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Water storage in the refractory lithospheric mantle

B.-X. Su[1,2*], W.-F. Zhang[2,3*], Q.-Q. Pan[1], Y. Bai[4], M.-M. Cui[1], X. Liu[1], I. Uysal[5], X.-P. Xia[6], X.-Y. Gu[7]

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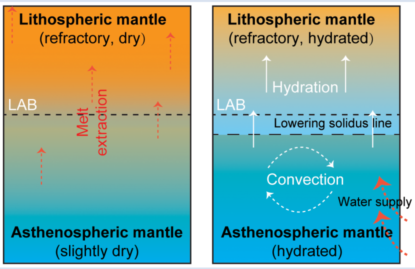
Abstract



This study reveals that ophiolitic olivine contains water, ranging from several to 170 μg.g^{-1}, contradicting expectations of a depleted refractory mantle. This range is comparable to xenolithic olivine from the continental lithospheric mantle. Both types of olivine show a positive correlation between water content and forsterite numbers, a typical proxy for refractory origin, which differs from patterns observed during partial melting or magma differentiation. Our findings indicate that water content in lithospheric mantle minerals may be primarily influenced by hydration after melt extraction rather than the extraction process itself. Thermodynamic factors (temperature, pressure, redox environment) and reduced competition from coexisting phases (*e.g.*, pyroxenes) enable olivine in refractory mantle to incorporate water. This hydration of olivine lowers the melting solidus temperature, promoting recurrent mantle melting and enhancing the refractory nature of mantle residues. The hydrated refractory mantle, sustained by deep mantle water or recycled materials, acts as a significant water reservoir. The interplay between melting and hydration plays a critical role in forming a thick lithospheric mantle.

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Introduction

Hydrogen, primarily present as OH within the Earth's mantle, is a volatile and highly incompatible element, predominantly extracted from the mantle source during partial melting (Williams and Hemley, 2001). It is assumed to be depleted in mantle minerals following melt extraction and in early crystallising phases (*e.g.*, olivine) of mantle-derived magmas. Olivine, comprising over 60 % by volume of the upper mantle, tends to be retained in the mantle during partial melting, and its forsterite (Fo) content serves as a refractory index. Recent advancements in analytical techniques have enabled precise determination of hydrogen abundance in olivine (*e.g.*, Peslier *et al.*, 2010; Xia *et al.*, 2019). Available data reveal that hydrogen significantly influences mantle rheology and thermal conductivity (Dixon *et al.*, 2004; Li *et al.*, 2008; Demouchy and Bolfan-Casanova, 2016; Chang *et al.*, 2017). However, the relationship between water storage capacity and compositional variations in refractory mantle olivine remains poorly constrained.

Ophiolites, fragments of ancient oceanic lithosphere, predominantly originate in the supra-subduction zone. Their mantle sequences undergo high degree partial melting, resulting in refractory compositions. These compositions generally remain uncontaminated by subsequent melt/fluid modification, which often introduce volatile components released from dehydrating slabs (Su *et al.*, 2023). The fate of subduction-derived volatiles, including water, remains enigmatic (Peacock and Hyndman, 1999) due to the scarcity of hydrous minerals in ophiolites.

In this study, we measured hydrogen abundance in olivine from 40 samples of the Bursa and Lycian ophiolites (90 Ma in age) in western Türkiye. These ophiolites lie within the Cretaceous Neo-Tethyan suture zone (Fig. 1a) and preserve extensive mantle peridotite massifs alongside overlying crustal units (Fig. 1b, c). The mantle peridotites are predominantly harzburgites hosting chromite deposits, with dunites occurring as lens/dykes or associated with chromitites (Pan *et al.*, 2022; Su *et al.*, 2023). Mineral assemblages of the three rock types include olivine and chromite, with additional orthopyroxene and clinopyroxene in harzburgites (Fig. 1d–f). Pristine samples with no or rare alteration features, comprising 4 harzburgites, 16 dunites and 20 chromitites, were selected for olivine separation and analyses (see Supplementary Information for detailed analytical methods).

Results

The analysed olivine grains exhibit Fo contents of 90 to 96 and water contents of 5–169 μg.g^{-1} (Table S-1), with values

1. State Key Laboratory of Lithospheric and Environmental Coevolution, Institute of Geology and Geophysics, Chinese Academy of Sciences, Beijing 100029, China
2. University of Chinese Academy of Sciences, Beijing 100049, China
3. State Key Laboratory of Isotope Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China
4. College of Geological and Surveying Engineering, Taiyuan University of Technology, Taiyuan 030024, China
5. Department of Geological Engineering, Karadeniz Technical University, 61080-Trabzon, Türkiye
6. YU-CUGW Joint Research Center on the Coupling between Earth Deep Processes and Shallow System, College of Resources and Environment, Yangtze University, Wuhan 430100, China
7. School of Earth Sciences, Zhejiang University, Hangzhou 310058, China
* Corresponding authors: B.-X. Su and W.-F. Zhang (emails: subenxun@mail.igcas.ac.cn; wfzhang@gig.ac.cn)

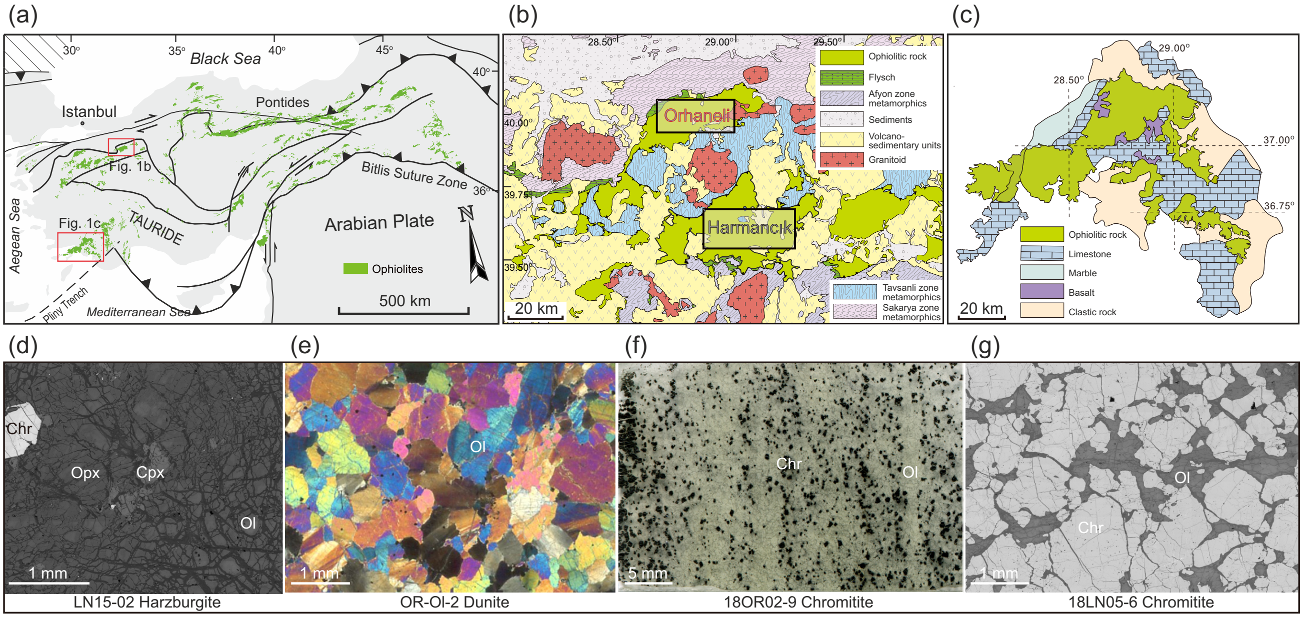

Figure 1 Distribution of ophiolites in Türkiye and locations of Bursa and Lycian ophiolites **(a)**; simplified geological maps of Bursa **(b)** and Lycian **(c)** ophiolites. Microphotographs of representive samples **(d–g)**. Back-scattered electron image of harzburgite from Lycian ophiolite showing mineral assemblage of olivine, orthopyroxene, clinopyroxene and chromite **(d)**. Crossed polarised image of dunite **(e)** and scanned image of chromitite **(f)** from Bursa ophiolite showing occurrence of olivine and chromite. Back-scattered electron image of pristine olivine occurrence in chromitite from Lycian ophiolite **(g)**.

increasing from harzburgites, to dunites and chromitite (Fig. 2). A strong covariation between Fo and water contents aligns with data from ophiolites and oceanic lithospheric massifs (Fig. 2). This trend contrasts with patterns expected from mantle peridotite melting, where water content should decrease with increasing Fo number (Fig. 3a). Some dunites and chromitites formed as magmatic cumulates within lithospheric mantle magma conduits or chambers (Su *et al.*, 2023). Their Fo and water contents of olivine exhibit a positive correlation, opposing the negative trend observed during magma differentiation (Fig. 3a), as exemplified by mafic–ultramafic intrusions (Tang *et al.*, 2022, 2023; Bai *et al.*, 2024).

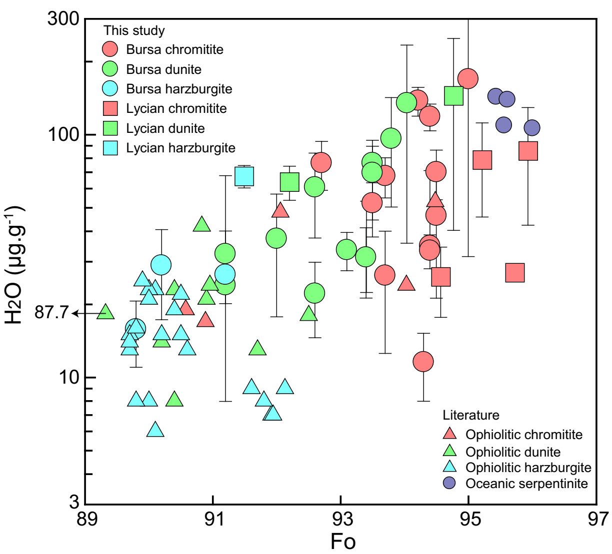

Figure 2 Correlation diagram of forsterite (Fo) *vs.* H_2O (μg.g^{-1}) contents in olivine from Bursa and Lycian ophiolitic chromitites, dunites and harzburgites. Literature data of ophiolites and Unterer Theodulgletscher oceanic lithospheric massif (see data sources in Supplementary Information) are plotted for comparison. Error bars of some analytical data are smaller than the symbols.

Discussion

Xenolithic olivine from the continental lithospheric mantle has been extensively studied (Wang, 2010). Its water contents (1 to 200 μg.g^{-1}) correlate positively with Fo (mostly 88–95), mirroring the trend observed in ophiolitic olivine (Fig. 3b). Olivine from deeper garnet-facies and diamond-facies sources generally exhibits higher water and Fo contents than its spinel-facies counterparts, consistent with pressure-dependent water storage (Peslier and Luhr, 2006; Hirschmann and Kohlstedt, 2012; Demouchy and Bolfan-Casanova, 2016). Ophiolitic olivine (spinel-facies) spans water contents equivalent to garnet-facies xenolithic olivine (Fig. 3b). Xenolithic olivine from diverse tectonic settings (craton, off-craton and oceanic) and ages (Archean to Cenozoic) follows the same covariation trend. Thus, water content variability in refractory mantle olivine is not controlled by locations, depths or ages, but predominantly by its chemical composition. We also note that the possibility for hydrogen loss from olivine during ascent (Peslier and Luhr, 2006) suggests that measured water contents represent minima.

The OH in olivine predominantly occupies point defects in the crystalline structure, and hydrogen incorporation is thus controlled by cation vacancies in olivine (Demouchy and Bolfan-Casanova, 2016). Water storage capacity depends on vacancy population, influenced by factors such as temperature, pressure, composition and oxygen fugacity (Withers and Hirschmann, 2008; Wang, 2010). Vacancy concentration is primarily controlled by temperature; that is, with increasing temperature, vacancy concentration increases (Nakamura and Schmalzried, 1983). Studies have revealed that the $Fe^{3+}/(Fe^{3+}+Fe^{2+})$ ratio in olivine tends to increase with rising pressure within lithospheric mantle conditions (Gaetani *et al.*, 2014; Muir *et al.*, 2023),

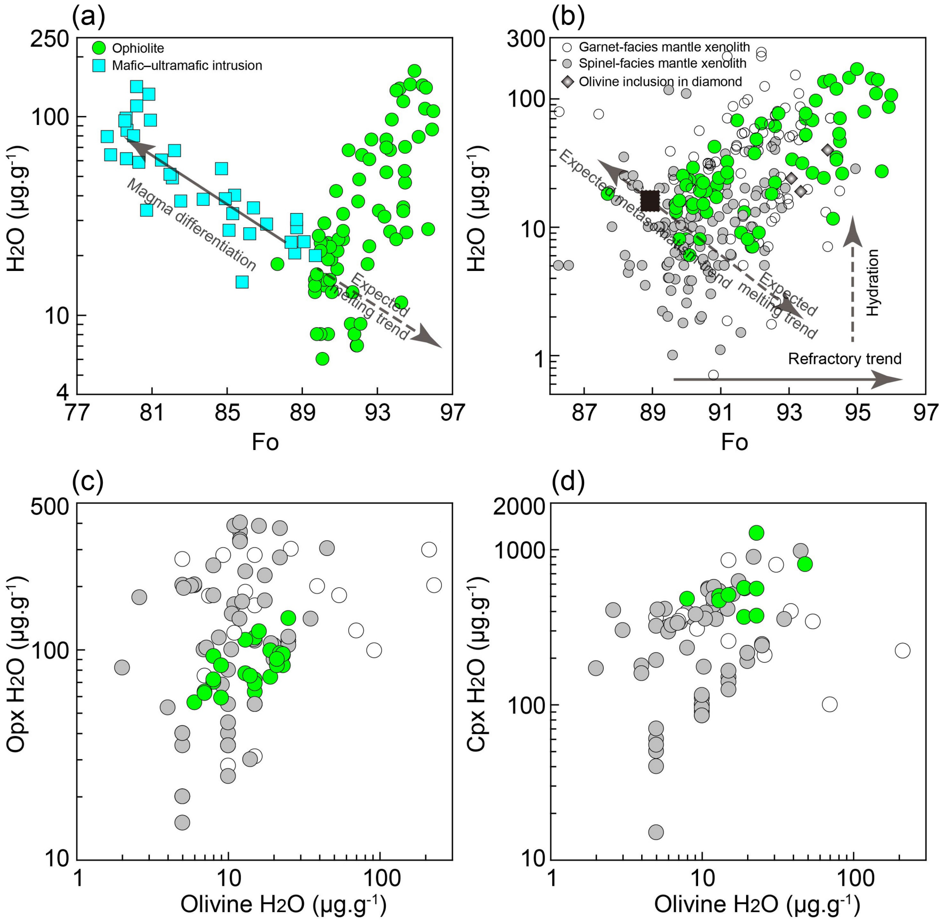

Figure 3 Comparisons of H_2O (µg.g^{-1}) *vs.* forsterite (Fo) contents in olivine between ophiolites, mafic–ultramafic intrusions, mantle xenoliths and inclusions in diamond. The magma differentiation trend is defined by the data from mafic–ultramafic intrusions, while expected melting and metasomatism trends are defined by incompatible behaviour of hydrogen in olivine. Spinel-facies mantle xenoliths, garnet-facies mantle xenoliths and olivine inclusion in diamond represent increasing depths of sub-continental lithospheric mantle. The refractory trend is defined by variation of forsterite content in olivine, while the hydration trend is expected from water variation observed in this study. The black square in panel b is the estimated water content of the lithospheric mantle beneath cratons (17 ± 13 µg.g^{-1}; Wang, 2010), with Fo 89 in olivine. The hydrated state of the lithospheric mantle depends on degree of refractoriness and is defined as the field above the line of expected melting trend in panel b. Plots of H_2O contents between olivine and **(c)** orthopyroxene (Opx) and **(d)** clinopyroxene (Cpx) in ophiolites and mantle xenoliths. See data sources in Supplementary Information.

generating more defects for the entry of hydrogen. This aligns with the observed variation in water content from spinel-facies to garnet-facies olivine from mantle xenoliths (Fig. 3b). Given that the $Fe^{3+}/(Fe^{3+}+Fe^{2+})$ ratio indicates oxygen fugacity, ophiolitic olivine formed under highly oxidised subduction conditions (Su *et al.*, 2023) is likely enriched in Fe^{3+} to enhance water storage capacity. However, determining this enrichment is challenging due to the extremely low Fe^{3+} content in olivine. Additionally, experimental results indicate that hydrogen in olivine occupies silicon vacancies at low Si activity (aSiO$_2$) and Mg vacancies at high aSiO$_2$ (Lemaire *et al.*, 2004). The Mg/Si ratios in various olivine are also covarying with water contents (Fig. S-1). Consequently, the lower Si content in refractory olivine enhances water storage compared to fertile compositions.

Water contents of olivine also depend on coexisting phases, particularly pyroxenes (Withers and Hirschmann, 2008), which have higher hydrogen partition coefficients (Kovács *et al.*, 2012; Demouchy and Bolfan-Casanova, 2016; Xia *et al.*, 2019). In ophiolites and mantle xenoliths, orthopyroxene shows variable and higher water contents than that of coexisting olivine (Fig. 3c), while the rough positive correlation of water contents is observed between clinopyroxene and olivine (Fig. 3c, d). The covariations suggest that olivine grains are equilibrated with pyroxenes in water contents for most, if not all, samples. In the less refractory or fertile lithospheric mantle, the increased presence of pyroxenes provides a competitive advantage relative to olivine, resulting in a low abundance of water in olivine. Conversely, the predominance of olivine in the refractory lithospheric mantle offers more opportunities for olivine to incorporate water during mantle hydration. Less competition from pyroxenes in the refractory mantle could potentially explain the higher water contents observed in olivine in dunites and chromitites (Fig. 2a), both of which notably lack pyroxenes (Fig. 1e–g). The significant water storage capacity of olivine might also hinder the crystallisation of hydrous minerals during mantle hydration, consistent with the rarity of hydrous minerals in ophiolitic rocks. Moreover, the hydrogen diffusion rate in defects in Fe-free olivine is one order of magnitude lower than that in Fe-bearing olivine at given *P-T* conditions (Padrón-Navarta *et al.*, 2014; Demouchy and Bolfan-Casanova, 2016; Fei *et al.*, 2018). Consequently, refractory olivine has a higher capacity for water preservation than fertile olivine. In addition, the tentative covariations of water contents between pyroxenes and olivine (Fig. 3c, d) indicate that orthopyroxene and clinopyroxene in the refractory mantle are also rich in water. Therefore, the refractory mantle should exhibit an even more pronounced

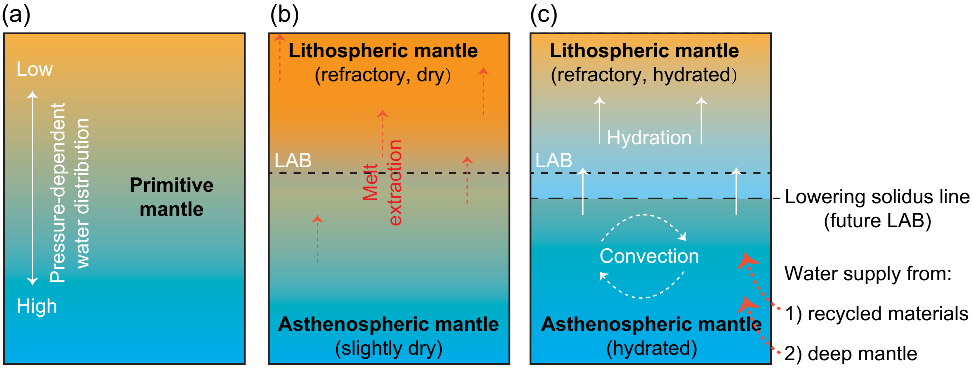

Figure 4 Illustration showing water distribution and variation during formation of local lithospheric mantle. **(a)** Water distribution in primitive mantle is mainly pressure-dependent. **(b)** Melt extraction results in removal of water from the asthenosphere and formation of lithospheric mantle. **(c)** Mantle convection supplies water from recycled materials or deep mantle to the top of the local asthenosphere where the lithospheric mantle just formed, and meanwhile water migration from the asthenosphere to the lithosphere occurs. See main text for more details. LAB, lithosphere-asthenosphere boundary. Hydration here refers to the infiltration of volatiles (hydrogen-dominated), leading to an increase in hydrogen abundance without affecting major and trace elements, which distinguishes it from melt/fluid metasomatism.

enrichment in water and would represent a significant water reservoir on a global scale.

The upward-decreasing water distribution in Earth's mantle (Hirschmann and Kohlstedt, 2012; Fig. 4a) is accentuated by melt extraction, forming a refractory, 'dry' lithospheric mantle (Fig. 4b). However, the rarity of olivine with Fo-water negative correlations (Fig. 3a) suggests that the 'dry' signature of the lithospheric mantle could not be preserved for a long time. Instead, the water enrichment observed in refractory olivine in this study and in literature suggests that the 'dry' signature of most lithospheric mantle should likely be immediately overridden by mantle hydration. Mantle hydration involves the infiltration of volatiles, predominantly hydrogen, resulting in an increase in hydrogen abundance without affecting the major and trace element contents (Nishi, 2015). This stands in contrast to typical melt- or fluid-induced metasomatism, which would normally enrich the mantle in incompatible elements, with reducing Fo contents in olivine. To discriminate typical melt/fluid metasomatism, "Simultaneous hydration and metasomatism of the lithospheric mantle by a water-rich silicate melt/fluid" and "hydrogen metasomatism" (Demouchy *et al.*, 2015) have been proposed to account for water enrichment of olivine (Liu *et al.*, 2017; Fig. 3b). The hydration process appears to be an ongoing and widespread phenomenon within the mantle, as the presence of hydrated refractory olivine remains consistent across various spatial and temporal settings. The water may originate from the decomposition of minerals in the deep mantle (Németh *et al.*, 2017) or from recycled materials mainly through plate subduction (Li *et al.*, 2018; Gose and Schmädicke, 2021; Su *et al.*, 2023). In comparison to the continental lithospheric mantle, the subduction zone is a water (over)saturated environment due to continuous dehydration of the subducting slab. Ophiolitic olivine could undergo more extensive hydration than those in the continental lithospheric mantle.

Partial melting removes water, creating a concentration gradient between the asthenosphere and lithospheric mantle. This gradient drives water migration into the lithospheric mantle, lowering solidus temperature of the refractory mantle (Hirschmann, 2006), prompting further melting of the already hydrated refractory mantle, enhancing its refractory nature. Concurrently, the asthenosphere beneath the refractory lithospheric mantle would receive water supply from recycled materials or deep mantle through mantle convection, and become more enriched in water. Similar to the overlying lithospheric mantle, partial melting could easily occur at the top of the locally hydrated asthenosphere, which would result in the formation of new lithospheric mantle, with a downward shift of the lithosphere-asthenosphere boundary (Fig. 4c). This melting-hydration feedback may facilitate lithospheric thickening.

Conclusion

This study challenges the paradigm of a dehydrated refractory lithospheric mantle by demonstrating that ophiolitic and xenolithic olivine can retain substantial water contents (5–170 μg.g^{-1}). The positive Fo-water correlation indicates that post-melting hydration—not initial melting—governs water storage. Hydration is enabled by favourable thermodynamic conditions and reduced competition from pyroxenes in refractory lithologies. Water incorporation lowers the solidus, triggering recurrent melting that enhances the refractory nature of mantle residues, while sustaining the refractory mantle as a major global water reservoir. This reservoir is replenished by fluids derived from deep mantle sources or subduction-related recycling. The cyclic feedback between hydration and melting preserves water in the lithospheric mantle and drives its thickening, offering a transformative framework for understanding upper mantle evolution.

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Additional Information

Supplementary Information accompanies this letter at https://www.geochemicalperspectivesletters.org/article2530.

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