

# JGR Solid Earth

## RESEARCH ARTICLE

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### Special Section:

Ophiolites and Oceanic Lithosphere, with a focus on the Samail ophiolite in Oman

### Key Points:

- An infrared spectral index using the area of OH/H<sub>2</sub>O absorptions at ~1,400 nm spatially maps and quantifies H<sub>2</sub>O in rocks with high accuracy
- The hydration ranges from 0 to 18 wt.% H<sub>2</sub>O with mean and medians for sheeted dike and gabbroic cores ranging from 1.5 to 3.2 wt.% H<sub>2</sub>O
- Deeper gabbroic oceanic crust is more hydrated due to relative prevalence of zeolite minerals and fault zones that facilitate hydration

### Supporting Information:

Supporting Information may be found in the online version of this article.

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## Characterizing Hydration of the Ocean Crust Using Shortwave Infrared Microimaging Spectroscopy of ICDP Oman Drilling Project Cores

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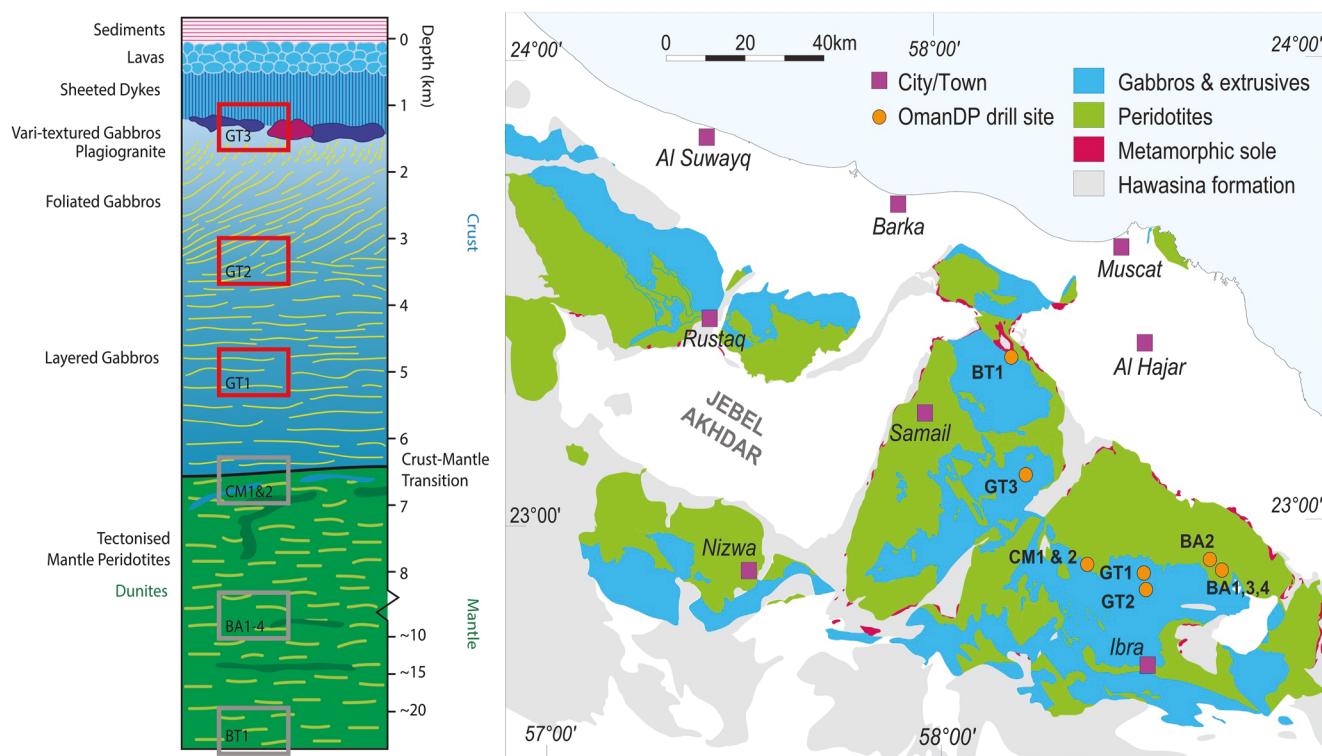
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**Abstract** Although ocean crust covers over 60% of Earth's surface, the processes that form, cool, and alter the ocean crust are not completely understood. We utilize shortwave infrared micro-imaging spectroscopy of ~1.2 km of rock cored by the International Continental Scientific Drilling Program's Oman Drilling Project to quantify hydration of basaltic dikes and gabbros from the Samail ophiolite as a function of depth, mineralogy and deformation. We develop a regression ( $R^2 = 0.66$ ) between area of the ~1,350–1,650 nm OH/H<sub>2</sub>O absorption and measurements of loss on ignition of samples and apply this relationship to generate quantitative ~250  $\mu\text{m}/\text{pixel}$  hydration maps for all cores. The lowest mean hydration is observed in the most pervasively altered dike-gabbro boundary (GT3A, H<sub>2</sub>O<sub>mean</sub> = 2.1  $\pm$  1.6 wt%), consistent with the low H<sub>2</sub>O content of the dominant alteration minerals, amphibole and epidote. The highest H<sub>2</sub>O content occurs in deeper foliated and layered gabbros (GT2A, H<sub>2</sub>O<sub>mean</sub> = 3.2  $\pm$  3.0 wt%) and layered gabbros (GT1A, H<sub>2</sub>O<sub>mean</sub> = 2.8  $\pm$  3.1 wt%). The greater prevalence with depth of zeolite alteration as opposed to lower wt% H<sub>2</sub>O amphibole at shallow stratigraphic depths, as well as the occurrence of zones of intensive hydration associated with fault zones (H<sub>2</sub>O<sub>mean</sub> = 5.7  $\pm$  4.0 wt%) lead to greater hydration of the lower ocean crust. This new approach provides an objective quantification of hydration in these cores, enabling an improved understanding of quantities and characteristics of ocean crust hydration. It highlights the importance of specific phases and faulting in controlling hydration, which has implications for ocean crust cooling, rheological properties, and the role of alteration in global biogeochemical cycling.

**Plain Language Summary** Ocean crust makes up much of Earth's crust. When it forms and cools, it reacts with seawater and other fluids to trap water in rock. The goal of this study was to determine how much water is stored within the ocean crust. We measured rock cores drilled from an ophiolite, a place where ocean crust has been thrust onto land. We used an infrared imaging spectrometer, commonly used on spacecraft to survey other planets, to measure reflected light as a function of wavelength to make maps of the amount of water in the core to determine what controls where the water is held in different minerals. We developed a method to determine the amount of water in the rocks using wavelength-dependent patterns in absorptions in reflected light. Most of the ocean crust contains 1.5%–3.2% water by weight. Surprisingly, the mid ocean crust sampled by our cores has the least amount of water, whereas the lower segments have more water. This is due to the presence in the cores of specific minerals called zeolites that form at low temperatures and the high extent of hydration in the rocks surrounding deep fault zones.

## 1. Introduction

Although the ocean crust covers over 60% of the Earth, it is not completely understood how the basaltic and gabbroic ocean crust forms, cools, and is chemically altered by seawater. The location and intensity of chemical exchanges between seawater and the ocean crust impacts global biogeochemical cycles and influences mechanisms for the accretion of new ocean crust. Consequently, it is important to improve our understanding of the ways in which the ocean crust formed and has been modified by fluid-rock interactions (e.g., Alt & Teagle, 1999; Coggon et al., 2016; Harris et al., 2017; Kelemen et al., 1997). Moreover, subduction of altered oceanic crust—together



**Figure 1.** Stratigraphic section (left) and geologic map (right) of the OmanDP boreholes. Boreholes considered in this study are outlined in red. Adapted from Kelemen et al. (2020).

with altered mantle peridotite and  $H_2O$ -bearing sediments—is the main input of molecularly bound  $H_2O$  into the Earth's upper mantle, where it drives volcanic arc magmatism, contributes to formation and evolution of continental crust, and reduces the viscosity of the convecting mantle (e.g., Blacic, 1972; Campbell & Taylor, 1983; Chopra & Paterson, 1984; Hacker, 2008). Thus, quantifying the  $H_2O$  content of oceanic crust is essential to understand the planetary water cycle.

It has been challenging to observe and measure relevant parameters because deep drilling of intact, in situ the ocean crust has proved very challenging (Michibayashi et al., 2019). Consequently, study of the magmatic accretion and hydrothermal exchange processes in the ocean crust has required the investigation of ophiolites: ancient sections of ocean crust and upper mantle formed at some style of ocean spreading center, now preserved on-land. Recently, the International Continental Scientific Drilling Program (ICDP) Oman Drilling Project (OmanDP) has drilled cores from ocean crust exposed in the mountains of Oman (Kelemen et al., 2013, 2020). These cores provide selected continuous sections of oceanic crust representative of specific intervals of the mid to lower ocean crust preserved in the Samail ophiolite. On-land diamond coring returned cores with near-100% recovery avoiding the drill sampling bias that commonly affects igneous rock sampling by scientific ocean drilling (e.g., Tominaga et al., 2009).

OmanDP recovered  $\sim 3.2$  km of rock from nine boreholes within the Samail ophiolite in Oman, which formed at a fast-spreading, submarine spreading center (e.g., Rioux et al., 2012; Tilton et al., 1981) and is the largest and best exposed section of oceanic lithosphere and upper mantle preserved on-land (Glennie et al., 1973; Searle & Cox, 1999) (Figure 1). Although trace element and geochronological data indicate that the Samail spreading center formed above a subduction zone, the crust formed via crystallization of primitive magmas very similar to parental mid-ocean ridge basalts (e.g., MacLeod et al., 2013; Pearce et al., 1981; Rioux et al., 2016). Although weathering and obduction may have further altered the ophiolite, the signatures of seafloor alteration remain. Key OmanDP objectives include better understanding of the formation of ocean lithosphere at mid-ocean

ridges, seafloor hydrothermal alteration, ocean-crust-mantle mass transfer, and weathering processes (Kelemen et al., 2013, 2020).

Processes in the mantle, the hydrosphere, the atmosphere, and the biosphere drive crustal hydration and alteration (e.g., Shuai & Yang, 2017). Water stored within minerals is the dominant reservoir of water on Earth, and the quantification of water in minerals and rocks illuminates the processes of hydration and alteration in the crust and, through subduction, impact the water budgets of the mantle (e.g., Bell & Rossman, 1992). Previous studies have measured bulk water contents of the ocean crust sampled by scientific ocean drilling ranging from 1 to 8 wt%  $H_2O$  in the upper oceanic crust and from 0.1 to 0.5 wt%  $H_2O$  in the lower gabbros (Agrinier, Hékinian, et al., 1995; Agrinier, Lavern & Tartarotti, 1995; Alt et al., 1996; Coggan et al., 2016; Jarrad, 2003; Godard et al., 2009; Kawahata et al., 1987; Kleine et al., 2020; Kusakabe et al., 1989; Shlobbreeva et al., 2011). However, core recovery is low for the lavas and dikes of the upper ocean crust (Alt et al., 1996), and cumulate gabbros have not been reached in intact ocean crust (Teagle et al., 2012). As a result of low recovery, more altered and fragile materials are underrepresented in the cores (Tominaga et al., 2009), leading to probable underestimates of the abundance of  $H_2O$  and other seawater-derived tracers in the ocean crust. In this study, access to core with near-complete recovery enables a more complete quantification of water content of the ocean crust, providing crustal hydration data for the wide range of materials, depths, and tectonic settings.

Spatially resolved infrared spectroscopy is non-contact, requires no sample preparation, and provides a method to map hydration non-destructively and contiguously at small spatial scales to address both the overall water content of the ocean crust and factors driving its heterogeneity. The entire OmanDP drill core was scanned with an imaging spectrometer at a spatial resolution of 250–260  $\mu m/pixel$  (Greenberger et al., 2021), and here we develop methods that utilize this data set to quantify and map the water content of the crust. Water in rocks occurs as both  $H_2O$  and hydroxyl OH groups, usually bonded with a metal in a mineral structure. In this study, we use “water” to refer to OH/ $H_2O$  collectively. Water has a number of absorptions in shortwave infrared, including the fundamental stretch of metal-OH ( $\sim 2,700$ – $2,800$  nm), the fundamental stretch of  $H_2O$  ( $2,760$ – $3,000$  nm), and the fundamental bend of  $H_2O$  ( $6,000$ – $6,200$  nm) (e.g., Bishop et al., 1994, 2002). Absorptions from  $2,700$  to  $3,000$  nm are commonly used in transmission spectroscopy to quantify water in minerals (Rossman, 2006). In single crystal mineral samples, infrared spectroscopy quantitatively determines the amount of water present within a mineral (Bell et al., 1995; Libowitzky & Rossman, 1996, 1997; Potter & Rossman, 1979; Rossman, 2006; Shuai & Yang, 2017). In geologic samples, the  $2,700$  nm OH and  $H_2O$  fundamental absorptions, which occur when a vibrational state is excited from  $\nu = 0$  to  $\nu = 1$ , have been shown to correlate directly with hydration in particulate minerals and mineral mixtures, irrespective of composition (Milliken & Mustard, 2005). However, for this study, we do not have spectral data at wavelengths longer than  $2,600$  nm. Instead, we develop and apply methods based on additional absorptions of OH/ $H_2O$  that do occur in our spectral range: combination absorptions that occur when two or more fundamental absorption modes combine, and overtone absorptions that occur near integer multiples of the fundamental absorption modes. For minerals in the OmanDP core, these include metal-OH overtones ( $1,380$ – $1,560$  nm),  $H_2O$  combination tones ( $1,410$ – $1,460$  nm), and  $H_2O$  combination tones ( $1,910$ – $1,970$  nm) (Bishop et al., 1994, 2002; White et al., 2017). Prior studies with particulate geologic materials have shown the  $1,900$ -nm  $H_2O$  combination band increases with increasing weight percent  $H_2O$  with mineral-specific dependencies (Milliken & Mustard, 2005).

We first develop and then apply infrared methods to quantify and map hydration at sub-millimeter scale over  $\sim 1.2$ -km of ocean crustal core from OmanDP Holes GT1A, GT2A, and GT3A. These three boreholes sample 400 m-deep sections of layered gabbros (GT1A), foliated and layered gabbros (GT2A), and the transition from sheeted dikes to gabbros (GT3A) (Figure 1; Kelemen et al., 2020). Using a combination of traditional geochemical measurements of loss on ignition (LOI) and imaging spectroscopy data, this study quantifies the spatially resolved hydration of the ocean crust and its variation, using a novel infrared spectral index. By examining the spatial patterns of hydration and alteration as a function of depth, mineralogy, and fault occurrence, we gain insight into the ways in which ocean crust cooled and was altered by hydrothermal fluids.

## 2. Methods

### 2.1. Determination of Loss on Ignition

Published loss on ignition (LOI) data, collected in the laboratories of the D/V *Chikyu* by the OmanDP science team during the ChikyuOman2017 core description campaign (Kelemen et al., 2020), were used to develop correlations between spectral indices and H<sub>2</sub>O content of rocks. Methods for determination of loss on ignition of OmanDP samples from Holes GT3A, GT2A, and GT1A are described in detail in Kelemen et al., (2020) and summarized here. After acquisition, the core was split into an archive half and a working half. Destructive analyses were performed on the working half following detailed core description. Representative whole-rock geochemical samples with adjacent oriented thin sections were cut with a diamond rock saw as either 5- or 8-cm long quarter-round sections. Core surface altered rinds were removed. The sections were then cut into thin slices, and saw marks were removed with 220  $\mu\text{m}$  sandpaper. Samples from GT1A and GT2A were then ultrasonicated in ethyl alcohol for 10 min and in deionized water three times for 10 min to clean them. Samples from borehole GT3A were ultrasonicated in deionized water three times for 10 min. Samples were dried for 6 hr at 105 °C, crushed to <5 mm, and then crushed to <2 mm in the Spex SamplePrep 3630 X-Press. To prepare fine powders for analysis, samples were further ground using a Fritsch Pulverisette 5 planetary mill with agate grinding bowls and agate balls.

Samples were weighed using the OHTI (Ocean High Technology, Institute, Inc., Tokyo, Japan) balance system into a ceramic crucible. A total of 5 g were used for samples from boreholes GT1A and GT2A, and 2–3 g were used for samples from borehole GT3A. The balance was calibrated at the beginning of the ChikyuOman2017 core description phase, and calibration was verified every 12 hr. Samples were then dried at 105°C for 2 hr to remove adsorbed water, cooled to room temperature in a desiccator, and reweighed. The powder was then ignited at 1000°C for 3 hr. The powder was cooled, placed in a glass desiccator, and reweighed to determine LOI. We correct those measurements for CO<sub>2</sub> and SO<sub>2</sub> also determined aboard (Table S1 in Supporting Information S1) by subtracting the measured CO<sub>2</sub> and volatile S contents from LOI. We then use the corrected LOI as the basis of comparison with water quantification from imaging spectroscopy data.

It has also been noted that the oxidation of Fe in samples during heating may increase the residual sample weight thereby reducing the measured LOI (Kelemen et al., 2020; Lechler & Desilets, 1987). If all Fe had been present as Fe<sup>2+</sup> and it was all oxidized, then the H<sub>2</sub>O from LOI would be on average 0.8 wt% greater. However, given that iron oxides and epidote containing Fe<sup>3+</sup> are present in the core, it would be incorrect to assume that all the iron was initially present as Fe<sup>2+</sup>. Because the Fe<sup>2+</sup> to Fe<sup>3+</sup> ratio is unknown as is the extent of any oxidation during the procedure, we choose not to correct for Fe oxidation. Different choices in method of H<sub>2</sub>O calculation from LOI only lead to H<sub>2</sub>O changes in all holes at the  $\leq 1$  wt% level, affecting each hole in a similar manner (Text S1 and Table S1 in Supporting Information S1).

### 2.2. Imaging Spectroscopy

Imaging spectroscopy, also known as hyperspectral imaging, is a technique which enables the user to combine both spatial and spectral data to determine information about a material and the abundance and/or state of an absorbing species (Clark et al., 2003; Goetz et al., 1985). Absorptions present in spectra correspond to chemical bonds and can be used to determine the mineralogy and the amount of water and hydroxide in minerals and rocks (e.g., Asimow et al., 2006; Bell et al., 2003, 2004; Burns, 1993; Clark et al., 1990; Hammer et al., 1996; Hunt, 1977; Jamtveit et al., 2001; Johnson & Rossman, 2003; Milliken & Mustard, 2005, 2007a, 2007b; Schuttlefield et al., 2007; Stolper, 1982). Our method of reflectance imaging spectroscopy of the core is non-destructive, enabling acquisition of whole core hydration at sub-millimeter scale with spatial context preserved and requiring no sample preparation. In contrast, although transmittance spectroscopy has demonstrated accuracy for hydration determinations, it requires destructive sampling of the core and consequently can only be applied to small regions of samples.

Methods for the imaging spectroscopy data acquisition and mineral mapping are described in detail in Kelemen et al. (2020), a companion paper (Greenberger et al., 2021), and summarized here. Imaging spectroscopy measurements were obtained on the archive halves of the OmanDP cores in August-September 2017 with the

Caltech Ehlmann laboratory's pushbroom imaging spectrometer, which acquires line-by-line a cross-core spatial row of pixels, measuring reflected light; each pixel has a corresponding spectrum. This instrument has both a visible-near infrared (VNIR; 0.4–1.0  $\mu\text{m}$ ) and a shortwave infrared (SWIR; 1.0–2.6  $\mu\text{m}$ ) sensor. For this study, the data from the SWIR sensor was used because the relevant OH/ $\text{H}_2\text{O}$  absorption features are in that wavelength range. The SWIR data were acquired at 6 nm spectral resolution, 6 nm spectral sampling, and 250–260  $\mu\text{m}/\text{pixel}$  spatial resolution for the HQ core and 260  $\mu\text{m}/\text{pixel}$  for the NQ core with exposure times of 4.5–5.4 ms. After every four to six images, the spectrometer was calibrated with measurements of dark current and a Spectralon reference target.

The data were stored as spectral image cubes, and regions of interest (ROIs) were defined and outlined in ENVI software using known depths of the geochemical samples used for LOI and thin section images. Using the automated methods of Greenberger et al. (2021), pixels in the images that were not the rock core (i.e., pixels containing the plastic core liner or Styrofoam) were identified and excluded from further analysis. We also used methods described in Greenberger et al. (2021) to combine analyses of all core sections from each borehole to create a continuous length of core with depths assigned. For calculation of mean, median, and standard deviation statistics, pixels 65–214 in the horizontal or  $x$ -direction were used for the larger diameter HQ-size core sampled at 250  $\mu\text{m}/\text{pixel}$  and pixels 65–220 for NQ-size core sampled at 260  $\mu\text{m}/\text{pixel}$ . There are 4% fewer pixels vertically in the coarser resolution of NQ core, so we compensate with 4% more pixels in the horizontal direction. This sub-sampling ensures that approximately the same number of pixels per meter are included in calculation of statistics and removes sample size biases from downhole changes in core diameter and spatial resolution. For downhole statistics aggregated at 10 cm or 1 m spacing, we use all pixels that contain the core within each interval. Data from miscellaneous core, designated “M” (Kelemen et al., 2020) where portions of holes were re-drilled, are excluded from our downhole analyses.

### 2.3. Calculation of Integrated Absorption Area

While simple absorption band intensities for a sample can vary depending on the instrument's polarizer and spectroscopic resolution, absorption band areas do not vary significantly as a result of changes in polarization and resolution (Rossman, 2006). The relationship between integrated absorbance and the amount of water in a mineral has been successfully determined for single crystals but has yet to be extensively studied in bulk rocks (Shuai & Yang, 2017).

The integrated band areas of the  $\text{H}_2\text{O}$  combination absorptions (1,900 nm) and the OH overtone and  $\text{H}_2\text{O}$  combination absorptions (1,350 nm–1,650 nm) were both initially calculated in this study (Figure 2). In order to obtain the integrated areas of these two bands, we first smoothed the imaging spectroscopy data with the boxcar averaging SMOOTH function in IDL with a width of three in all dimensions (spatial  $x$  and  $y$  and spectral). The average reflectance value is then plotted at the central wavelength and pixel value for the sample. This treatment was particularly important for the low  $\text{H}_2\text{O}$ , dark samples, where the  $\text{H}_2\text{O}$  and OH absorptions are small in magnitude and a positive or negative spike from noise has a large effect on the subsequent continuum removal and band area quantification (Figure 2i). By smoothing the spectrum, the magnitude of noise is decreased, which provides a more accurate measure of the depth of the  $\text{H}_2\text{O}$  combination and OH overtone absorptions. Although some of the sharpness of hydroxyl absorptions is lost, perhaps yielding a slight underestimation, the overall effect is to remove spurious data points. The effects of smoothing are shown in Figure 2 for a high hydration, moderate hydration, and low hydration pixel.

A continuum removal was then performed on spectral subsets of the smoothed spectrum utilizing the CONTINUUM\_REMOVE\_DOIT function in IDL. This function uses a convex hull fit to remove the continuum from the spectrum in every pixel in the hyperspectral images of the cores. To calculate  $\text{H}_2\text{O}$  concentrations pixel-by-pixel in the core, we first derived integrated band areas for each pixel using the continuum removed spectra and a rectangle approximation method. With this method, a rectangle width is defined as halfway between the prior bandpass and the center bandpass to halfway between the center bandpass and the next bandpass. The height of the rectangle is defined as one minus the reflectance measurement of each spectral band, that is, the area between the continuum (1.0) and the continuum removed spectrum. The area of the rectangle is then the width of the rectangle multiplied by the height of the midpoint, and the total area of the absorption is defined as the sum of the areas of all the rectangles defined across the range. The results

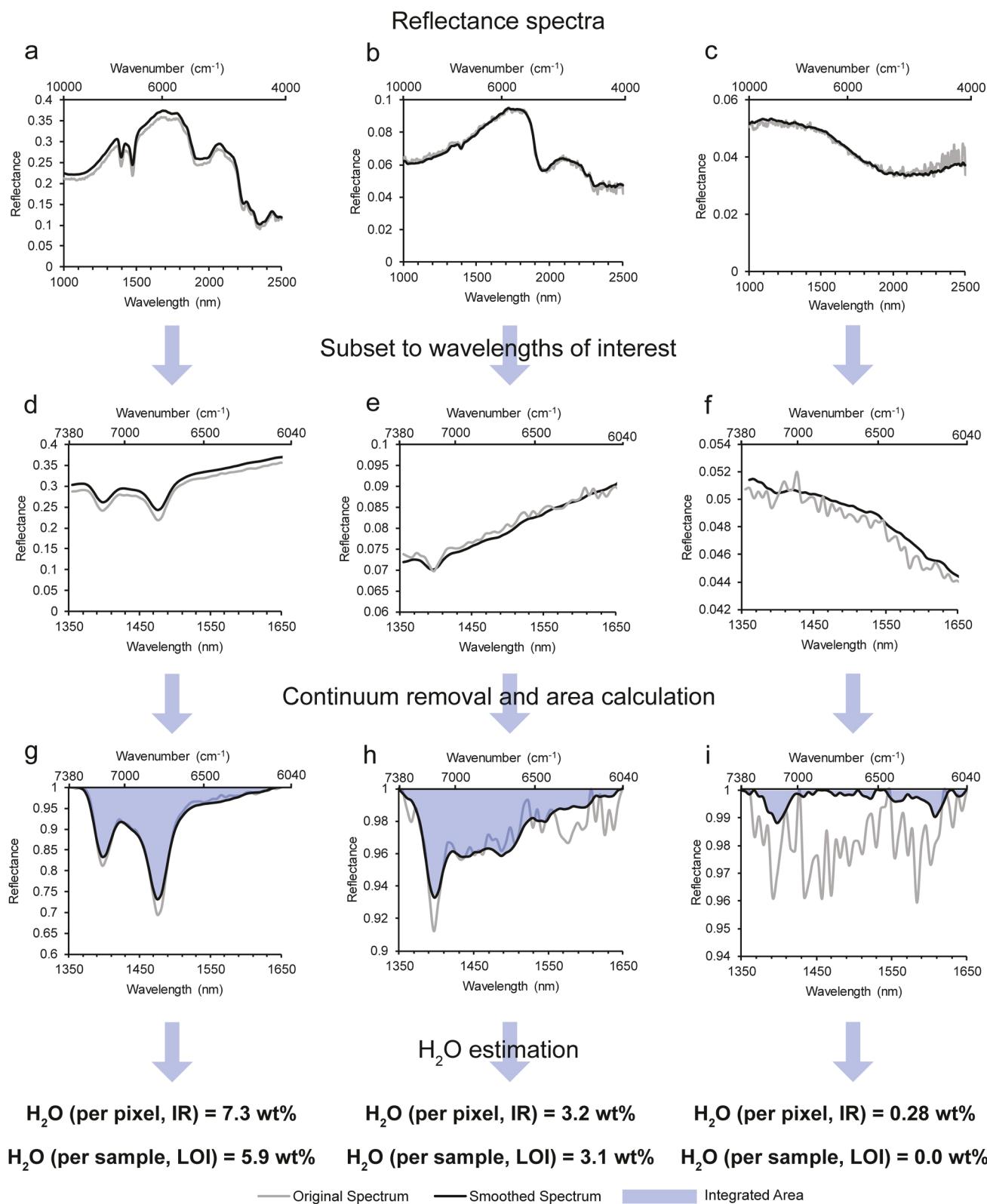


Figure 2.

of the rectangular approximation varied by <1% from the computationally slower five-point Newton-Cotes approximation using the INT\_TABULATED function in IDL.

We continuum removed and calculated band area for the  $\text{H}_2\text{O}$  combination region from 1,823–2,100 nm (5,485–4,762  $\text{cm}^{-1}$ ) and the OH overtone and  $\text{H}_2\text{O}$  combination region from 1,355 nm–1,654 nm (7,380–6,046  $\text{cm}^{-1}$ ). We evaluated four different means of calculating continuum-removed band areas: (a) the 1,900-nm band area with units of wavenumber, (b) 1,900-nm band area with units of wavelength, (c) the 1,400-nm band area with units of wavenumber, and (d) the 1,400-nm band area with units of wavelength.

Although raw data from the instrument were in units of wavelength, the motivation for the evaluation of the continuum removal in units of wavenumber is because it is a unit of energy. Absorption of light energy is directly related the type and quantity of minerals, and wavenumber is therefore commonly used for quantification of materials with spectroscopy (e.g., Bell et al., 1995; Libowitzky & Rossman, 1997; Potter & Rossman, 1979; Rossman, 2006; Shuai & Yang, 2017; Sunshine et al., 1990). Because the conversion between wavelength and wavenumber is not linear, band area calculations in units of wavelength and wavenumber result in slight differences in the distribution of values.

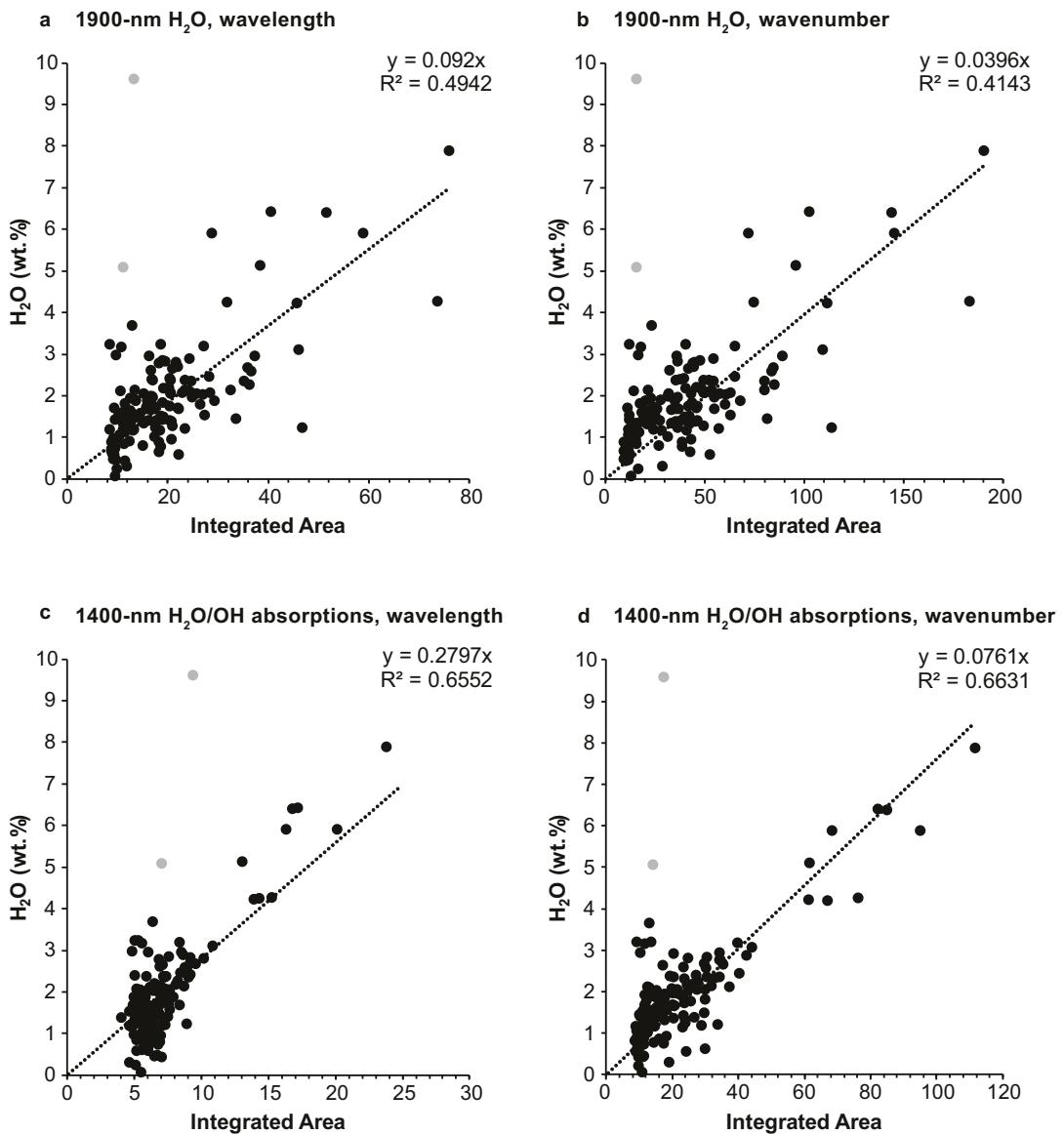
#### 2.4. Mapping Hydration With Integrated Area Versus Hydration Relationships

In order to determine the relationship between hydration and integrated band area of the four different calculated OH/ $\text{H}_2\text{O}$  band areas, we considered the LOI measurements obtained from whole rock samples in Holes GT1A, GT2A, and GT3A. ROIs were defined in the imaging spectroscopy data of the archive half of core that corresponded to the depths and core widths sampled in the working core. The mean integrated area of each sample was calculated by taking the mean of all integrated areas calculated for each pixel in the defined ROIs with each of the four different band area calculation methods (Table S2 in Supporting Information S1).

Linear regressions fit through (0, 0) were calculated for each of the four different band area calculations between integrated area and measured LOI (Figure 3). We excluded data from two samples of uncommon rock types that had weak absorption features with low integrated areas but high measured  $\text{H}_2\text{O}$  values. The inclusion of these samples in the regression decreased the accuracy of our  $\text{H}_2\text{O}$  estimation for more common rock types composing the vast majority of the rock from these three boreholes. One sample excluded from the regression came from GT1A 41Z-2, 98.26–99.16 m. In the igneous core description logs, this site was defined as a disseminated oxide olivine mela gabbro, present only between 96.36–101.23 m in Hole GT1A (1.2% of Hole GT1A and ~0.4% of all the core considered in this study) (Kelemen et al., 2020). The second excluded ROI was dunite (serpentinized) from GT1A 107Z-3, which only occurs in two depth intervals totaling 23 cm in Hole GT1A (~0.06% of Hole GT1A and ~0.02% of all core included in this study) (Kelemen et al., 2020). Both rock types are very dark, and prior work has shown that the depths of absorption features of OH and  $\text{H}_2\text{O}$  are affected by darkening agents such a magnetite, spinels, and sulfides (Milliken & Mustard, 2007a). These rock types are rare in the OmanDP holes considered in this study and the great majority of the plutonic ocean crust. However, these rock types are present in some IODP sites and dunites and other highly serpentized rocks are common around the crust-mantle boundary. For the quantification of hydration in cores where these rock types are abundant, different calibrations may need to be determined specifically for these very dark colored rocks.

For all four band area calculations, integrated band area increases with increasing hydration as measured by LOI in an approximately linear manner (Figure 3). This relationship determines wt.%  $\text{H}_2\text{O}$  with average errors of  $0.52 \pm 0.49$  wt.% (Table S3 in Supporting Information S1) and shows more scatter at <2–3 wt. %

**Figure 2.** Three example spectra showing the impacts of smoothing and continuum removal on the calculation of integrated area for single pixels. (a, d, g) The high hydration example is a pixel from Hole GT1A, Section 140Z-2. (b, e, h) The medium hydration sample is a pixel from Hole GT2A, Section 31Z-1. (c, f, i) The low hydration example is a pixel from Hole GT2A, Section 108Z-1. (a–c) Reflectance spectra. (d)–(f) Reflectance spectra at wavelengths used in final regression. (g–i) Continuum removed spectra at wavelengths used in final regression. The shaded region represents the integrated area calculated for each pixel.  $\text{H}_2\text{O}$  (pixel, IR) corresponds to the calculated wt%  $\text{H}_2\text{O}$  calculated based on the integrated area of the smoothed IR spectrum for the single pixels shown in (a–i) using Equation 1.  $\text{H}_2\text{O}$  (sample, LOI) gives the measured  $\text{H}_2\text{O}$  from loss on ignition (LOI) for the entire regions of interest (ROI) representing the sampled area. The single pixel is located within the ROI. Minor differences between the smoothed and unsmoothed spectra may occur due variability within the surrounding pixels.

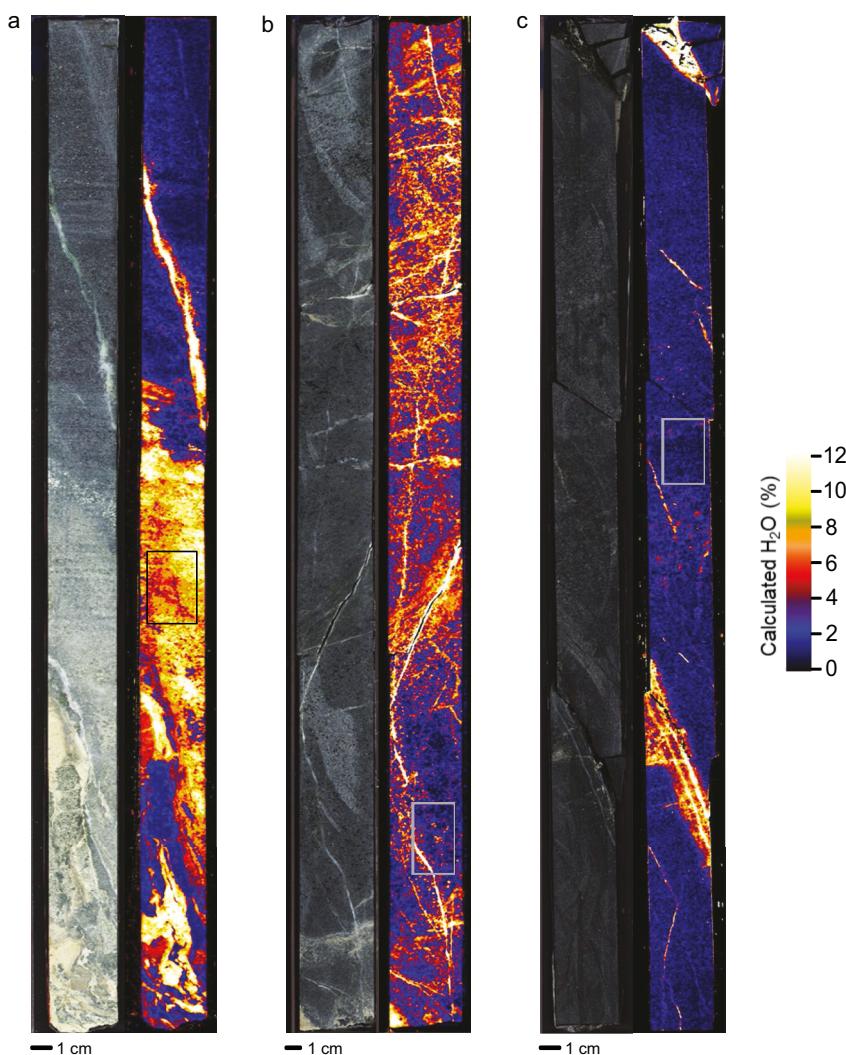


**Figure 3.** Results from each set of parameters for the area calculations: (a) Integrated area of the  $H_2O$  combination absorption at 1,900 nm calculated in wavelength space versus  $H_2O$  measured via loss on ignition (LOI); (b) Integrated area of the  $H_2O$  combination absorption at 1,900 nm calculated in wavenumber space versus  $H_2O$  measured via LOI. (c) Integrated area of the  $\sim 1,400$  nm  $H_2O/OH$  absorptions calculated in wavelength space versus  $H_2O$  measured via LOI. (d) Integrated area of the  $\sim 1,400$  nm  $H_2O/OH$  absorptions calculated in wavenumber space versus  $H_2O$  measured via LOI. Gray circles are outliers and rare rock types excluded from the regression.

$H_2O$ . The  $R^2$  value between the integrated area and measured LOI is the highest ( $R^2 = 0.66$ ) when using the continuum removed spectrum for the  $\sim 1400$ -nm absorption in wavenumber units ( $BA_{1400, \text{wavenum}}$ ) with the equation

$$\text{Calculated wt.\% } H_2O = 0.0761 \times BA_{1400, \text{wavenum}} \quad (1)$$

To map hydration in the entire core, the area of the OH overtone absorption was calculated for each pixel utilizing the smoothed and continuum removed spectrum in wavenumbers for each pixel and Equation 1.



**Figure 4.** Example of hydration maps (right) shown with standard color scans of core sections from the multi-sensor core logger (Kelemen et al., 2020). Approximate location sampled for geochemical analysis is outlined in a rectangular box on the hydration image (left). (a) A high hydration sample from Hole GT1A, Section 140Z-2 (LOI H<sub>2</sub>O = 5.9 wt%; spectral estimate 7.2 wt%). (b) A medium hydration sample Hole GT2A, Section 31Z-1 (LOI H<sub>2</sub>O = 3.1 wt%; spectral estimate 3.3 wt%). (c) A low hydration sample from Hole GT2A, Section 108Z-1 (LOI H<sub>2</sub>O = 0.0 wt%; spectral estimate 0.7 wt%).

### 3. Results

The LOI measurements used in this paper yield H<sub>2</sub>O contents of 0.058–9.75 wt%. For further discussion of the LOI results, see Kelemen et al. (2020).

#### 3.1. Mapping Core Hydration

Hydration maps were calculated for all core sections using the equation derived in Section 2 (Figures S1–S3 in Supporting Information S1). Examples of the hydration maps for core sections with LOI samples indicating high OH/H<sub>2</sub>O, intermediate OH/H<sub>2</sub>O, and low OH/H<sub>2</sub>O are shown in Figure 4. Hydration maps of the core show a high degree of spatial coherency and detail. Transitions between high hydration and low hydration regions correspond to and commonly follow the boundaries of subtle color differences present in color images of core section that indicate different rock types or degrees of alteration. There is very little speckling in hydration maps that could indicate noise or discrepancies in band calculation. Instead, the hydration maps

in fact reveal more structural complexity in veins and fractures than the standard color imagery. Mineralized fractures and veins present in the cores are commonly sites of higher hydration. Most veins and fractures are surrounded by halos of slightly more hydrated rock.

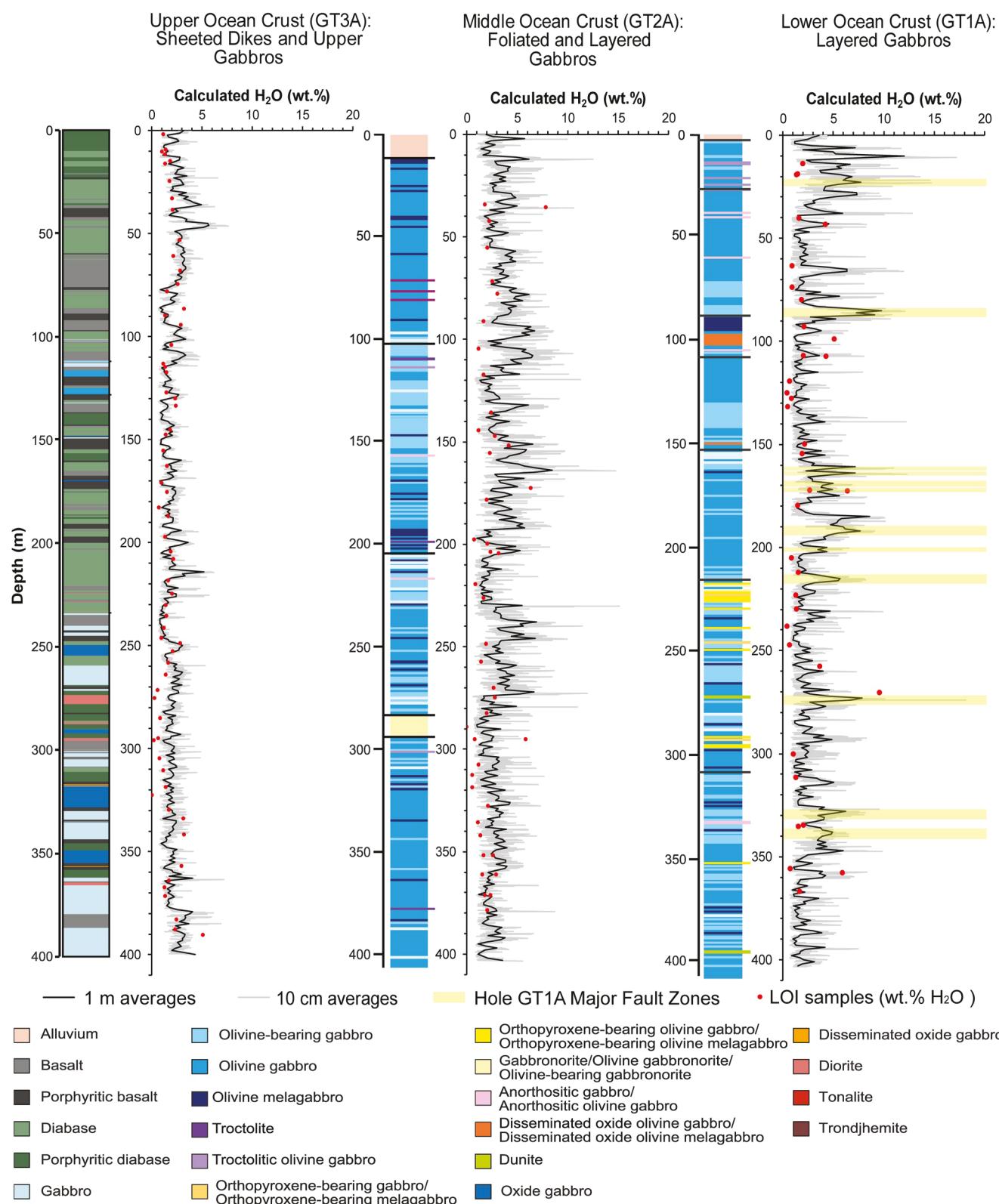
Downhole plots of calculated  $H_2O$  estimates with 1 m and 10 cm averages for each borehole are shown in Figure 5. At a spatial resolution of 250–260  $\mu m$ /pixel, hydration in these boreholes ranges from 0 to 20 wt%  $H_2O$ . Over 85% of the ocean crust has hydration values of less than 5 wt%  $H_2O$ . Qualitatively, all three holes have similar values for most intervals without systematic hydration change as a function of depth (Figure 5). However, Holes GT2A and GT1A have substantially more high hydration excursions. The dike-gabbro transition Hole GT3A exhibits the lowest mean  $H_2O$  ( $2.1 \pm 1.6$  wt%), whereas intermediate depth Hole GT2A that samples mid-crustal foliated and layered gabbros, exhibits the highest mean  $H_2O$  ( $3.2 \pm 3.0$  wt%) (Table 1). The mean hydration of lower crustal Hole GT1A ( $2.8 \pm 3.1$  wt%) is between the means of Hole GT2A and Hole GT3A. However, in terms of median hydration, a different trend is observed. Across all three holes, the median  $H_2O$  is lower than the mean  $H_2O$ . This is likely due to zones containing very hydrated pixel band areas, such as pixels containing primarily zeolite, particularly thomsonite and laumontite, that drive up the mean  $H_2O$  significantly. Hole GT2A has the highest median  $H_2O$  (2.2 wt%), but Holes GT3A and GT1A have much lower medians (1.7 wt% and 1.5 wt%, respectively). In GT1A, many of the high hydration depth intervals are associated with faults (Figure 5). Other high hydration intervals are caused by proximity to minor fault or damage zones (e.g., at 315 and 360 m in Hole GT1A, Figure 5) or increased abundances of high- $H_2O$  zeolite minerals (e.g., at 65 m in Hole GT1A, Figure 5; see Greenberger et al., 2021 for mineral occurrence maps). The controls on  $H_2O$  in the ocean crust is an important question and a topic that we are continuing to explore. However, the purpose of this paper is to provide quantitative estimates of  $H_2O$  within the ocean crust and some initial discussion of the controls on its distribution. A thorough analysis of the causes of all high  $H_2O$  zones and their significance—as well as increases in concentration of key minerals—is beyond the scope of this paper, and a prime topic for future research.

In order to understand the calculated mean and median  $H_2O$  and the range of  $H_2O$  contents between holes GT3A, GT2A, and GT1A, we consider the variations in hydration values and generate histograms of  $H_2O$  content for each hole (Figure 6). From Figure 6, it is evident that, while the much of the core from all boreholes has 0.5%–2%  $H_2O$ , GT2A and GT1A have longer tails toward higher  $H_2O$  values. ~37% of Hole GT2A by area has  $H_2O$  concentrations  $>3$  wt%. In contrast, only ~30% of Hole GT1A and ~19% of Hole GT3A by area have  $H_2O$  concentrations  $>3$  wt%. Approximately 60 wt% of Holes GT1A and GT3A are  $<2$  wt%  $H_2O$  by area whereas only ~47% of Hole GT2A by area is  $<2$  wt%  $H_2O$ . Thus, it is evident that Hole GT2A has more zones of elevated  $H_2O$ , likely composed of high  $H_2O$  content minerals, relative to Holes GT1A and GT3A.

### 3.2. Mineral Carriers of Hydration

Both the visual core logging (Kelemen et al., 2020) and determination of spatial occurrences of key minerals (Greenberger et al., 2021) found that Hole GT3A exhibits the most pervasive alteration as measured by its lowest occurrence of primary minerals and highest occurrence of secondary minerals, yet we find the lowest levels of hydration in Hole GT3A. Thus, there are factors beyond simply alteration driving the hydration of the ocean crust, likely the type of secondary minerals formed. Comparing mean hydration of each 1-m length of core and the percentage of pixels within that meter that contained the mineral of interest from Greenberger et al. (2021) provides a means of understanding the carrier phases of hydration in the ocean crust. While overall hydration increases with alteration of primary igneous minerals, the specific phases of the alteration mineralogy appear to be one of the most important drivers of changes in degree of bulk hydration with depth (Figure 7).

Natural pyroxenes commonly host ~50–100 ppm OH (Skogby et al., 1990), but to first order, in its unaltered form, pyroxene ((Ca,Mg,Fe)Si<sub>2</sub>O<sub>6</sub>) contains very little water. As expected, with increasing frequency of pyroxene mapped in a given meter of core, the predicted  $H_2O$  decreases. This trend is observed in Figure 7a, indicating that the mineralogical and the hydration data are consistent with each other. Holes GT1A and GT2A have more unaltered pyroxene than Hole GT3A, clearly exhibit the expected correlation of hydration increase and pyroxene decrease, indicating that alteration drives hydration.

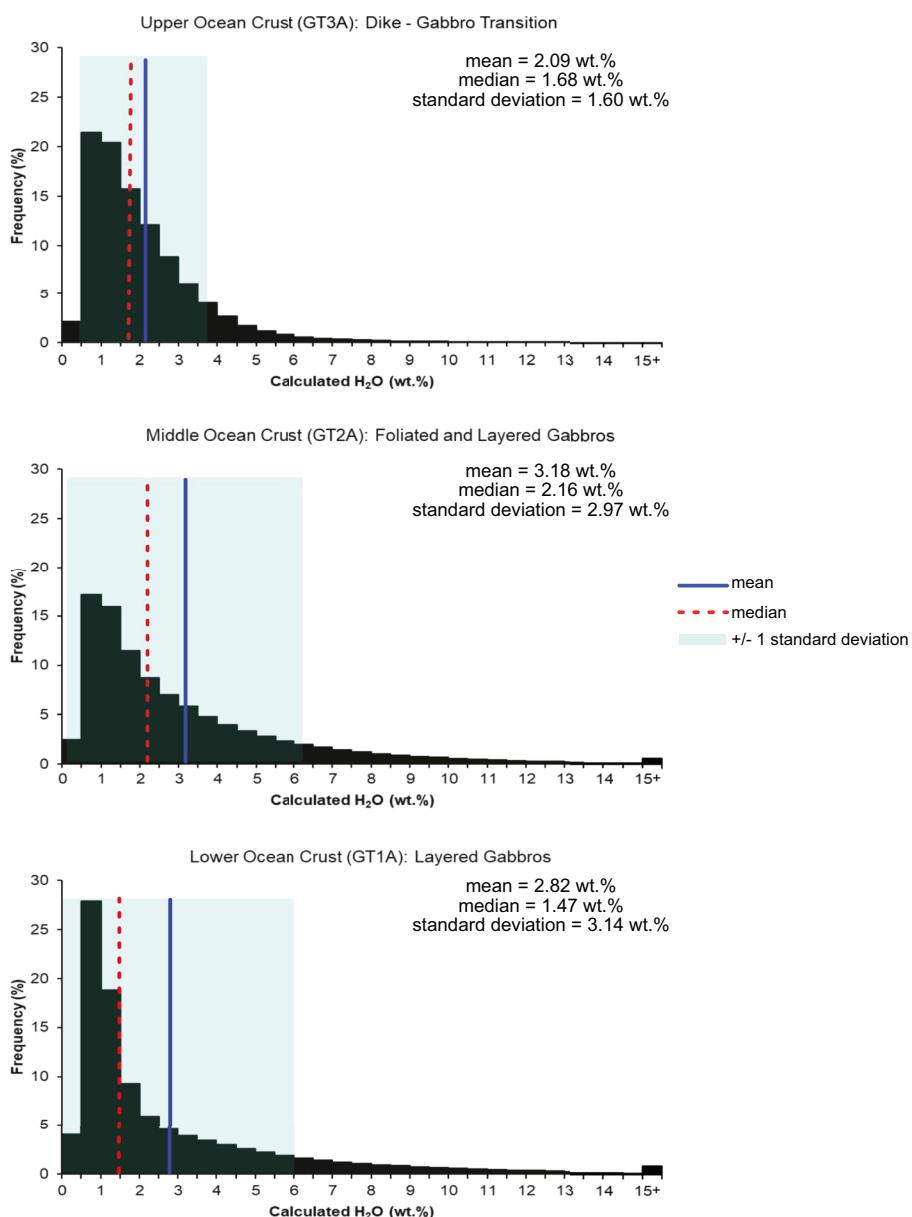


**Figure 5.** Downhole plots of calculated wt%  $H_2O$  versus depth for Holes GT3A, GT2A, and GT1A. 1 m averages of calculated wt%  $H_2O$  are shown in black, and 10 centimeter averages of calculated wt%  $H_2O$  are shown in light gray. For comparison, red dots are the wt%  $H_2O$  of samples with LOI measurements. The main fault zones in GT1A are marked in yellow from Kelemen et al. (2020). Major fault zones were only identified and described for Hole GT1A, so we only consider fault zones from GT1A here. All fault zones regardless of size were grouped for Holes GT3A and GT2A.

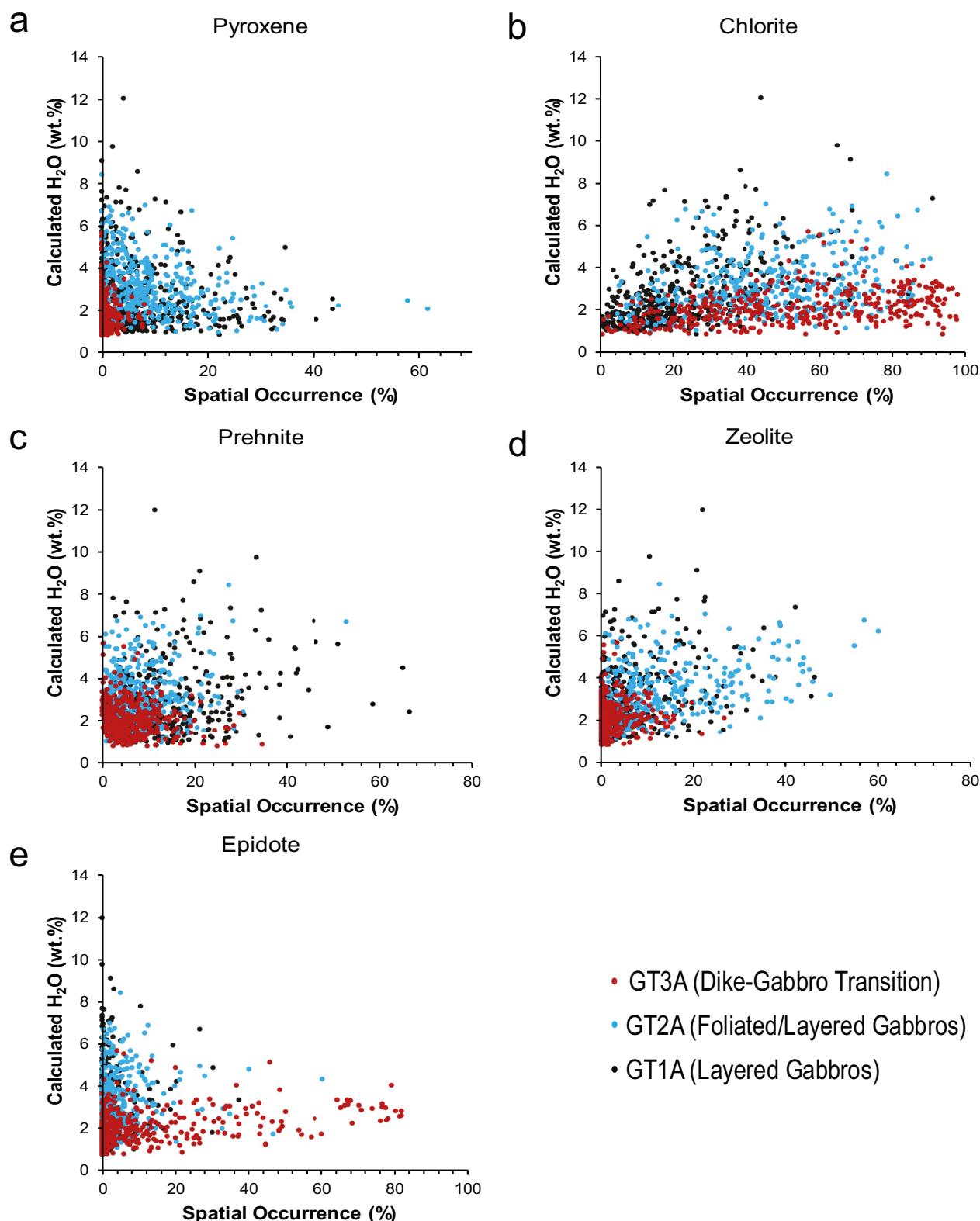
**Table 1**

*Mean, Median, and Standard Deviation of Calculated Weight Percent  $H_2O$  Values for Holes GT3A, GT2A, and GT1A Using Imaging Spectroscopy and, for Comparison, the LOI Geochemistry Measurements (Kelemen et al., 2020)*

Hole	H <sub>2</sub> O (wt%) from spectroscopy			# Of samples	H <sub>2</sub> O (wt%) from LOI samples		
	Mean	Median	Standard deviation		Mean H <sub>2</sub> O	Median H <sub>2</sub> O	Standard deviation
GT3A (dike—gabbro transition)	2.1	1.7	1.6	64	1.7	1.5	0.8
GT2A (foliated and layered gabbros)	3.2	2.2	3.0	43	2.2	2.0	1.5
GT1A (layered gabbros)	2.8	1.5	3.1	34	2.2	1.6	2.0



**Figure 6.** Histograms of calculated wt%  $H_2O$  content for pixels in Holes GT3A, GT2A, and GT1A. All values greater than or equal to 15 wt% calculated  $H_2O$  were combined into one bin.



**Figure 7.** A comparison between the spatial occurrence frequency of a mineral (the percentage of pixels in which the mineral occurs) within each meter of core versus the mean calculated  $\text{H}_2\text{O}$  in that meter. The relationships between the spatial occurrence frequency of (a) pyroxene, (b) chlorite, (c) prehnite, (d) zeolite, and (e) epidote versus calculated wt%  $\text{H}_2\text{O}$ .

**Table 2**

$H_2O$  wt% Inside and Outside of Fault Zones in GT1A, Measured by the Spectral Index

	Mean $H_2O$ (wt%)	Median $H_2O$ (wt%)	Standard deviation (wt%)
All GT1A	2.8	1.5	3.1
In fault zones	5.7	4.7	4.0
Out of fault zones	2.6	1.3	2.9

*Note.* Depths corresponding to fault zones are 21.00 m–24.85 m, 84.14 m–87.62, 161.20 m–162.16 m, 164.2 m–165.84 m, 168.60 m–170.29 m, 172.27 m–173.33 m, 190.59 m–194.24 m, 200.20 m–201.98 m, 213.95–216.81 m, 272.59 m–275.13 m, 329.05 m–333.85 m, and 336.03–340.40 m (Kelemen et al., 2020).

The most commonly occurring alteration mineral is chlorite, and hydration is observed to increase monotonically with abundance of chlorite (Figure 7b). The relationship is different for each hole, however, with a relatively modest increase in hydration for each wt% chlorite for GT3A and greater increases for GT1A and GT2A. This may be due to the overall abundance of chlorite and the role of accessory phases that are in mineral assemblages with chlorite. Prehnite and zeolite are more common spatially in Hole GT1A and Hole GT2A compared to Hole GT3A (Greenberger et al., 2021). Zeolite minerals contain on the order of ~15 wt%  $H_2O$ . Regions of the core which have the highest prevalence of epidote tend to converge to hydration values of ~3% (Figure 7e). Areas of very high hydration have relatively low epidote abundance of less than 20%, indicating that the water contents in regions of high hydration in the ocean crust are dominated by water in zeolites. Higher abundances of zeolite and zeolite-prehnite assemblages in Hole GT2A and GT1A may be driving their higher mean hydration values, and the lower frequency of these minerals in Hole GT3A may explain its lower predicted  $H_2O$  values

(Figures 7c and 7d). (We note that limitations in the mapping by Greenberger et al., 2021; prohibit identification of zeolites in pixels with the 1.48  $\mu m$  overtone of OH that is diagnostic of prehnite, resulting in likely underestimation of zeolite proportion.) Thus, although hydration increases with increased alteration, the specific minerals formed as a result of alteration processes are important in affecting the overall amount of water sequestered at different depths of the crust.

### 3.3. Hydration and its Correlation With Structural Features

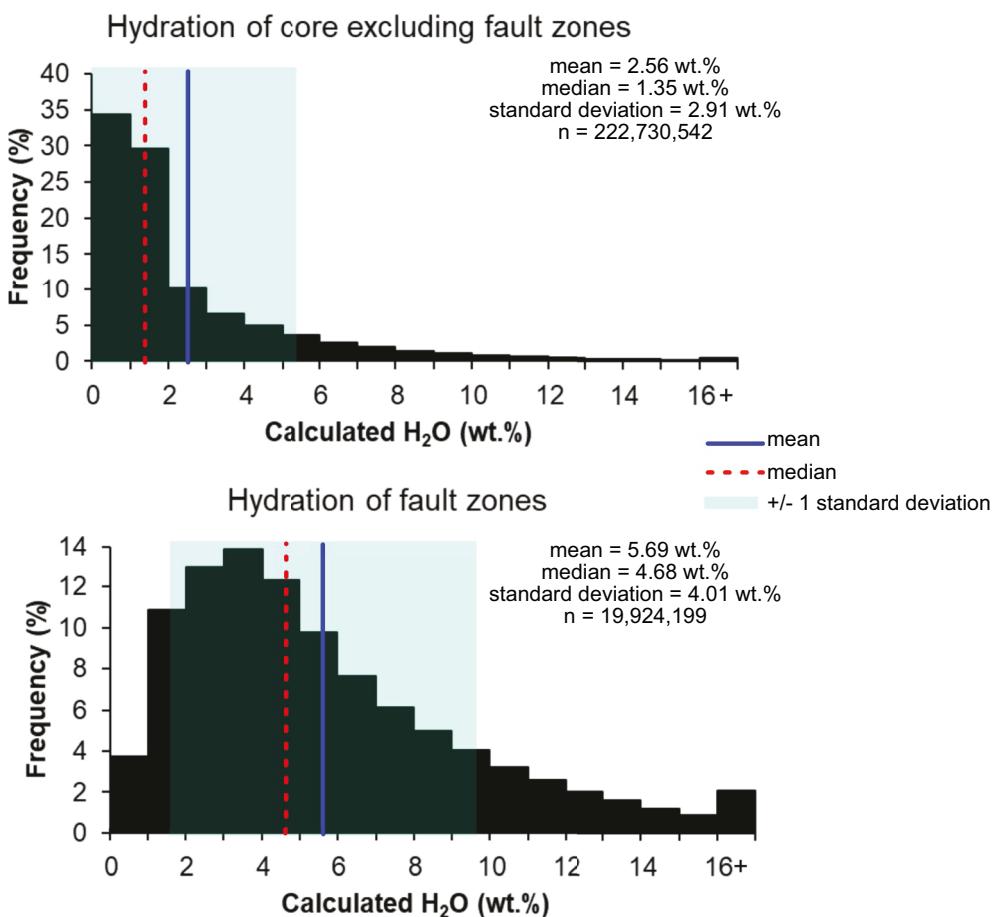
Faults are observed throughout the boreholes (Kelemen et al., 2020). Although fault zones are common in Holes GT3A and GT2A, we use only the major fault zones in Hole GT1A in this study to understand the role of major fault zones in hydration, alteration, and cooling of the ocean crust because this hole was located to sample such horizons (Kelemen et al., 2020) and has surface correlation with a major fault zone (Zihlmann et al., 2018). Fault zones have previously been shown to be associated with intense hydrothermal alteration (Bruhn et al., 1994; Zihlmann et al., 2018) and have been proposed as important conduits for fluid circulation in and cooling of the lower oceanic crust (Coogan et al., 2006; Harris et al., 2017). Using the depths of major fault zones recorded by the OmanDP core description teams (Kelemen et al., 2020), we compare  $H_2O$  concentrations for rock within fault zones and outside of major fault zones in Hole GT1A (Table 2; Figure 8) and find dramatic differences in hydration. Hydration is significantly higher in the major fault zones (mean  $H_2O$  = 5.7 wt%) compared to the regions outside of major fault zones (mean  $H_2O$  = 2.6 wt%) (Table 2). The median hydration of the fault zones is still high (4.7 wt%), indicating that the very high hydration pixels are common throughout the fault zones.

The high hydration within the fault zones of Hole GT1A correlates with the significant occurrences of alteration minerals, particularly amphibole and chlorite + prehnite + zeolite assemblages, detected by Greenberger et al. (2021). Additionally, the regions of Hole GT1A outside of major fault zones generally exhibit minimal alteration and low hydration, providing evidence that hydration and alteration of the lower ocean crust are driven by fault zones as conduits of hydrothermal fluid, highlighting the combined roles of physical fracturing and chemical alteration.

## 4. Discussion

### 4.1. Accuracy of Spectral Indices and Sources of Error

The infrared spectral index developed in this paper, band area of the 1,400–1,600 nm OH/ $H_2O$  absorptions calculated in wavenumber, displays a strong positive correlation with measured wt%  $H_2O$ . On average, the errors between the measured  $H_2O$  samples and the  $H_2O$  calculated by our model are of magnitude  $0.52 \pm 0.49$  wt%  $H_2O$  (Table S3 in Supporting Information S1). The  $R^2$  value for this regression (0.66) is only slightly lower than  $R^2$  values determined in other studies that use reflectance data. With their controlled grain size particulate mixtures of bright, water-bearing samples, Milliken and Mustard (2005), found  $R^2$  values ranging



**Figure 8.** Calculated H<sub>2</sub>O weight percent for pixels within the fault zones for GT1A versus those outside the major fault zones of GT1A. Major fault zones compose ~8.3% of Hole GT1A (Kelemen et al., 2020). The total number of pixels including in each of the histograms is given by n.

from 0.74 to 0.96 using methods including the mean optical path length, normalized optical path length, integrated band area, and effective single-particle absorption thickness for the OH fundamental absorption at ~2,900 nm. Our more complex, dark, multi-mineralic rock samples unsurprisingly have lower  $R^2$  values due to use of the weaker overtone and combination absorptions and variations in grain size, which can affect absorption. Nevertheless, the correlation between the area of the OH overtone absorption and the measured wt% H<sub>2</sub>O still provides means for effective tracking of spatial variability non-destructively and comprehensively at very fine scale.

There are a few instances where we more significantly overestimate or underestimate H<sub>2</sub>O. In the geochemical samples, ~5% of samples had low 1,400-nm band areas (<20) but higher LOI H<sub>2</sub>O measurements (>2 wt%) (Table S3 in Supporting Information S1). These 13 samples can be observed in Figure 3d. Our method therefore could result in the underestimation of the H<sub>2</sub>O content in some regions of the core. The weaker than expected OH absorption features in these samples are likely due to textural and mineralogical effects, such as the presence of darkening agents, particularly serpentinized, opaque, or dark colored minerals, that affect the depths of absorption features (Milliken & Mustard, 2007a).

One unavoidable source of error in the regression pertains to the way in which geochemical measurements were obtained. Samples for geochemical measurements including LOI were taken from the working half of the core, whereas the imaging spectroscopy data was obtained from the archive half (Kelemen et al., 2020). Although these two halves are close to mirror images of each other, they are not identical. In addition, geochemical measurements of LOI are three-dimensional measurements of a volume of rock crushed to powder that included material from below the surface of the split face of the working half. In contrast, imaging spectroscopy maps the hydration for a

two-dimensional measurement of the surface. The depth of penetration of light at the wavelengths of the imaging spectrometer is approximately tens to hundreds of micrometers (Hapke, 2012). Thus, the regression developed between integrated area of the 1.4- $\mu\text{m}$  band and the measurement of LOI actually compares measurements of slightly different materials. If a vein is present in the interior of the rock cut for geochemical measurements but not in the archive half of the core or vice versa, the calculated  $\text{H}_2\text{O}$  from spectra would and should differ from the volume measured. A second related inherent error in our data is the fact that the precise location of the ROIs is difficult to constrain. Through the use of the core logging tables and images of thin sections (Kelemen et al., 2020), we worked to locate as precisely as possible exactly where the samples were taken. However, the exact regions we defined as ROIs in the imaging spectroscopy data may be slightly offset from their actual location and differ in exact size and shape.

Although two LOI samples were excluded from the regression, these samples represent rock types that are uncommon in the ocean crust considered in this study. Consequently, their exclusion does not introduce significant sampling bias. Our methods work well for the analysis of gabbros and basalts, which are predominant rock types in the ocean crust. Both highly altered and unaltered sections of the ocean crust are included in this analysis, enabling a more complete understanding of ocean crust hydration. Samples with low wt%  $\text{H}_2\text{O}$  determined via LOI measurements also almost always have low  $\text{H}_2\text{O}$  as determined via spectroscopy, with errors as described previously, and the technique works well with higher  $\text{H}_2\text{O}$  samples, with the exception of a couple uncommon rock types, different from the calibration set.

Overall, this study has shown the value of microimaging spectroscopy as a tool to use in conjunction with conventional LOI methods. While LOI measurements do provide wt%  $\text{H}_2\text{O}$  data with high precision, they are time-consuming and destructive. Microimaging spectroscopy can be used to take millions or billions of measurements quickly and non-destructively. Although less precise, the average error of spectroscopic  $\text{H}_2\text{O}$  calculations in comparison with traditional LOI measurements was  $\sim 0.5$  wt%  $\text{H}_2\text{O}$ . In addition, microimaging spectroscopy can be performed on any sample and does not require complete core recovery or unharmed core. Furthermore, there are no biases related to sampling strategy. For different rock types and other core samples, a similar workflow could be used to obtain a regression equation relating integrated area via imaging spectroscopy to  $\text{H}_2\text{O}$  LOI measurements and then apply this equation to construct whole-core maps.

#### 4.2. Hydration of the Ocean Crust

The Oman ophiolite records hydrothermal alteration and metamorphism, mostly absent overprinting by weathering or obduction (e.g., A'Shaikh et al., 2006; Kawahata et al., 2001; Pflumio, 1991). Overall, we find that the  $\text{H}_2\text{O}$  content of the ocean crust at 250–260  $\mu\text{m}/\text{pixel}$  spatial resolution in the OmanDP core ranges between approximately 0 and 18 wt%  $\text{H}_2\text{O}$ , with the majority of the ocean crust containing 0.5–3 wt%  $\text{H}_2\text{O}$ . Hole GT2A (foliated and layered gabbros) is the most hydrated borehole in this study, with both the highest mean and median hydration values. Hole GT1A (layered gabbros) has a slightly lower mean  $\text{H}_2\text{O}$  than GT2A, but a much lower median  $\text{H}_2\text{O}$  (Table 1), suggesting that the majority of GT1A is low hydration, with a small volume of zones characterized by high hydration. Interestingly, despite being closest to the ocean floor, the uppermost Hole GT3A (dike-gabbro transition) has the lowest calculated mean and median  $\text{H}_2\text{O}$ . There are more very hydrated zones in GT1A compared to GT3A, driving the mean  $\text{H}_2\text{O}$  of GT1A higher than that of GT3A. The larger amount of high  $\text{H}_2\text{O}$  zones in GT1A is likely a result of the fact that Hole GT1A contains  $\sim$ twice as much high  $\text{H}_2\text{O}$  content zeolite as Hole GT3A, likely due to the presence of relatively more low temperature fluid circulation (Greenberger et al., 2021), and has extensive mineralized fault zones (Kelemen et al., 2020).

Overall, our results differ somewhat from existing estimates of oceanic crust  $\text{H}_2\text{O}$  content. Our results fall in the range measured in the Troodos Massif gabbros, where  $\text{H}_2\text{O}$  values ranged between 0 – 7.5 wt%  $\text{H}_2\text{O}$  (Moores & Vine, 1971). More recently, Kleine et al. (2020) estimated  $\text{H}_2\text{O}$  in altered basalts from the on-land section of the Mid-Atlantic Ridge ranging from 0.5 – 9.1 wt%. Using  $\delta\text{D}$  values measured in previous work (Agrinier, Hékinian, et al., 1995; Agrinier, Lavren & Tartarotti, 1995; Coogon et al., 2016; Kawahata et al., 1987; Kusakabe et al., 1989; Shilobreeva et al., 2011),  $\text{H}_2\text{O}$  has been estimated to be  $1.6 \pm 1.3$  wt% in the volcanic ocean crust,  $1.6 \pm 0.6$  wt% in the sheeted dikes, and  $0.8 \pm 0.1$  wt% in the gabbros (Kleine et al., 2020). Their average  $\text{H}_2\text{O}$  estimate in the sheeted dikes is similar to our study (2.1 wt% for GT3A). However, the mean values of 2.8 wt%

for GT1A and 3.2 wt% for GT2A are indicative of higher values for oceanic gabbro  $H_2O$  than those previously calculated. This is likely a result of the fact that the core recovery in previous ocean drilling is biased toward mostly unaltered sections of the core, whereas our whole core mapping includes crustal  $H_2O$  estimates for both the altered and unaltered sections of the ocean crust since onland diamond drilling provides  $\sim 100\%$  recovery of cored sections, both altered and unaltered. Previous IODP studies of ocean crust have generally suffered from lower core recovery. For example, Hole 1309 on the Atlantis Massif recovered an average of 75% of the core (Blackman et al., 2006). Similarly, Site U1473 on the Southwest Indian Ridge achieved an average of 59% core recovery (Macleod et al., 2017). Site 894 at Hess Deep (Leg 147) only achieved a core recovery of 27.5% in the upper mantle rocks (Früh-Green et al., 1996). Typical samples in previous studies focused on a single rock type and tended to exclude veins and other major structural features (e.g., Shilobreeva et al., 2011). In contrast, we include veins, fault zones, and damage zones in our calculations, and these features tend to have higher  $H_2O$  values as a result of their higher  $H_2O$  mineralogy and alteration, leading to higher hydration values. Thus, the range of  $H_2O$  values observed in our study is larger than ranges observed in previous studies, likely a result of our more complete sampling. In addition, we are able to spatially resolve variation at the  $\sim 250\ \mu m$  scale, which better enables our understanding of the processes that cause variations in hydration. Finally, though our mean and median values overlap across the cores, we note that our values are likely more accurate because we better capture the full range of alteration and lithologies present in the ocean crust. We also calculate hydration in the correct volumetric proportions because the highly altered material is not under-sampled relative to the less altered crust.

In comparing our results to previous  $H_2O$  measurements of ocean gabbroic core sections, we find that our results are similar. Results from IODP Site 1309 on the Atlantis Massif measured  $H_2O$  ranging from 0.93 wt% to 12.71 wt%, consistent with the ranges we observed in the OmanDP cores (Blackman et al., 2006). The gabbros in Site 1309 range from 1.20 wt% to 3.10 wt%  $H_2O$  (Blackman et al., 2006), which are similar to the means of 3.2 wt% for the foliated and layered gabbros (Hole GT2A) and 2.8 wt% for the lower layered gabbros (Hole GT1A) that were calculated in our study. At another site, samples from Hole 1473A on the Atlantis Bank in the Indian Ocean were found to contain 0.2–8 wt%  $H_2O$ , with a mean of  $1.0 \pm 0.9$  wt%  $H_2O$  (Macleod et al., 2017). In the nearby ODP Hole 735B, also on the Atlantis Bank of the Southwest Indian Ridge,  $H_2O$  contents range from 0–4 wt%  $H_2O$ , with a majority of the  $H_2O$  values falling between 0.5–2 wt%  $H_2O$  (Hertogen et al., 2002). Gabbroic rocks from Holes 894F-G at Hess Deep had LOI values ranging from 0.13–4.05 wt% (Lecuyer & Reynard, 1996). The overall ranges in these studies are broadly similar to our results, although each study only measured 99 to 451 samples.

Our results indicating that the lower gabbroic ocean crust is more hydrated than the sheeted dikes crust differ from previous estimates. However, we note that the lower sections of intact ocean crust formed at fast spreading rates, analogous to the Samail ophiolite, have not yet been sampled directly by ocean drilling. We attribute our results to two factors. First, the lower gabbroic ocean crust has experienced modestly less alteration than the upper sheeted dikes and gabbros, but the alteration phases formed at deeper depths have much higher wt%  $H_2O$ . Specifically, hydrothermal alteration processes that form large volumes of zeolite minerals are an important carrier of  $H_2O$  in the lower ocean crust. While the sheeted dikes and uppermost gabbros of the ocean crust (Hole GT3A) experienced more intense alteration, the greenschist facies alteration resulted in minerals with lower  $H_2O$  content, such as amphibole, and formed fewer high  $H_2O$  minerals than Holes GT1A and GT2A (Greenberger et al., 2021). Given that high frequencies of prehnite and zeolite are observed in GT2A and GT1A, which are deeper in the ocean crust, penetration of hydrothermal fluid is likely the cause of the lower temperature alteration occurring deeper in the ocean crust, as previously suggested (e.g., Alt et al., 2010; Dygert et al., 2017; Garrido et al., 2001; Hanghøj et al., 2010; Heft et al., 2008; Wilson et al., 2006; VanTongeren et al., 2008).

Second, throughout the lower gabbroic oceanic crust, veins and fault zones are present (see full maps in Files S1–S2 in Supporting Information S1), and these zones have substantially increased hydration (Table 2; Figure 8). Faulting may enable fluid flow, which would increase alteration, and thus hydration, within and surrounding faulted areas. By design, a significant portion of the core from Hole GT1A sampled fault zones ( $\sim 8\%$ ) that appear to be oceanic in origin (Kelemen et al., 2020; Zihlmann et al., 2018), though we note that additional later fault zones generated during obduction might also be contributing to hydration, unrelated to seafloor processes. Similarly, Coogan et al. (2006) estimated that such fault zones in the same block of the Samail ophiolite have 10–50 m widths with a spacing of  $\sim 1$  km, thus comprising  $\sim 1$  to 5 volume percent of the lower crust. Our results suggest that fault zones provide conduits for channeled fluid flow and locations for increased hydration and alteration

within the deep ocean crust, supporting previous predictions. Because fault zones appear to be a major driver of hydration within the ocean crust, hydration of any particular locale may be dependent on the local faulting and structure of the ocean crust in the region (Bruhn et al., 1994).

In applying these results to “typical” oceanic crust formed at fast-spreading ridges, several caveats should be considered. First, although the primitive lavas that formed the ophiolite had major element contents very similar to those of mid-ocean ridge basalts, trace element and geochronological data indicate that the crust of the Samail ophiolite formed via crystallization of hydrous magmas above a subduction zone (e.g., MacLeod et al., 2013; Pearce et al., 1981; Rioux et al., 2016). Second, to the extent that the average water contents of core from Hole GT1A are influenced by the presence of highly altered fault zones, readers should keep in mind that the frequency and width of such zones have not yet been fully quantified for the Samail ophiolite, and are almost unknown for lower oceanic crust formed at intermediate-to fast-spreading, Pacific crust. Third, while OmanDP investigators have, like prior investigators of the Oman ophiolite (e.g., A'Shaikh et al., 2006; Kawahata et al., 2001; Pflumio, 1991), inferred that most of the low temperature, high  $H_2O$ -content alteration minerals in the core (i.e., zeolites) formed by water-rock reactions at or near the Samail spreading center, we did not do independent work to fully rule out the alternative that some of these minerals may have formed during or post-obduction.

High proportions of hydrous minerals within the crust influence the physical properties of the ocean crust. For example, the presence of faults, fractures, and damage zones have been demonstrated to impact seismic wave propagation (Berryman, 2007). The high levels of hydration in the lower oceanic crust and its fault zones relative to the rest of the crust may be related to the relatively low observed seismic wave speeds observed for oceanic lower crust, as compared to calculated wave speeds in fresh gabbro lithologies (Behn & Kelemen, 2003; Korenaga et al., 2002). Our data on hydration as a function of depth can be used to update interpretations of seismic data for the ocean crust. In particular, the seismic wave speeds of zeolites have rarely if ever been considered in this context.

## 5. Conclusions

We quantified hydration of the ocean crust in drill core from the Samail ophiolite by developing a regression between geochemical measurements of LOI and spectral indices of  $OH/H_2O$  utilizing data from shortwave infrared micro-imaging spectroscopy of ICDP OmanDP Holes GT3A (sheeted dikes and upper gabbros), GT2A (middle ocean crust; foliated and layered gabbros), and the GT1A (lower ocean crust; layered gabbros). The integrated area of the  $OH/H_2O$  absorptions at  $\sim 1,350$ – $1,650$  nm in wavenumber determined  $H_2O$  wt. % concentrations with  $R^2 = 0.66$  and a typical absolute accuracy of  $0.52 \pm 0.49$  wt%  $H_2O$ . We applied our regression to each  $\sim 250$ – $260$   $\mu m$  pixel in 1.2 km of rock core, comprising 1.07 billion pixels, to produce spatially resolved maps of ocean crust hydration.

Our results indicate that the majority of the ocean crust contains 0.5–3 wt%  $H_2O$ , and that overall ocean crust hydration ranges from 0 to  $\sim 18$  wt%  $H_2O$ . Pixels sampling material with high  $H_2O$  occur where there is a higher prevalence of minerals with high  $H_2O$  contents (e.g., zeolites) while lower  $H_2O$  areas contain primary anhydrous mineralogy (pyroxene and plagioclase) and/or minerals with lower  $H_2O$  contents, such as amphibole. Hydration does increase to a certain degree with extent of alteration, as expected, but the nature of the mineral phases formed as a result of alteration is an important determinant of bulk hydration of sections of the crust. Somewhat counterintuitively, we find the overall less altered lower crust is, in bulk, more hydrous by 1–2 wt. %, mainly due to the ubiquitous presence of  $H_2O$ -rich zeolites, together with localized, mineralized fault zones. The lowest mean  $H_2O$  (2.1 wt%) is in Hole GT3A, which experienced the most extensive alteration. Its secondary mineralogy is dominated by amphibole and epidote, relatively low wt%  $H_2O$  minerals, along with some chlorite. The mean  $H_2O$  wt. % contents in Holes GT2A (3.2 wt. %) and GT1A (2.8 wt. %) were considerably higher. Fault zones serve as conduits for the enhanced hydration of the lower ocean crust; gabbroic GT1A has relatively high levels of  $H_2O$  in fault zones (5.7 wt%), which are 2 $\times$  as hydrated as the non-faulted regions of Hole GT1A (2.6 wt. %  $H_2O$ ).

Overall, our ability to use the imaging spectroscopy data set of the entire core, for a core with near 100% recovery, enables us to quantify  $H_2O$  in both the unaltered sections of the oceanic crust and the more altered sections that have been excluded from other studies, ultimately increasing estimates of bulk crustal hydration relative to previous work. The higher estimates for hydration of the lower ocean crust have implications for its rheologic properties in other processes.

## Appendix A

### Phase 1

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## Data Availability Statement

The LOI measurements used in this study were published in Kelemen et al., (2020), and data are available from ICDP. The underlying imaging spectroscopy data set used is archived through CaltechDATA (Greenberger et al., 2021, <http://dx.doi.org/10.22002/D1.2009>). H<sub>2</sub>O maps of each core section are also available as PDF images (Crotteau et al., 2021a, <http://dx.doi.org/10.22002/D1.2141>) and .img files through CaltechDATA (Crotteau et al., 2021b, <http://dx.doi.org/10.22002/D1.2142>). Regions of interest corresponding to the LOI samples are also archived on CaltechDATA (Crotteau et al., 2021a, <http://dx.doi.org/10.22002/D1.2141>).

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