High water content in primitive continental flood basalts

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As the main constituent of large igneous provinces, the generation of continental flood basalts (CFB) that are characterized by huge eruption volume (>10^5 km^3) within short time span (<1–3 Ma) is in principle caused by an abnormally high temperature, extended decompression, a certain amount of mafic source rocks (e.g., pyroxenite), or an elevated H2O content in the mantle source. These four factors are not mutually exclusive. There are growing evidences for high temperature, decompression and mafic source rocks, albeit with hot debate. However, there is currently no convincing evidence of high water content in the source of CFB. We retrieved the initial H2O content of the primitive CFB in the early Permian Tarim large igneous province (NW China), using the H2O content of ten early-formed clinopyroxene (cpx) crystals that recorded the composition of the primitive Tarim basaltic melts and the partition coefficient of H2O between cpx and basaltic melt. The arc-like H2O content (4.82 ± 1.00 wt.%) provides the first clear evidence that H2O plays an important role in the generation of CFB.

As the main constituent of continental large igneous provinces (LIPs)1, continental flood basalts (CFB) are characterized by huge eruptive volumes within a relatively short time span. The estimated eruptive basalt volumes range from ~2 × 10^5 km^3 for the Columbia River Basalts to ~2 × 10^6 km^3 for the Siberian Traps2,3. The time span is usually as short as <1–3 My4–8. These features imply a special geodynamic process in the mantle and may trigger prominent environmental effects (climate change, mass extinction, etc.) and contribute to the formation of giant metal ore deposits9.

In principle, the generation of CFB requires an abnormally high temperature, extended decompression, a certain amount of mafic rocks in the mantle source, or the addition of H2O and/or CO2 into the mantle source10,11. These four factors are not mutually exclusive, and it is likely that several or all factors contribute together to generate CFB. The elevated H2O and/or CO2 content allows melting to start in the deeper mantle and enlarges the whole melting regime, consequently contributing to the enormous melt. The CO2 content is much less than the H2O content in the mantle12, and the magnitude of the lowering solidus of the upper mantle by CO2 is less than that of H2O13. Therefore, adding H2O is expected to be more important for the genesis of CFB. High temperature, decreased pressure and mafic source lithology have been extensively discussed, albeit debated, for three decades14–19, but the evidence of high H2O content is scarce.

Indeed, there were attempts to obtain H2O content of mineral-hosted melt inclusions in CFB, but the extent to which they can reflect the initial H2O content of primitive basaltic magmas (i.e. the magmas that after being extracted from their source regions have experienced little modification) was controversial. Stefano et al.20 and Cabato et al.21 measured melt inclusions hosted by olivine phenocrysts in the CFB of the Yellowstone hotspot track and the Columbia River, respectively. They found that the H2O content in melt inclusions is as high as 2.4 wt.% for the Yellowstone and 3.3 wt.% for the Columbia River. However, they also found that the melt inclusions with the highest H2O content are not hosted by the earliest-formed (i.e., with highest Fo value) olivine phenocrysts, so they may represent the H2O content of the evolved melts rather than that of the initial ones. Ten melt inclusions in olivine phenocrysts from the Siberian Traps basalts have H2O contents ranging from 0.01 wt.% to 1.6 wt.%22,23, almost falling in the range of mid-ocean ridge basalts (MORB, ~0.1–0.3 wt.%)24–29 and ocean island basalts (OIB, 0.3–1.0 wt.%29–33). However, the possibility of loss of H2O due to late-stage degassing processes was not evaluated for the Siberian melt inclusions.

In addition, Michael et al.34 and Wallace et al.35 analysed basaltic glasses of the Ontong Java and Kerguelen oceanic plateaus (the oceanic counterpart of CFB), respectively. The H2O content in these glasses ranges from

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0.13 to 0.49 wt.% for the Ontong Java and from 0.24 to 0.69 wt.% for the Kerguelen oceanic plateaus and is only slightly higher than that of MORB. However, the low Mg# (Mg/(Mg + Fe) mol.) of 40–60 indicated that the basaltic glasses they analysed are evolved melts, again arguing against the representativeness of the initial melts. Overall, there is currently no unarguable evidence to show whether the generation of CFB is related to the high H2O content of the mantle source.

Here, we calculate the initial H2O content of the early Permian Tarim CFB in NW China (>200,000 km² flood basalts) using the H2O content of clinopyroxene (cpx) macrocrysts crystallized from the primitive Tarim flood basalts and the H2O partition coefficient between cpx and basaltic melt. The inferred high H2O content in the initial basaltic melt provides the first firm evidence that H2O plays an important role in the generation of CFB.

Samples and previous study

Many cpx macrocrysts (1–15 mm of grain size) were hosted by one basaltic dyke that crosscuts to the Early Permian (~280 Ma) Xiaohaizi wehrlite intrusion in the Tarim large igneous province, NW China (Fig. 1a,b). They are fresh and usually prismatic and sub- to euhedral shapes (Fig. 1c), and they commonly have a high-Mg (Mg# = 80–89) core and a thin low-Mg rim (Mg# down to 70) that is resulted from the interaction with the host basalt. Wei et al. carried out a detailed geochemical analysis on these macrocrysts. These cpx generally have low TiO₂ (0.26–1.09 wt.%), Al₂O₃ (1.15–3.10 wt.%) and Na₂O (0.16–0.37 wt.%), so they are not likely to be xenocrysts from mantle peridotites. They are not in chemical equilibrium with the host basaltic dyke, arguing against a phenocryst genesis. In addition, the cpx macrocrysts define a coherent compositional trend (e.g., negative correlations between Mg# and Ti, Al, Na, La, Nd, Yb) with the cpx from the wehrlites crosscut by the basaltic dyke hosting the cpx macrocrysts, and these cpx have identical trace element distribution patterns, demonstrating a comagmatic origin. Accordingly, these
which is within the range reported by Wei37. The cpx Mg# values are 85.2 to 87.8 (Table 1), corresponding to a calculated H2O contents of the equilibrated basaltic melts are 3.69 wt.% to 6.61 wt.% (Average: 4.82 wt.%).

Mg# of ~70 for the equilibrated basaltic melts using the experimental Mg-Fe partition coefficient (0.34 ± 0.04)38. Cpx macrocrysts were formed from a nearly primary basaltic melt. Although an assimilation and fractional crystallization process may operate during the formation of the Xiaohaizi intrusion that was evidenced by higher 87Sr/86Sr and lower εNd (0.7035–0.7037, and εNd = 4.5–4.8) suggests that the cores of these cpx macrocrysts may have recorded the composition of the primitive Tarim basaltic melts, with little crustal contamination37. The Cpx macrocrysts in this paper were from the same dyke studied by Wei et al.37.

### Table 1. Chemical composition and H2O content of the Tarim clinopyroxenes and H2O content of the corresponding basaltic melts. Mg# = 100Mg/(Mg + Fe), 40Al and Ca are atomic numbers calculated based on 6 oxygen atoms. D(cpx/melt) is calculated by the equation 10 in O’Leary et al.40. Cpx H2O is measured by FTIR, melt H2O = Cpx H2O/D(cpx/melt).

Table 1 displays the chemical composition and H2O content of the Tarim clinopyroxenes and the corresponding basaltic melts. The Cpx H2O content ranges from 300 to 550 ppm (Average: 384 ppm), and the calculated H2O content of the melts equilibrated with the analysed cpx to represent the H2O content of the initial and primitive Tarim basaltic melt. Although bearing an uncertainty of up to 40%, such an H2O content is apparently higher than those of MORB, OIB and back-arc basin basalts (BABB, 0.2–2.0 wt.%)38–41 and falls in the range of island arc basalts (IAB, 2.0–8.0 wt.%)36–40 (Fig. 2).

| Sample | xu05-01 | xu05-02 | xu05-03 | xu06-01 | xu06-02 | xu06-03 | xu06-04 | xu06-07 | xu06-09 | xu06-10 | Average | 1 SD |
|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|---------|------|
| wt.%    |         |         |         |         |         |         |         |         |         |         |         |      |
| SiO2    | 53.76   | 53.43   | 53.07   | 53.81   | 53.94   | 53.63   | 53.79   | 53.87   | 54.16   | 53.58   |         |      |
| TiO2    | 0.41    | 0.39    | 0.58    | 0.43    | 0.55    | 0.52    | 0.47    | 0.59    | 0.48    | 0.66    |         |      |
| Al2O3   | 1.33    | 1.49    | 1.70    | 1.23    | 1.43    | 1.53    | 1.51    | 1.59    | 1.55    | 1.77    |         |      |
| Cr2O3   | 0.36    | 0.44    | 0.32    | 0.52    | 0.24    | 0.68    | 0.26    | 0.33    | 0.33    | 0.24    |         |      |
| FeO     | 4.88    | 4.41    | 5.19    | 4.31    | 4.75    | 4.32    | 4.54    | 4.95    | 4.66    | 5.12    |         |      |
| NiO     | 0.018   | 0.08    | 0.03    | 0.011   | 0.005   | 0.061   | 0.031   | 0.037   | 0.029   | 0.065   |         |      |
| MnO     | 0.079   | 0.08    | 0.09    | 0.082   | 0.068   | 0.048   | 0.067   | 0.084   | 0.079   | 0.062   |         |      |
| MgO     | 16.98   | 17.43   | 16.69   | 17.44   | 17.10   | 17.03   | 16.54   | 16.78   | 17.05   | 16.64   |         |      |
| CaO     | 21.84   | 21.51   | 21.98   | 21.79   | 21.91   | 21.78   | 21.63   | 21.71   | 21.98   | 21.89   |         |      |
| Na2O    | 0.15    | 0.18    | 0.19    | 0.20    | 0.21    | 0.26    | 0.23    | 0.19    | 0.19    | 0.22    |         |      |
| K2O     | 0.00    | 0.00    | 0.01    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.00    | 0.01    |         |      |
| Total   | 99.79   | 99.44   | 99.84   | 99.83   | 100.20  | 99.86   | 99.07   | 100.13  | 100.49  | 100.26  |         |      |
| Mg#     | 86.1    | 87.6    | 85.2    | 87.8    | 86.5    | 87.5    | 86.7    | 85.8    | 86.7    | 85.3    |         |      |
| 40Al    | 0.029   | 0.037   | 0.048   | 0.032   | 0.032   | 0.037   | 0.019   | 0.032   | 0.031   | 0.041   |         |      |
| Ca      | 0.858   | 0.847   | 0.866   | 0.854   | 0.856   | 0.854   | 0.853   | 0.850   | 0.856   | 0.857   |         |      |
| D(cpx/melt) | 0.0077 | 0.0082 | 0.0086 | 0.0079 | 0.0079 | 0.0081 | 0.0072 | 0.0079 | 0.0078 | 0.0083 |         |      |
| Cpx H2O (ppm) | 380 | 310 | 380 | 385 | 300 | 300 | 350 | 500 | 380 | 550 | 384 | 83 |
| melt H2O(wt.%) | 4.94 | 3.78 | 4.40 | 4.89 | 3.82 | 3.69 | 4.84 | 6.32 | 4.89 | 6.61 | 4.82 | 1.00 |

### Results

The chemical composition and H2O content in 10 cpx grains were obtained by an electron probe micro-analyzer (EPMA) and a Fourier transform infrared spectrometer (FTIR), respectively (see Methods). Wei et al.37 have shown that the rims of the Tarim cpx macrocrysts may have reacted with the host basalt, so only the clean core area of each cpx grain was measured here, in order to retrieve the information about the initial and primitive basaltic melts. 4–6 clean analysed spots in the core area were selected to run EPMA and FTIR for each grain (Fig. 1c), and in individual grains they show same chemical compositions and IR spectra (Fig. 1d). The average values of the analysed spots of each grain were, therefore, used to represent the element and H2O contents of that grain. Ten cpx grains have TiO2 (0.39–0.66 wt.%), Al2O3 (1.23–1.77 wt.%) and Na2O (0.15–0.26 wt.%) (Table 1), which is within the range reported by Wei37. The cpx Mg# values are 85.2 to 87.8 (Table 1), corresponding to a Mg# of ~70 for the equilibrated basaltic melts using the experimental Mg-Fe partition coefficient (0.34 ± 0.04)38. This suggests that the analysed cpx grains were crystallized from a nearly primary basaltic source39, in agreement with the trace element and Sr-Nd isotope characteristics of the Tarim cpx macrocrysts37.

The IR absorption spectra of the Tarim cpx can be subdivided into four groups, namely: 3630–3620 cm−1, 3540–3520 cm−1, 3470–3450 cm−1 and 3360–3350 cm−1 (Fig. 1d). The band at 3630 ~ 3620 cm−1 occurs in few grains, consistent with the structural OH bands in the cpx phenocrysts in Mesozoic-Cenozoic basalts of eastern China40–42. The calculated H2O contents of 10 cpx grains are 300–550 wt. ppm (Average: 384 ± 83 wt. ppm), and the calculated H2O contents of the equilibrated basaltic melts are 3.69 wt.% to 6.61 wt.% (Average: 4.82 ± 1.00 wt.%).
The arc-like H₂O contents in the early Permian Tarim primary basaltic melts indicate an addition of water from subduction-related processes. In the mid-Proterozoic, the Tarim was surrounded by subduction zones. In addition, ophiolite mélanges and arc-like magmatic events along the northern margin of the Tarim were dated at 600–418 Ma and 422–363 Ma, respectively, suggesting an active convergent margin. Experimental and natural investigations have demonstrated that minerals (cpx, garnet, olivine, etc.) in dehydrated plates can carry at least several thousands ppm (wt.) of H₂O into the Earth’s mantle. Garnets and omphacites from ultra-high pressure metamorphic eclogites have also been shown containing ~2000–3000 ppm wt. H₂O. If we consider that (1) the partition coefficient of H₂O between the mantle rock (peridotite, eclogite, pyroxenite) and melt is ~0.01 and (2) the degree of partial melting of the Tarim basalts is < 10%, then ~5000 ppm wt. H₂O in the source can produce 5% H₂O in basaltic melts, regardless of the melting model (batch or fractional) involved.

The upper mantle can accommodate several hundred ppm (wt.) of H₂O, and the lower mantle contains much less. Only the mantle transition zone (MTZ) can contain up to >1 wt.% H₂O. Several thousands ppm (wt.) of H₂O in the source of the Tarim basalts is, therefore, likely from the MTZ where the subducted plates stagnated and provided water. If so, the classic core-mantle boundary-derived plume model cannot be applied to the Tarim large igneous province.

In conclusion, the high water content in the primary early Permian Tarim basalts provides clear evidence that water, in addition to the temperature, pressure and source lithology, plays an important role in the generation of continental flood basalts. Furthermore, when high water content is considered, abnormally high temperature and extended decompression that are two critical factors in the widely accepted mantle plume model are not always to be prerequisites in the generation of CFB (and LIPs).

Methods
The H₂O content of cpx was determined with a Nicolet iso50 FTIR coupled with a Continuum microscope in School of Earth Sciences, Zhejiang University, following the unpolarized method described in Xia et al. For each cpx grain, several analysed spots (~20 μm × 20 μm) were set in the clean core area and they display almost same spectra, therefore the average spectrum was used to calculate the H₂O content of that grain. The modified Beer-Lambert law \( I = \frac{A}{I(t)} \) was used to calculate to H₂O content, in which \( I \) is the integral specific absorption coefficient (7.09 ppm \( \times \) cm \( ^{-2} \)) and \( t \) is the thickness (cm). The uncertainty of H₂O content is less than 30%.

The major element contents of cpx were analysed using a Shimadzu EPMA 1600 at University of Science and Technology of China. The 15 kV accelerating voltage, 20 nA beam current and 1 μm beam diameter were used. Standards are natural minerals and synthetic oxides. Data correction was obtained by a program based on the ZAF procedure. The reproducibility is <1% for elements with concentration >5% and <3% for elements with concentration >1%. The analysed points were set within the FTIR analysed area. Several points in each cpx grain have homogeneous element contents, and the average values were used (Table 1).

The H₂O content of the basaltic melts equilibrated with cpx is estimated by the H₂O content of cpx and the H₂O partition coefficients (Dcpx/melt) between cpx and melt. Dcpx/melt can be calculated by the equation 10 in O’Leary et al. \( D = \exp(-4.2 + 6.5 \times X(Al) - X(Ca)) \), where X(Al) and X(Ca) are the concentration of octahedrally coordinated Al³⁺ in tetrahedral site and Ca²⁺ in cpx calculated on the basis of 6 oxygen. This equation was derived by compiling experimental results run at temperatures between 1025 °C and 1440 °C, pressures between 0.5–5.0 GPa, melt H₂O contents between 1.09 wt.% and 24.9 wt.%, and cpx Al between 0.002 and 0.306. Considering the uncertainties from Dcpx/melt (~10%) and H₂O content in cpx (<30%), the total uncertainty of H₂O contents in melts is estimated to be less than 40%.
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Author Contributions

Q.-K.X. had the idea for the study and Y.B. and P.L. carried out all analyses. All authors contributed to the interpretation of the data and Q.-K.X. took the lead in preparing the manuscript with input from Y.B., P.L., W.T., X.W. and H.-L.C.

Additional Information

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