single step. The gas was purified in an extra low-volume stainless steel extraction line of 240cc, using two SAES AP10 and one GP50 getter. Ar isotopes were measured in static mode using a low volume (600 cc) ARGUS VI mass spectrometer from Thermofisher© ([Jourdan et al., 2019](#)) set with a permanent resolution of ~200. Measurements were carried out in multi-collection mode using four faraday cups to measure mass 40 to 37 and a low background compact discrete dynode ion counter to measure mass 36. We measured the relative abundance of each mass simultaneously using 10 cycles of peak-hopping and 33 seconds of integration time for each mass. Detectors were calibrated to each other electronically using Air shot beam signals.

The raw data were processed using the ArArCALC software ([Koppers, 2002](#)) and the ages were calculated using the decay constants recommended by [Renne et al. \(2011\)](#). All parameters and relative abundance values are provided in [Table S4](#) and have been corrected for blanks, mass discrimination and radioactive decay. Our criteria for the determination of plateau are as follows: plateaus must include at least 70% of ³⁹Ar and the plateau should be distributed over a minimum of 3 consecutive steps agreeing at 95% confidence level and satisfying a probability of fit (P) of at least 0.05. Plateau ages are given at the 2σ level and are calculated using the mean of all the plateau steps, each weighted by the inverse variance of their individual analytical error. Uncertainties include analytical and J-value errors.

PHASE EQUILIBRIUM MODELLING

Pseudosections for samples of blueschist (17QL27-1c), a glaucophane-bearing mafic schist (17QL25-1) from the NE NQOB were calculated in the Na₂O–CaO–K₂O–FeO–MgO–Al₂O₃–SiO₂–H₂O–TiO₂–Fe₂O₃ (NCKFMASHTO) system using THERMOCALC 3.45 and the internally consistent ds62 thermodynamic dataset (6 February 2012 updated version; ([Holland and Powell, 2011](#))). Activity–composition solution models include metabasite melt, clinopyroxene, amphibole ([Green et al., 2016](#)), biotite, garnet, ilmenite, chlorite and muscovite-paragonite ([White et al., 2014](#)),

epidote (Holland and Powell, 2011), and plagioclase (Holland and Powell, 2003). Quartz, rutile, titanite and H₂O are considered as pure end-member phases; and H₂O is considered to be in excess.

Bulk compositions used for calculations for sample 17QL27-1c were based on whole-rock XRF analyses, for which CaO content was reduced, based on P₂O₅ content to account for the presence of apatite; values of $X\text{Fe}^{3+}$ (atomic $\text{Fe}^{3+}/\Sigma\text{Fe}^{2+}$) were set to 0.2. The modelled bulk composition for sample 17QL25-1 was calculated based on mineral proportions and compositions analyzed by EPMA, following the method of Wei and Song (2008). The compositions (in mol.%) are listed in Table S2.

1. Blueschist sample 17QL27-1c

A P – T pseudosection for blueschist sample 17QL27-1c, calculated over the P – T range of 5–18 kbar and 410–530°C, is shown in Fig. S2. Glaucophane and chlorite occur in all assemblage fields. Epidote is present at pressures of 7–14.5 kbar. Phengite is only stable at $P > 7$ kbar. Biotite occupies a large stability field at $P < 16$ –18 kbar. Actinolite occurs at temperature $> 470^\circ\text{C}$ and pressure < 7 –12 kbar. Rutile is stable at temperature > 460 –480°C, whereas titanite is stable at temperature < 460 –490°C. Omphacite occurs at low temperature and pressure ($< 470^\circ\text{C}$, < 7 kbar) and at high pressure (> 7.5 –14 kbar). The interpreted peak assemblage of glaucophane–phengite–chlorite–titanite is predicted to be stable in the P – T field (outlined by the pink solid line in Fig. S2) ranging from ~ 7.5 kbar and 410°C to 17.5 kbar and 460°C. Within this assemblage field, compositional isopleths of Si in phengite show moderately positive slopes, with Si content increasing with P . The measured Si contents of 3.38–3.49 (c.p.f.u., based on 11 oxygen atoms) constrain maximum pressures to < 14.5 kbar. Thus, the interpreted peak P – T conditions are 7.5–14.5 kbar/410–490°C.

2. Mafic schist sample 17QL25-1

The pseudosection for the epidote schist sample (17QL25-1) was calculated in the range of 2–15 kbar and 350–600°C (Fig. S3). H₂O contents are estimated to be in excess. Epidote is stable in a large field except at pressures > 11 kbar at a temperature

of ~350–420°C and pressures <5.5 kbar at a temperature of ~507–600°C. Chlorite is stable at temperatures lower than ~466–560°C. Glaucophane occurs at pressures >6.2 kbar. The interpreted peak mineral association (albite–chlorite–epidote–glaucophane–phengite–titanite) is estimated in the *P–T* field of ~6.8–8.5 kbar/~350–475°C (the area is identified by the pink solid line in Fig. S3), which is constrained by rutile-in to the high temperature side and paragonite-in to the high pressure side.

GEOLOGICAL DATA

Zircon grains from the mafic schist matrix (18QL03) are characterized by broad oscillatory zoning, consistent with an igneous origin (Fig. S4A). Laser ablation-inductively-coupled plasma-mass spectrometer (LA-ICP-MS) and sensitive high-resolution ion microprobe (SHRIMP) U–Pb zircon analyses yield identical weighted mean $^{206}\text{Pb}/^{238}\text{U}$ ages of 488 ± 2 Ma (MSWD = 0.32) (Fig. S4B) and 488 ± 4 Ma (MSWD = 1.5, Fig. S4C; Table S3), respectively. LA-ICP-MS U–Pb analysis of 55 titanite grains from the blueschist (17QL27-1) define a discordant array with a lower intercept age of 492 ± 27 Ma (MSWD = 1.1) (Fig. S4D; Table S3). These ages are interpreted as the protolith age of the blueschist and meta-mafic schist matrix.

One analysis of phengite (18QL02) and two of glaucophane from the blueschist (17QL27-1, 18QL02) give robust $^{40}\text{Ar}/^{39}\text{Ar}$ ages. Phengite yields a plateau age of 454.8 ± 0.6 Ma (MSWD = 0.86, $\rho = 0.62$; Fig. S5A; Table S4), which is interpreted as the age of peak blueschist facies metamorphism. The glaucophane yields younger plateau ages of 432 ± 5 Ma (MSWD = 0.83, $\rho = 0.66$) and 425 ± 13 Ma (MSWD = 0.70, $\rho = 0.81$; Figs. S5B–S5C; Table S4) that are indistinguishable in age and which likely record cooling and exhumation.

EVALUATION of ELEMENT MOBILITY by ALTERATION and METAMORPHISM

As the studied rocks experienced greenschist-blueschist facies metamorphism, we first evaluate the effects of alteration and metamorphism before discussing the geochemical characteristics of the blueschist and mafic schist samples. Generally,

most large ion lithophile elements (LILEs; e.g., K, Rb, Ba, Sr) are mobile elements, while rare earth elements (REEs), high field strength elements (HFSEs; e.g., Zr, Hf) and transition metal elements (e.g., Cr, Ni, V) are relatively immobile (Polat et al., 2002; Pearce and Peate, 1995; Pearce et al., 1992; Winchester and Floyd, 1977; Pearce and Reagan, 2019), and can therefore be used to evaluate alteration and mobility of other elements according to their geochemical covariations. The blueschist and metabasite samples have relatively low LOI values (<4 wt. %), suggesting insignificant carbonization. Some samples have lower MgO and higher SiO₂ contents, which suggest that these high Si and low Mg blueschist samples probably experienced a certain degree of Mg loss and were metasomatized by variable amounts of silica. For some blueschist samples, thin quartz bands (~1–15 mm) and larger quartz lenses (~2–15 cm) are distributed parallel to the foliation, consistent with the incorporation of silica during metamorphism. Therefore, the concentrations of MgO, SiO₂ and other major elements in quartz-rich samples were likely modified during metamorphism. Some blueschist samples have relatively low LOI, SiO₂, and high MgO contents, suggesting they retained their original compositions. In the following major element based diagrams, we just plot the less-altered samples with low SiO₂, LOI, and high MgO contents.

In contrast, all blueschist and metabasite samples show coherent trace element patterns (REEs and HFSEs) without anomalies of Ce/Ce* (Polat et al., 2003), suggesting that the REEs and HFSEs of the blueschist and metabasite samples were not significantly modified. Therefore, we use these less mobile elements to fingerprint mantle source of the protolith of the blueschist and metabasite samples.

Figure S1. Field photographs and photomicrographs of representative rocks from the eastern North Qilian Orogenic Belt, North Tibet. (A) Blueschist block in mafic schist matrix. (B) Typical view of blueschist (17QL27-1). (C) Backscattered electron (BSE) images of blueschist (17QL27-1). (D) BSE images of glaucophane-bearing mafic schist matrix (18QL03). Gln–Glaucophane; Ph–Phengite; Ab–Albite; Ttn–Titanite; Qz–Quartz; Ap–Apatite

Figure S2. P – T pseudosection for blueschist sample (17QL27-1c) in the system NCKFMASHTO ($\text{H}_2\text{O}+\text{q}$) with bulk composition from Table S2. The pseudosection is contoured with isopleths of Si content in phengite.

Figure S3. P – T pseudosection for mafic schist matrix sample (17QL25-1) in the system NCKFMASHTO ($\text{H}_2\text{O}+\text{q}$) with bulk composition from Table S2.

Figure S4. (A) Representative zircon cathodoluminescence (CL) images. (B) Concordia diagram of zircon LA–ICP–MS dating of mafic schist matrix (18QL03) from eastern North Qilian Orogenic Belt. (C) Concordia diagram of zircon SHRIMP U–Pb dating of mafic schist matrix (18QL03). (D) Tera-Wasserburg concordia plot of titanite in blueschist (17QL27-1) from the eastern North Qilian Orogenic Belt.

Figure S5. $^{40}\text{Ar}/^{39}\text{Ar}$ apparent age of (A) phengite (18QL02) and (B–C) glaucophane separated from the blueschist (18QL02, 17QL27-1). MSWD—Mean square of weighted deviates; p —probability.

Figure S6. Geochemical classification diagram for blueschist and metabasites (including mafic schist matrix and greenschist) from the eastern NQOB. (A) Zr/TiO₂ versus Nb/Y diagram. (B) MgO versus SiO₂ diagram. (C) TiO₂ versus MgO diagram. (D) CaO/Al₂O₃ versus MgO diagram. Data sources: low-Si boninite, high Si boninite and HIMA (Bonin Island) (Li et al., 2019); Chichijima boninite (Taylor et al., 1994); Dachadaban tholeiite and boninite in the NQOB (Xia et al., 2012); Bonin Ridge FAB and boninite samples (Ishizuka et al., 2011); Mariana FAB (Reagan et al., 2010). Bonin Island arc volcanic rocks (Ishizuka et al., 2020). FAB—forearc basalt; HIMA—High-Mg andesite; BADR—Basalt-andesite-dacite-rhyolite.

Figure S7. Tectonic discriminant diagram and trace element diagram evaluating the slab-derived compositions for blueschist and metabasite (including mafic schist matrix and greenschist) samples from the eastern NQOB. (A) Ba/Th versus Th diagram. (B) Nb/N-MORB versus Th/N-MORB diagram (Saccani, 2015). Data sources for comparisons are same as in Figure S6.

Figure S8. Summary of peak P - T conditions and P - T paths of HP metamorphic rocks from northern and southern metamorphic belts in the North Qilian Orogenic Belt. The

subduction zone geotherms of 5°C/km, 10°C/km and 20°C/km are shown for reference. P – T conditions of representative samples from this study in the Laohushan area are labelled and the P – T path (pink line) is marked. The dark blue field shows the peak P – T conditions of eclogite in the Qilian area, central NQOB after Zhang et al. (2007), Wei et al. (2009) and the peak P – T conditions of the Sunnan blueschist after Song et al. (2009). The P – T paths for eclogitic rocks in the Qilian area are after Song et al. (2009) (green dashed line) and Zhang et al. (2007) (blue dashed line) and the P – T path for blueschists in the Sunan area are after Zhang et al. (2009) (orange dashed line). The P – T paths for chloritoid-glaucophane schist in the Qilian area are after Wei and Song (2008) (purple dashed line).

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