High Water Contents in Zircons Suggest Water-Fluxed Crustal Melting During Cratonic Destruction

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Abstract Water is essential for the formation of granites, but its origin and role in granite generation (i.e., dehydration vs. water-fluxed melting) remain uncertain. These issues are addressed by combining water abundances and other geochemical indices in zircons from Late Mesozoic granites generated during the destruction of the North China Craton (NCC). The water contents in zircons from the NCC Early Cretaceous granites (763 ppm, median) are much higher than those of the NCC Jurassic granites (424–513 ppm), upper mantle and continental arc magmas (92–477 ppm). More importantly, the higher water contents in the voluminous Early Cretaceous granites also have higher zircon saturation temperatures, εHf(t), and lower δ18O values. These observations suggest a predominantly mantle origin for the water, and water-fluxed crustal melting, in which larger water ingress produced more voluminous melts. The high-water flux was likely related to the subduction of the Paleo-Pacific Plate, which ultimately destabilized the NCC.

Plain Language Summary The fact that water is essential in generating granites has been known for a long time. However, its detailed role is poorly understood due to heterogeneous source and complex melting reactions involved in the generation of granites. As a fundamental issue of granite genesis, it remains a long-standing problem to distinguish the two major mechanisms, that is, hydrous-mineral-dehydration melting versus external-water-added melting. In this study, the water content of zircon combined with other lines of clues of I-type granites that generated during the destruction of North China Craton (NCC) in Late Mesozoic collectively points to water-added crustal melting rather than dehydration melting. The isotope composition of zircon suggests a mantle provenance of water. The highest water contents occurred in the Early Cretaceous granites, corresponding to the climax of the NCC destruction. Higher zircon water contents in Early Cretaceous granites indicate higher water-flux into the lithospheric mantle and overlying crust by the subduction of the paleo-Pacific plate. Accordingly, water played a significant role in cratonic destruction.

1. Introduction

Water is essential in the generation and evolution of granites (Campbell & Taylor, 1983; Collins et al., 2020) and usually is derived from one of two sources (Brown, 2013; Johannes & Holtz, 1996; Weinberg & Hasalová, 2015): (a) dehydration of hydrous minerals (e.g., amphibole and biotite) in the crust; and (b) ingress of external water to the magmatic system. In the latter case, the presence of free water lowers the crustal solidus and facilitates water-fluxed melting. Different crustal melting mechanisms have significant implications for reconstructions of the geodynamic evolution of a given region (Collins et al., 2020; Weinberg & Hasalová, 2015); but distinguishing methods between these mechanisms usually yield ambiguous results, probably because of the heterogeneous source nature of granites and the complex melting reactions involved in their generation (Bartoli, 2021; Schwindinger et al., 2019).

Given that dehydration melting requires hydrous phases to be present in the melting region, the extent of melting and volume of melts generated in intraplate setting are limited because the hydrous phases cannot be replenished (Brown, 2013; Weinberg & Hasalová, 2015). Dehydration melting can be considered a closed system, where water behaves incompatibly, and its abundance in melts depends on the degree of melting and water abundance in the source (Brown, 2013; Weinberg & Hasalová, 2015). In this case, a higher water content results from a smaller
degree of melting and generation of a smaller volume of granitic melt. The isotopic compositions of the granites produced reflect those of the crustal source (Weinberg & Hasalová, 2015). In contrast, if crustal melting is triggered by fluid ingression, the degree of melting can be large and voluminous melts will be generated (Weinberg & Hasalová, 2015). In this regard, direct measurement of water content in granites can provide more prominent insights into crustal melting processes.

The diffusion rate of H\textsubscript{2}O in zircon, a nominally anhydrous mineral (NAM), is slower than other NAMs (e.g., quartz, clinoproxene, garnet) (Ingrin & Zhang, 2016; Zhang, 2015) and zircon saturates early in felsic and non-peralkaline melts (Boehnke et al., 2013; Watson, 1979), which makes melts of this composition an ideal subject to study their initial water content. Zircon water content, in combination with other zircon tracers (e.g., zircon saturation temperatures T\textsubscript{Zr}, ε\textsubscript{Hf}(t) and δ\textsubscript{18}O), is a potentially powerful tool to constrain crustal melting mechanism, and crust-mantle interactions (Kemp et al., 2007; Miller et al., 2003; Valley et al., 1994). Here we describe an application of this new methodology to the North China Craton (NCC; Figure 1a), which lost its lithospheric root during the Late Mesozoic (Menzies et al., 1993; Xu, 2001). Cratonic destruction was associated with the emplacement of voluminous granites over an intraplate area of ∼70,000 km\textsuperscript{2}. Two giant granite sub-provinces of Jurassic (200–145 Ma) and Early Cretaceous (145–110 Ma) age have been identified (Wu et al., 2019), with the latter accounting for ∼75% of the entire province (Figure 1a). The zircons from 28 I-type granite samples of nine plutons in Liaodong Peninsula have been investigated in detail (Table S1 in Supporting Information S1), which were formed in three stages: Early Jurassic (172–178 Ma), Late Jurassic (154–163 Ma) and Early Cretaceous (119–125 Ma) (Figure 1b).

2. Results

We first screened zircons by Cathodoluminescence (CL) image and La concentration (<10 ppm) to preclude the possible hydrothermal alteration (Figures S1 and S2a in Supporting Information S1) (Hoskin, 2005). The data with transitional La concentration of 0.1–10 ppm (Hoskin, 2005; Zou et al., 2019) are retained because they are demonstrated to be the impacts of deep-sited micro-inclusions (Figure S2b in Supporting Information S1). SIMS pits are shallower and atop of the LA-ICP-MS craters, consequently, the measured water contents are not affected by deeper inclusions, as evidenced by the uncorrelated water contents and La concentrations (Figure S2c in Supporting Information S1). Additionally, the lattice of zircons can be damaged due to the radioactivity of U and Th, which allows absorption of secondary water (Nasdala et al., 2001). We carefully selected the least metamict zircons for water measurements following the restricted screening criterion established by Yang et al. (2022) (Figure S2d in Supporting Information S1 and Text S2 in Supporting Information S1). Of the 28 samples, a portion of zircons of 23 samples passed the filter (Table S1 in Supporting Information S1).
Eleven Early Cretaceous zircon samples have median water contents of 446–889 ppm (Figure S3 in Supporting Information S1) and only one sample (08JF95) has a significantly higher median water content of 1,323 ppm (Figure S3 in Supporting Information S1). In contrast, the median water contents of five Late Jurassic samples are lower and ranges from 162 to 512 ppm. The median water contents (677–801 ppm) of zircon in the remaining three Late Jurassic samples (11JF76, 13JF46, 13JF61) are comparable with those in the Early Cretaceous zircons (Figure S3 in Supporting Information S1). The water contents in the Early Jurassic zircons (360–687 ppm, medians) are transitional between the other two groups (Figure S3 in Supporting Information S1). Three different aged granite groups in this study are also distinguishable in terms of Hf-O isotopes of zircon (Table S1 in Supporting Information S1). The Late Jurassic granites show high δ^{18}O (7.1%–9.1‰) and low ε_{Hf}(t) (−24.2 to −28.1). The Early Cretaceous granites are characterized by lower δ^{18}O (5.5%–6.9‰) and relatively higher ε_{Hf}(t) (−6.1 to −22.1, Table S1 in Supporting Information S1). The Early Jurassic pluton shows transitional Hf-O isotope compositions between the above-mentioned groups; they overlap the Late Jurassic granites in ε_{Hf}(t) and the Early Cretaceous granites in δ^{18}O (Table S1 in Supporting Information S1).

3. Discussion

3.1. Factors Responsible for Water Variation in Zircon

Previous studies on substitution mechanisms in zircon have revealed that the hydrogen could be incorporated by buffering reaction (i.e., coupled substitution with trivalent cations). Water content in zircon may be controlled by the trace element concentrations, not a reflection of the melts (De Hoog et al., 2014; Trail et al., 2011). As shown in Figure S5b in Supporting Information S1, only small amount of water (<20% in most zircons) was incorporated into zircon through this coupled substitution mechanism (Text S3 in Supporting Information S1). The majority of water may enter zircon by hydrograssular substitution of the form of 4OH\(^-\)↔(SiO\(_4\))\(^{4-}\), which is directly related with water content in melt (Woodhead et al., 1991).

Recent studies on the zircons from Gangdese batholith delineate a positive correlation between zircon Hf concentration and water content (Xia et al., 2021) (Figure S7b in Supporting Information S1), indicating the variation of zircon water content is dominated by the water content in melt and that zircon, once crystallized, is not subjected to later evolved melts. Therefore the initial water content incorporated in zircon during crystallization can be preserved under the P-T conditions of magma chamber. This assessment is further supported by the distinct water contents between cores and rims of zircons from sample 13JF70 (Figure S1 in Supporting Information S1). We have also checked the relationship between grain sizes of zircons and their water contents. In general, Early Cretaceous zircons have higher water contents and larger grain sizes than Jurassic zircons (Figure S1 in Supporting Information S1). But Jurassic zircons with similar grain size (e.g., 13JF03, 13JF05, 13JF29) to Early Cretaceous zircons still have conspicuously lower water contents (Figure S1 in Supporting Information S1). These observations suggest that measured water contents are irrespective of grain size and the initial water in zircon has been well preserved.

Compared with highly evolved granites that generally contain >1.2 wt.% Hf concentrations and <0.05 Eu/Eu* (chondrite normalized Eu/√Sm×Gd) (Breiter et al., 2014; Deering et al., 2016; Wang et al., 2010; Xia et al., 2021), relatively lower Hf concentrations (<1.2 wt.%) and higher Eu/Eu* (>0.05) of zircons in this study suggest these zircons crystallized in the less evolved granites (Figure S7 in Supporting Information S1). Thus, zircons most likely record water contents in early magmas. The lack of correlation between whole rock SiO\(_2\) contents and zircon water contents argues against a significant influence of magmatic differentiation on zircon water contents (Figure 2a). Although experimentally determined partition coefficients for zircon are not available, it can be reasonably assumed to be negatively and positively correlated with temperature and pressure respectively, similar to other NAMs (Keppler, 2006; O'Leary et al., 2010). The broadly negative correlation between Sr/Y ratio (a proxy of the depth of magma origin) (Chapman et al., 2015) and water contents (Figure 2b) is the opposite to that expected if a pressure effect was significant.

The temperature effect is apparent for the Late Jurassic granites whose water contents in zircon negatively correlate with T\(_{2z}\) (Figure 2c), as expected. The decreasing temperature would not only increase the zircon/melt partition coefficients for water but also result in a high solid/melt ratio, both of which would increase zircon water contents. That the highest water contents were measured in the samples (11JF76 and 13JF61) with the lowest T\(_{2z}\) (<730°C; Figure 2c) points to the temperature effect. Consequently, these two samples are not considered
in further discussion. The temperature effect does not apply to the Early Jurassic and Early Cretaceous granites either, given lack of obvious correlation between TZr and H2O (Figure 2c). Comparison of water contents between different samples is only feasible over a similar temperature range. Over a comparable TZr (750°C–850°C), the Early Cretaceous zircons have higher water contents (median of 763 ppm) than the Late Jurassic zircons (424 ppm), with the Early Jurassic granites (513 ppm) being transitional (Figure 2d).

3.2. Comparison With Zircons From Other Tectonic Environments

The water contents of zircon in Late Mesozoic granites in NCC are comparable to or much higher than those of zircons from other tectonic settings (Figure 2d). Two in-house zircon standards, that is, Penglai and Qinghu zircons, are interpreted to be generated in the intraplate mantle derived melts. The low water contents (medians of 92–259 ppm) characterize that of the intraplate mantle source. Zircons from subduction-related rocks tend to have relatively higher water contents (medians of 250–477 ppm; Figure 2d), reflecting slab to mantle wedge transfer of fluids/melts released by the downgoing plate. In comparison, the zircons from the Early Cretaceous granites of the NCC are conspicuously higher in water contents with a median of 763 ppm than that from any other tectonic settings investigated here (Figure 2d).
3.3. Evidence for Water-Fluxed Crustal Melting

The crustal thickness of the NCC was estimated >30 km in the Jurassic, but <30 km in the Early Cretaceous using the method of Chapman et al. (2015). When the adiabatic ascent effect from 8 to 2 kbar (∼50°C) is taken into account (Johannes & Holtz, 1996), the inferred melting temperatures of Jurassic and early Cretaceous granites are ∼790°C–850°C and ∼820°C–900°C, respectively. These estimates are lower than the dry solidus of granites and the dehydration melting temperatures of amphibole (Figure 3). This, together with the general absence of hydrous minerals in the lower crust beneath the NCC (<10 wt.% amphibole + biotite, i.e., <∼0.3 wt.% water) (Zhai et al., 2001), precludes dehydration melting being the main mechanism to form Late Mesozoic granites.

Another message from Figure 3 is that the minimum water abundances, required to intercept the liquidus, are ∼4–5 wt.% and ∼2–3 wt.% for the Jurassic and Early Cretaceous granites, respectively. As a consequence, conditions were more favorable for crustal melting during Early Cretaceous than Jurassic. The higher water contents in the Early Cretaceous granites compared to Jurassic suites suggests that the granites were not generated under such minimum water content melting conditions. More likely, crustal melting was triggered by excess water ingression. Further supports for water-fluxed melting come from the positive correlations between water content in zircon and the volume of granites (Figure 1). Higher water flux in the Early Cretaceous granite source resulted in a larger degree of melting and the generation of more voluminous granites.

3.4. A Mantle Origin of Water in the NCC Granites

The Hf-O isotope correlation indicates more mantle-derived material in the source of the Early Cretaceous granites than in the Jurassic granites (Figure 4a). This is consistent with the more depleted whole-rock Sr-Nd isotope compositions in the Early Cretaceous granites compared to the Jurassic granites (Wu et al., 2005; Yang et al., 2004). The higher water contents and εHf(t) in zircons from the former (Figure 4b) further suggests a dominant mantle origin for the water in the studied granites. This is surprising but is understandable in the context of regional geology.

The Late Mesozoic sub-continental lithospheric mantle (SCLM) beneath the NCC was unusually enriched in water (up to >1,000 ppm) (Figure S8a in Supporting Information S1) (Xia et al., 2013) compared to ∼120 ppm for the typical upper mantle (Bell & Rossman, 1992). It is possible that, in the NCC case, melting of the SCLM was induced by infiltration of hydrous fluids/melts, followed by melting of the crust due to fluxing by underplating of SCLM-derived hydrous mafic melts like Feixian basalts (Xia et al., 2013). Calculation consolidates that dry SCLM and lower continental crust (dehydration melting) cannot generate water-rich granites under such a two-stage melting, but the hydrous SCLM could (Figure S8 in Supporting Information S1). For instance, the water contents of lower crust of in Early Cretaceous and Jurassic increase to 4.78 wt.% and 2.08 wt.% respectively by water fluxing from the mantle, assuming 70% and 30% mantle-derived water-rich fluid/melt is added to the lower crust (Text S4 in Supporting Information S1). In consideration of a smaller degree of partial melting may result in a higher water content in melt, our calculation suggests that higher water ingestion was responsible for the high-water content in Early Cretaceous granites (Figure S8b in Supporting Information S1).

3.5. Lithospheric Hydration and Cratonic Destruction

If the Late Mesozoic NCC granites were formed by water-fluxed melting, it requires a substantial water supply to ensure the availability of water over a period of 60–70 Ma. Such a high-water flux was made possible by subduction of the Paleo-Pacific Plate, which started at least ca. 180 Ma (Ma and Xu, 2021), and is the only known process on Earth that can transport significant amounts of water into deep Earth (Zheng, 2009). The coherent variation between the volume of these two episodes of magmatism, and their zircon water contents (Figure 1b), point to the increasing influence of the subduction of the western Pacific Plate on the SCLM beneath east Asia.
During the Jurassic, the Paleo-Pacific Plate subducted westward at a low-angle beneath east Asia, which expelled the asthenospheric mantle between the lithospheric mantle and subducted slab (Figure 5a) (Wu et al., 2019) and the cold slab directly underlay the bottom of the SCLM. The associated cooling effect (Dumitru et al., 1991; Kusky et al., 2014) limited the transfer of material from slab to the SCLM, and resulted in a smaller water flux to the crust (Figure 5a), and a predominant crustal signature and relatively small volume of Jurassic granites (Figures 1 and 4). From the Late Jurassic to Early Cretaceous, the retreated Pacific Plate steepened subduction angle (Figure 5b) (Wu et al., 2019), the space left by was filled by the convecting asthenosphere. The SCLM and the slab were heated, resulting in the release of a large amount of water from the slab and extensive hydration of the SCLM (Figure 5b) (Wu et al., 2019). As a consequence, voluminous Early Cretaceous granites with a significant mantle signature were produced due to a high-water flux (Figures 1 and 4).

The cratonic lithosphere is compositionally refractory, buoyant, and can be stable for billions of years. However, its strength is considerably weakened in the presence of water (Hirth & Kohlstedt, 1996), leading to thinning and refertilization of the lithosphere (Niu, 2005). The water-fluxed crustal melting model proposed here involves hydration of the SCLM and overlying deep crust beneath the NCC during the Late Mesozoic, emphasizing the role of water in the destruction of the NCC.

4. Conclusions

The water content in zircon, combined with other geochemical indices, provides new insights into the formation of late Mesozoic I-type granites from the NCC. The water abundances in the Liaodong granites are significantly higher than (Early Cretaceous) or similar to (Jurassic) those of intraplate mantle-derived magmas and continental arc granitic magmas. The highest water contents and more voluminous of the Early Cretaceous granites corresponding to the climax of the North China Craton destruction indicate higher water-flux into the lithospheric mantle and overlying crust by the subduction of the paleo-Pacific plate. Accordingly, this study thus provides evidence for the role of water in the cratonic destruction.
Data Availability Statement

The supporting informations and the analyses data of SIMS water content and oxygen isotope composition, LA-ICP-MS Hf isotope and trace element file for this paper are available at https://zenodo.org/record/5912119#.YfNV8PtBzIV (http://doi.org/10.5281/zenodo.5912119).

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