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**Electrical conductivity of omphacite and garnet indicates limited  
deep water recycling by crust subduction**

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## Abstract

It is usually stated that oceanic crust recycling at subduction zones introduces large quantities of water into Earth's interior. However, it remains clouded how much water is recycled to the deep mantle. This is largely due to the challenge in directly sampling, and analyzing the water content of, a deep subducting crust. The subducting crust below ~30 km depth is dominated by omphacite and garnet in eclogite-facies rocks. Here we have determined the electrical conductivity of omphacite and garnet in representative subduction-related eclogites, each with varying contents of Fe and H<sub>2</sub>O that are key in electrical conduction. Considering the measured conductivity, the eclogite chemistry by geochemical investigations and the highly resistive property of subducting crusts by geophysical surveys, we demonstrate that, at 70-120 km depths in the subducting crust, the H<sub>2</sub>O contents of omphacite and garnet are strikingly small, with the maximum value being <400 and <80 ppm in the former and latter, respectively. The very small water contents indicate extremely water-poor conditions, or very low water activity, during the eclogite-facies metamorphism and in the system. This further implies the absence of appreciated amounts of hydrous phases such as amphibole and chlorite in the matrix, because of the strong ability of omphacite and garnet in hosting water as documented in natural samples. We suggest that the recycling of water to the deep mantle by oceanic crust subduction is limited. The results are important for modeling the conductivity of subducting slabs and understanding the deep water circulation.

## 1. Introduction

The transport of water from the hydrosphere to the mantle plays a critical role in affecting mantle dynamics, magma genesis and Earth's habitability. Water is recycled back to the mantle by slab subduction. At very shallow depths (e.g., <20 km), the input

of water into the mantle is primarily controlled by pore water (Peacock, 1990; Cai et al., 2018). In a downgoing slab at greater depths, however, water chiefly occurs as OH groups bound in the structure of the constitutive minerals, both hydrous and nominally anhydrous, note that H<sub>2</sub> dissolution in minerals is significant only at very high pressure and reducing conditions (Yang et al., 2016). As a slab descends, water is gradually lost by the breakdown of hydrous minerals such as amphibole and chlorite at depths mostly <100 km, due to their instabilities at high temperature (Peacock, 1990; Liu et al., 1996; Frost, 2006; Schmidt and Poli, 2014). Driven by its low density, the released water rises, metasomatizes wallrocks and causes melting in the wedge. This causes the development and widespread distribution of subduction zone volcanism and ore deposits.

It is widely believed that the crust layer in the slab is important for water recycling, because most of the slab water is initially stored there (Ito et al., 1983; Peacock, 1990; Dixon et al., 2002; Van Keken et al., 2002; Schmidt and Poli, 2014). Beyond ~30 km depth, the slab crust is dominated by eclogites (via phase change), consisting mainly of nominally anhydrous omphacite and garnet that are the main water carriers in eclogites after hydrous minerals decompose (Lu and Keppeler, 1997; Bromiley and Keppeler, 2004; Schmidt and Poli, 2014). As such, the water contents of omphacite and garnet are key to understanding water recycling to the deep mantle. Omphacite and garnet in terrane eclogites, that were once subducted to mantle depths and then returned to surface, could hold up to several thousands of ppm H<sub>2</sub>O as structural OH, leading to the suggestion that the subducting crust is able to carry large quantities of water into the deep mantle beyond the stability of the hydrous phases (Katayama and Nakashima, 2003; Katayama et al., 2006; Sheng et al., 2007). The high water contents of omphacite and garnet in the eclogites may, however, have suffered from complex secondary fluid-rock interactions postdating eclogite peak-metamorphism (e.g., Sheng et al., 2007; Schmädicke and Gose,

2017). It is thus obscure how much water is present in omphacite and garnet in the deep subducting crust.

The electrical conductivity of nominally anhydrous minerals (NAMs) is sensitive to water (Karato, 1990; Dai and Karato, 2009; Yoshino et al., 2009; Yang et al., 2011, 2012; Yang and McCammon, 2012; Zhang et al., 2012, 2019; Zhao and Yoshino, 2016; Liu et al., 2019). Meanwhile, electromagnetic surveys offer a direct window into Earth's interior. In this context, constraints can be provided on the water contents of omphacite and garnet in the subducting crust (and water recycling), if the conductivity structure of subducting crusts is resolved and the electrical property of subduction-related materials is determined. In a recent work, Liu et al. (2019) have quantified the effect of water on the conductivity of omphacite and garnet, and provided a preliminary constraint on the likely water content in the subducting crust. However, Liu et al. have not evaluated the effect of Fe, which along with H are crucial in electrical conduction, and the question arises how convincing their results might be applied to Earth's interior. In this report, we have by experimental work measured the conductivity of omphacite and garnet from representative eclogites, which plus the samples of Liu et al. (2019) show a wide range of contents in Fe and H<sub>2</sub>O. The data are used to infer the water contents of the minerals in the subducting crust, by combining with geophysically-based electrical structure and slab petrology. The results demonstrate clearly that a small amount of water is present in omphacite and garnet in deep subducting crusts.

## **2. Experiments and Methods**

### **2.1 Starting materials and sample characterization**

Three starting eclogites were from Bixiling (omp1 + grt1) and Shuanghe (omp3 + grt3) in China and Weißenstein (omp2 + grt2) in Germany, and were all related to slab subduction and exhumation (Franz et al., 1986; Sheng et al., 2007). The Weißenstein

eclogite was studied in Liu et al. (2019). These samples were chosen by considering Fe contents of constituting omphacite and garnet and quality of grains from >30 eclogites (*note*: the electrical property of silicates is independent of the origin). Natural eclogites were not used for conductivity studies directly, because of the occurrence of accessory materials (e.g., secondary hydrous phases, fluid inclusions and/or other impurities) and zoned water distribution that affect conductivity analyses. We analyzed the conductivity of omphacite and garnet separately and then modeled the bulk conductivity. Omphacite and garnet are representative in composition, in particular in Fe that almost covers the content ranges in subduction-involved eclogites (omp1 and omp3: Appendix). Optically clear omphacite/garnet grains, ~100-700  $\mu\text{m}$  in size, were handpicked under a binocular microscope. Each sample is chemically homogeneous in major-/minor-elements (Table 1), as measured by a JEOL JXA-8230 electron microprobe (15 kV accelerating voltage, 10 nA current and <5  $\mu\text{m}$  beam size). Grains were ground to powder, mostly 30-80  $\mu\text{m}$ , and H-annealed with minor distilled water in Ni capsules (ID 4.2 mm, OD 5.0 mm and length 10 mm) at 1-3.5 GPa and 650-700  $^{\circ}\text{C}$  (150-180 h duration) in a piston cylinder press. By this, water homogenization was achieved in each sample, and samples with different water contents were prepared. Dry samples were not prepared, because OH is common in natural omphacite and garnet. Recovered samples were cored into cylinders of 3 mm diameter and 1.7 mm length for subsequent conductivity runs, note that the conductivity of silicate minerals by lattice conduction is independent of grain size above ~5  $\mu\text{m}$  that is typical for minerals in the deep Earth (Yang and Heidelbach, 2012).

A precise analysis of water content is key for evaluating its effect on conductivity. The general principle for water quantification in silicate minerals by Fourier-transform infrared (FTIR) spectroscopy is given by the modified Beer-Lambert law:  $C_{\text{H}} = \text{Abs}_{\text{total}}/I$ ,

where  $C_H$  is OH content,  $Abs_{total}$  is the total OH integrated absorbance normalized to 1 cm thickness, and  $I$  is the mineral-specific integral molar absorption coefficient. As such,  $C_H$  is determined by  $Abs_{total}$ , once the coefficient  $I$  is externally calibrated. For optically anisotropic minerals, the intensity of OH bands is sensitive to the orientation of the IR active dipole relative to incident beam, and  $Abs_{total}$  is the sum of thickness-normalized integrated absorbance along the three principal axes ( $a$ ,  $b$  and  $c$ ). This requires orienting samples and polarized FTIR analyses along the axes, which are difficult for small grains and are time-consuming for sample preparation. Recently, Shuai and Yang (2017) have shown that  $Abs_{total}$  can be well characterized by polarized FTIR analyses along any three mutually perpendicular directions of a given sample, without the necessity to orient the samples; in case very small grains which are unable to be prepared for polarized FTIR work, Qiu et al. (2018) have demonstrated that  $Abs_{total}$  can be obtained by unpolarized FTIR analyses on randomly oriented grains.

Water contents were measured by a Bruker Vertex 70V FTIR spectrometer coupled with a Hyperion 2000 microscope. Analyses were performed on optically clear grains with an aperture of  $60 \times 60 \mu\text{m}$  and a resolution of  $4 \text{ cm}^{-1}$  (128 scans for each spectrum by a global source, KBr-Ge beam splitter, MCT detector and wire-grid Ze-Se polarizer). For anisotropic omphacite, polarized spectra were acquired for relatively large grains in annealed samples before conductivity runs (Shuai and Yang, 2017), and unpolarized spectra on 15-20 randomly oriented grains were obtained for fine-grained samples after conductivity runs (Qiu et al., 2018). For cubic garnet, unpolarized spectra were recorded over 6-9 grains. Water contents were calibrated by the mineral-specific coefficients of Katayama et al. (2006) for omphacite and garnet in eclogites, and uncertainty is mostly  $<10\%$ , as documented in Shuai and Yang (2017) and Qiu et al. (2018). For a comparison under the same framework, the same calibration was used to recalculate the  $\text{H}_2\text{O}$  content

of minerals in natural eclogites in available studies. This yields values different from some early work, e.g., the 1840 ppm OH of omphacite in Smyth et al. (1991), estimated by the OH peak linear intensity, is actually ~310 ppm H<sub>2</sub>O; in contrast, the H<sub>2</sub>O contents of omphacite and garnet in recent reports (Katayama and Nakashima, 2003; Katayama et al., 2006; Sheng et al., 2007; Schmädicke and Gose, 2017) are less affected. The general results of this work are not affected by adopting the calibration coefficients.

## **2.2 Conductivity experiments at elevated conditions**

Slabs are rather oxidizing above ~200 km depth, with redox state similar to the Ni-NiO buffer (McCammon, 2005). Therefore, conductivity runs were carried out with Ni-NiO at 1-2.5 GPa and 250-850 °C in an end-loaded piston cylinder press. The assembly design resembles that in our previous studies (Yang et al., 2011, 2012; Yang, 2012; Yang and Heidelbach, 2012; Yang and McCammon, 2012; Li et al., 2016, 2017; Liu et al., 2019), and only a brief description is offered. In each run, a BN-Ni double capsule was adopted to maintain sample geometry and yield a relatively sealed chamber. BN causes reducing conditions only when O<sub>2</sub> is rich (e.g., in air) and run duration is long above ~1000 °C, by  $4\text{BN} + 3\text{O}_2 = 2\text{B}_2\text{O}_3 + 2\text{N}_2$ , and BN itself in a sealed system does not influence the redox state. It has been demonstrated that, for the design at <1000 °C and short duration (typical of conductivity runs), the reaction of Pt and Fe in samples is negligible and the redox state in the chamber is well buffered, as tested with pyroxenes and olivine (Yang et al., 2011, 2012; Yang, 2012; Yang and McCammon, 2012).

Before assembly, Al<sub>2</sub>O<sub>3</sub> parts were heated at 1000 °C to remove absorbed water, and during assembly, no cement/glue was used to immobilize the various parts to avoid volatile release at high temperature that affects the analyses. Completed assembly was heated at 136 °C overnight, and was heated again at 200 °C after loading into the press. Impedance spectra were recorded in the heating-cooling cycles, with a Solartron 1260



Impedance/Gain Phase analyser by frequency sweeping at  $10^6$ -1 Hz and 0.5 V applied voltage. Temperature of the analyses was up to 850 °C, to minimize water diffusion loss of samples upon heating. Duration was typically ~1 min or less per analysis (depending on temperature). After each run, recovered capsule was polished for optical, FTIR and backscattered electron examinations. Ni-NiO pairs were present, and sample distortions and grain growths were negligible, as observed in similar work (Yang et al., 2011, 2012; Yang, 2012; Yang and Heidelbach, 2012; Yang and McCammon, 2012; Li et al., 2017; Liu et al., 2019). Resistance ( $R$ ) was obtained from the impedance spectra (see below), and conductivity ( $\sigma$ ) was calculated by  $\sigma = L/SR$ , where  $L$  and  $S$  are the effective sample length and cross-section area, respectively. Uncertainty is usually <5% for conductivity and <20 °C for temperature.

### 3. Results

Representative FTIR spectra of annealed samples are shown in Fig. 1. The spectra show absorption peaks at 3610-3625, 3500-3520 and 3440-3460  $\text{cm}^{-1}$  in the omphacites and at 3610-3640 and 3530-3550  $\text{cm}^{-1}$  in the garnets, which are typical of OH bands in the eclogite minerals (Smyth et al., 1991; Katayama and Nakashima, 2003; Katayama et al., 2006; Sheng et al., 2007; Schmädicke and Gose, 2017). In each mineral, the peak frequency positions are similar, suggesting similar H incorporation mechanism, and the relative difference of peak intensity between different samples are due to different water contents and/or grain orientations. Sample  $\text{H}_2\text{O}$  contents are basically the same prior to and after each conductivity run (Table 2). Fe and other major-/minor-elements are more sluggish than H, and their contents are unchanged in the runs. Representative complex spectra are presented in Fig. 2. The spectra usually show a high frequency arc and a low frequency tail, although the tail is sometimes not obvious, and the spectral shapes are similar to those reported for other minerals (Dai and Karato, 2009; Yoshino et al., 2009;

Yang et al., 2011, 2012; Yang, 2012; Zhang et al., 2012, 2019; Zhao and Yoshino, 2016; Li et al., 2016, 2017; Liu et al., 2019). The arc and tail was by sample lattice conduction and electrode effects, respectively.

The conductivity of NAMs (and other Earth materials) is quantitatively described by the Arrhenius relation:

$$\sigma = \sigma_0 \cdot \exp(-\Delta H / RT) \quad (1)$$

where  $\sigma_0$  is a constant,  $\Delta H$  is the activation enthalpy,  $R$  is the ideal gas constant, and  $T$  is temperature. Conductivities in the first heating were sometimes affected by residual moisture in the chamber, yielding data not obeying Eq. (1) as observed in similar studies (Yang et al., 2012; Yang and Heidelbach, 2012; Li et al., 2017; Liu et al., 2019), and the influenced data were excluded. Conductivity data and the fittings to Eq. (1) are shown in Fig. 3, and fit parameters are given in Table 2. Sample conductivity is much greater than the assembly background conductivity (Yang et al., 2011). The data are consistent between different heating-cooling cycles in each run (see also Appendix), suggesting the absence of system hysteresis. The conductivity is slightly larger at 2.5 than at 1 GPa, indicated by the runs with an OH-bearing omphacite (omp3). This is consistent with the previous study on an OH-bearing pyrope that shows similar conductivity enhancement by pressure (Dai and Karato, 2009). In contrast, the conductivity of dry olivine or garnet decreases slightly with increasing pressure (Xu et al., 2000; Dai and Karato, 2009). The difference is caused by the different pressure dependence between small polaron and H conduction in the minerals (see Section 4), but the effect of pressure (at a small variation) on the conductivity of silicate minerals is in general insignificant.

#### **4. Fe- and H-dependent conductivity**

In Fe- and H-bearing NAMs, the conductivity is related to the conduction of small

218 polarons (electron-hole hopping between  $\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ ) and protons (H). Our data show  
 219 that, at otherwise identical conditions, the conductivity of both omphacite and garnet  
 220 increases with Fe and  $\text{H}_2\text{O}$  contents (Fig. 3), agreeing with studies on other NAMs (Dai  
 221 and Karato, 2009; Yoshino et al., 2009; Yang et al., 2011, 2012; Yang and McCammon,  
 222 2012; Zhang et al., 2012; Zhao and Yoshino, 2016). The data trends of our omphacites  
 223 are consistent with those of synthetic omphacites at 3 GPa (Zhang et al., 2019), and the  
 224 conductivity of the omphacite (omp3) with 7.92 wt.% FeO and 70 ppm  $\text{H}_2\text{O}$  is close to  
 225 that of an augite with 8.31 wt.% FeO and 75 ppm  $\text{H}_2\text{O}$  at 1 GPa (Yang and McCammon,  
 226 2012) (Fig. 3a). The conductivity of the omphacite (omp1) with 1.46 wt.% and 200 ppm  
 227  $\text{H}_2\text{O}$  resembles that of the one (omp2) with 4.27 wt.% and 150-290 ppm  $\text{H}_2\text{O}$ , implying  
 228 an insignificant effect of Fe at that content range. At a similar 155-160 ppm  $\text{H}_2\text{O}$ , the  
 229 conductivity of the Fe-rich garnet (15.40 wt.% FeO) is less than that reported by Dai  
 230 and Karato (2009) for a Fe-poor pyrope (7.42 wt.% FeO) at 8 GPa (Fig. 3b). Probably,  
 231 this is related to the different pressures (1 vs. 8 GPa) and H-species (note that different  
 232 H-defects could have different mobility, contributing differently to the conduction), e.g.,  
 233 narrow OH bands at  $3700\text{-}3500\text{ cm}^{-1}$  in this study vs. broad bands at  $3800\text{-}3000\text{ cm}^{-1}$  in  
 234 Dai and Karato (2009). The conductivity of our garnets is close at similar contents of  
 235 FeO (26.55-26.58 wt.%) and  $\text{H}_2\text{O}$  (100-110 ppm, with similar OH band shapes, Fig. 1).  
 236 The activation enthalpy is Fe-dependent in omphacite,  $\sim 87$ , 81-84 and 65 kJ/mol at 1.64,  
 237 4.27 and 7.92 wt.% FeO, respectively, but is broadly Fe-independent in garnet, 88-91  
 238 kJ/mol over a large range of Fe contents, 15.40-26.58 wt.% FeO (Table 1). Therefore,  
 239 the effect of Fe on the conductivity of omphacite and garnet is mineral dependent.

240 For the same omphacite or garnet in eclogite samples under similar conditions, the  
 241 conductivity is linearly proportional to water content below  $\sim 1000^\circ\text{C}$  and at moderate  
 242  $\text{H}_2\text{O}$  contents (Liu et al., 2019; Zhang et al., 2019). This has also been reported for many

other NAMs such as augite, diopside, enstatite, plagioclase, pyrope and olivine (Yang et al., 2011, 2012; Yang and McCammon, 2012; Zhang et al., 2012; Zhao and Yoshino, 2016). For an OH-bearing mineral with a fixed content of Fe (and other major elements), the relative contribution of small polaron conduction is usually much smaller than that of proton conduction at relatively low temperature. Eq. (1) can be approximated by:

$$\sigma \propto C_H \cdot \exp(-\Delta H_H / RT) \quad (2)$$

where  $\Delta H_H$  is the activation enthalpy by H conduction and is numerically equal to  $\Delta H$  in Eq. (1). As such, the conductivity of both omphacite and garnet can be modeled for a range of H<sub>2</sub>O contents at each of the starting FeO contents (Fig. 4). The essentially same conductivity of omphacite with 1.64 and 4.27 wt.% FeO at the same H<sub>2</sub>O content, as noted above, indicates a more significant role of H than Fe at such Fe content levels. The N-type semiconductor relation is used to model the effect of both Fe and H on the conductivity of both minerals:

$$\sigma = A \cdot X_{Fe}^n \cdot C_H \cdot \exp[-(\Delta E - \alpha X_{Fe}^{1/3} - \beta C_H^{1/3}) / RT] \quad (3)$$

where  $X_{Fe}$  is the molar fraction of Fe (= Fe/(Fe + Mg)),  $\Delta E$  is the activation energy, and  $A$ ,  $n$ ,  $\alpha$  and  $\beta$  are constants. The insensitivity of omphacite conductivity to FeO content at 1.64-4.27 wt.% makes it hard to model the whole dataset with Eq. (3). In this context, the modeling is carried out for omphacite using the conductivity data of our samples starting with omp1 and omp3, that restrict the boundaries of Fe contents in subduction-related omphacites, and the synthetic omphacite (5.81 wt.% FeO and 420 ppm H<sub>2</sub>O with similar OH band positions) of Zhang et al. (2019), and for garnet using the data of all our samples (the Dai and Karato (2009) pyropes are not used because of the different OH bands). This yields  $\Delta E$ ,  $A$ ,  $n$ ,  $\alpha$  and  $\beta$  of  $106 \pm 6$  kJ/mol,  $2754 \pm 380$  S/m,  $-0.05 \pm 0.09$ ,  $64 \pm 6$  kJ/mol and  $-26 \pm 8$  kJ/mol for omphacite, and  $89 \pm 3$  kJ/mol,  $61660 \pm 4085$  S/m,  $4.25 \pm 1.32$ ,  $-4 \pm 31$  kJ/mol and  $15 \pm 9$  kJ/mol for garnet. For both minerals, the modeled

results by Eq. (2) and (3) are well consistent at the same H<sub>2</sub>O and boundary Fe contents, especially at >500 °C (Fig. 4).

The bulk conductivity of eclogite is calculated from omphacite and garnet with the Hashin and Shtrikman (1962) bounds, which offer the narrowest restrictions of a two-phase composite despite the geometrical configurations:

$$\begin{aligned}\sigma_{HS+} &= \sigma_1 + x_2[(\sigma_2 - \sigma_1)^{-1} + x_1 / (3\sigma_1)]^{-1} \\ \sigma_{HS-} &= \sigma_2 + x_1[(\sigma_1 - \sigma_2)^{-1} + x_2 / (3\sigma_2)]^{-1}\end{aligned}\tag{4}$$

where  $\sigma_{HS+}$  and  $\sigma_{HS-}$  are the upper and lower bounds, respectively, given  $\sigma_1 > \sigma_2$  ( $x$  is the volume proportion). A key prerequisite for the modeling is the equilibrium distribution of major/minor elements and OH between coexisting minerals. This is a factor that must be considered when modeling the bulk conductivity of any matrix consisting of several phases. The calculation is carried out by Eq. (2) using the conductivity data of minerals at different H<sub>2</sub>O contents for each starting Fe content, in which case the effect of Fe is constrained at the same time (Fig. 4). Eq. (3) is not adopted, because of the difficulty in controlling chemical equilibrium between the two minerals. Each of the mineral pairs, omp1-grt1, omp2-grt2 and omp3-grt3, was separated from the same starting eclogite, and the major/minor elements are already in equilibrium. The water partition coefficient between omphacite and garnet in eclogite, ~0.2 from Katayama et al. (2006), is adopted to obtain the equilibrium H<sub>2</sub>O contents (assuming 100, 200 and 400 ppm in omphacite). The bulk conductivity of eclogite is not affected by likely minor impurities, because if present, they are usually isolated (not connected) in the matrix and do not contribute to bulk conduction. The results are shown in Fig. 5a and b for omphacite-rich and garnet-rich rocks, respectively. This produces the modeled bulk conductivity at variable Fe and H<sub>2</sub>O contents and high temperature. In particular, The FeO contents reflect the range of the two minerals in subducting crusts (as noted before). Given other conditions,  $\sigma_{HS+}$

and  $\sigma_{HS}$  of each sample show a small variation above 600 °C, and are up to ~0.1 S/m at 900 °C (for the modeled composition). Considering the positive pressure effect on the conductivity of OH-bearing omphacite and garnet as mentioned above, the bulk conductivity is only slightly greater at higher pressure.

## **5. Water contents in subducting omphacite and garnet**

Electromagnetic induction is sensitive to conductive rather than resistive bodies in Earth's interior. This makes it hard to accurately resolve the conductivity of subducting crusts that are highly resistive. Usually, high conductivity is not detected in subducting slabs, and slab conductivity is much smaller than that of mantle wedge and surrounding mantle in the inversion results. Conductivity of the subducting crust, either fast or slow subduction in diverse settings, is estimated to be mostly  $10^{-4}$  to  $10^{-3}$  S/m at ~70-120 km depth (Vanyan et al., 2002; McGary et al., 2014; Ichiki et al., 2015; Vargas et al., 2019). This is a reasonable estimate by imposing a 1000, 5000 or 10000  $\Omega$  slab resistivity in the inversion models (Ichiki et al., 2015). A typical electrical structure of a subducting crust is given in Fig. 5c (after McGary et al. (2014) who focused on conductive channels in the wedge). We want to note that, although Evans et al. (2014) have offered a variety of outputs (mainly in the wedge) with the same electromagnetic data of McGary et al. (2014) by applying different inversion methods, the crust portion at ~70-120 km depth is very resistive in most of their models. Moreover, Fig. 5c is for the Cascadia area, but broadly similar results on resistive slab vs. depth have been reported for other tectonic environments (e.g., Vanyan et al., 2002; Ichiki et al., 2015; Vargas et al., 2019).

It has been shown that, despite a wide range of ages, geometries and convergence rates, slabs actually share many common features with their thermal structures (Peacock, 1996; Syracuse et al., 2010; Penniston-Dorland et al., 2015). At ~70-120 km depth, the temperature is mostly ~600-900 °C in the crust layer, for subduction zones over various

314 tectonic settings. The FeO content is ~1-8 wt.% of omphacite and 14-27 wt.% of garnet  
315 in terrane eclogites that were once brought to ~200 km depth (Katayama and Nakashima,  
316 2003; Katayama et al., 2006; Sheng et al., 2007; and Appendix), suggesting that deeply  
317 subducting eclogites are in fact not depleted in Fe. As such, to cause the  $10^{-4}$ - $10^{-3}$  S/m  
318 conductivity of subducting crusts at ~600-900 °C, the only possibility is that omphacite  
319 and garnet are water-poor. According to the modeling in Fig. 5a-b, the H<sub>2</sub>O contents are  
320 <400 ppm in omphacite and <80 ppm in garnet at 600 °C and <100 ppm in omphacite  
321 and <20 ppm in garnet at 900 °C for the bottom Fe boundaries (omp1-grt1), and <30  
322 ppm for the top Fe boundaries (omp3-grt3). These H<sub>2</sub>O-content ranges are meaningful,  
323 in particular if considering the weaker effect of a minor variation in Fe than in H content  
324 (Section 4), and can be even smaller, if the effect of pressure on the conductivity of OH-  
325 bearing omphacite and garnet is considered (Section 3). The maximum <400 ppm H<sub>2</sub>O  
326 content resembles that estimated in Liu et al. (2019), which is based on omp2 in contrast  
327 to omp1 in this study (due to the more significant effect of H at the Fe content levels as  
328 noted above). The inferred H<sub>2</sub>O contents of omphacite and garnet in subducting crusts  
329 (~70-120 km depth) are much less than those up to 2000-3000 ppm in terrane eclogites  
330 (Katayama and Nakashima, 2003; Katayama et al., 2006). This suggests that OH-rich  
331 omphacite and garnet in the terrane samples were H-enriched by secondary processes  
332 during their exhumation to surface, in line with the arguments by analyzing mineral OH  
333 patterns and D/H ratios (Sheng et al., 2007; Schmädicke and Gose, 2017). Interestingly,  
334 the H<sub>2</sub>O values resemble the mostly <360 ppm of omphacite and <65 ppm of garnet in  
335 volcano-hosted xenolith eclogites (Smyth et al., 1991; Bell and Rossman, 1992; Koch-  
336 Müller et al., 2004). This makes more sense if xenolith eclogites represent the various  
337 dehydrated products of subducted crusts that were accidentally entrained by volcanoes  
338 and transferred to the surface; however, things could be complicated because a general

consensus has not been reached for the origin of xenolith eclogites, e.g., some may form by melt accumulation (Jacob, 2004).

Consequently, the conductivity data and inferred water contents of omphacite and garnet are broadly consistent with the constraints from other approaches. The inversion of electromagnetic data at subduction zones has long been subjected to the difficulty in adopting a good reference resistor in the starting model. In most cases, the inversion is carried out by highlighting the conductivity contrast between the resistive slab and the conductive paths in the wedge (linked to melting and/or dehydrated fluids). In order to identify fine-scale variation of conductivity in the subducting slab, e.g., due to change of temperature and release of water with subduction, it is important to impose a reliable slab conductivity in the inversion model. The water contents of minerals in the slab, the electrical properties of eclogites and the conductivity laws, as provided here, allow to model the conductivity of oceanic crusts as a function of temperature and water content as the starting reference resistor for the inversion of electromagnetic data.

## **6. Implications for deep water recycling**

The very low water contents of omphacite and garnet at ~70-120 km depth imply a much smaller water content in the subducting crust than the original value dominated by hydrous phases (e.g., amphibole and chlorite), e.g., ~1-2 wt.% H<sub>2</sub>O (Peacock, 1990). Water in the subducting crust is mostly released at shallow depths, and the dehydration of hydrous phases and omphacite and garnet in eclogites produces conductive channels in the wedge (McGary et al., 2014). Eclogite-facies metamorphism, which involves all materials in the system, occurs at >30 km depths in the subducting crust (e.g., Peacock, 1990; Schmidt and Poli, 2014). The inferred small water contents of omphacite and garnet are indicative of extremely water-poor conditions, or very low water activity, during the metamorphism and in the system. Otherwise, the water contents of the two



minerals would dramatically increase, due to their strong ability in accommodating OH (Katayama and Nakashima, 2003; Katayama et al., 2006; Sheng et al., 2007), and their conductivity would be greatly enhanced, leading to conductive zones in the subducting slab that is inconsistent with geophysical mappings (Vanyan et al., 2002; McGary et al., 2014; Ichiki et al., 2015; Vargas et al., 2019). The water-poor conditions also imply very minor to negligible amounts of hydrous phases in the system, because of their extensive dehydration and water escape at shallow depths. This agrees with the arguments that subducting crusts may contain no hydrous minerals beyond ~3 GPa and 700 °C, as advocated by considering dehydration in natural rocks, numerical modeling and phase stabilities (Liu et al., 1996; Schmidt and Poli, 2014).

In this context, the amount of water carried to the deep mantle by subducting crust is regulated by omphacite and garnet in eclogites, but not hydrous phases. This implies a minor amount of water that could be brought to the deep mantle, although it might be significant at very shallow depths (Peacock, 1990; Cai et al., 2018). Assuming the total length, thickness and density of subducting oceanic crusts of 40000 km (Syracuse et al., 2010), 6 km and 3 g/cm<sup>3</sup>, respectively, and the average rate of subduction of 5 cm/year, the mass of water transferred to the mantle exceeding ~70 km depth in the past 3 billion years would correspond to <3% of the oceans. A direct implication is that the exchange of water between Earth's exterior and interior reservoirs might not be efficient. This fits well with the high dehydration efficiency (>92%) of subducting crusts at shallow depths, estimated from the H<sub>2</sub>O/Ce ratios of mid-ocean ridge basalts (Dixon et al., 2002), and the preservation of protosolar nebular water in the deep mantle over Earth's evolution, constrained by the D/H ratios of plume-related lavas (Hallis et al., 2015). The limited water transfer by crust deep subduction does not mean a water-poor deep Earth, because surface water (and atmosphere) formed by degassing of the Earth (Allègre et al., 1987)

and its deep interior was already hydrous.

Consequently, the water flux into the deep mantle is much smaller than that simply estimated from the stabilities of hydrous minerals such as amphibole, chlorite and dense hydrous magnesium silicates, by assuming their continuous presence in appreciated amounts in the downgoing crust. The stability fields of hydrous phases are established by experiments at water saturated/super-saturated conditions, e.g., with up to >10 wt.% water in sealed capsules (Liu et al., 1996; Frost, 2006; Schmidt and Poli, 2014). This requirement is hard to always fulfil in the subducting crust that is not a closed system and contains an initial H<sub>2</sub>O content of ~1-2 wt.% prior to subduction (Peacock, 1990), which is mostly released at shallow depths. Thus, there is a gap between the water-poor state in the deep slab and the water-rich state in laboratory. Finally, if slab mantle bears serpentine, e.g., by the serpentinization of peridotites providing there is enough water for serpentine formation prior to subduction, then its role in conveying water to Earth's interior should be evaluated. Key to this issue is to map the actual amount of serpentine in the slab mantle and the dynamics of the low density with subduction, e.g., 2.6 g/cm<sup>3</sup> of serpentine vs. 3.4 g/cm<sup>3</sup> of peridotite and 3.6 g/cm<sup>3</sup> of eclogite. This is beyond the scope of this work, and more studies are required.

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## Figure captions

**Fig. 1** Representative FTIR spectra of annealed omphacite and garnet. (a) polarized spectra of omphacite along three orthogonal directions of each grain ( $X'$ ,  $Y'$  and  $Z'$ ), and (b) unpolarized spectra of garnet from different grains. The omp2 and grt2 spectra were reproduced from Liu et al. (2019). Spectra were normalized to 1 cm thickness and vertically offset.

**Fig. 2** Representative complex spectra of (a) omphacite (B260) and (b) garnet (B256).  $Z'/Z''$  is the real/imaginary part of complex impedance, and frequency decreases along the  $Z'$  axis from left (1 MHz) to right (1 Hz). An equivalent circuit of a single  $R$ -CPE, a resistor ( $R$ ) and a constant phase element (CPE) in parallel, was used to fit the high-frequency arc. Tail and data scatter at low frequency were due to electrode effects that did not affect sample lattice resistance measurements. Number close to each spectrum is temperature ( $^{\circ}\text{C}$ ), and inset in (a) shows spectra at 600-700  $^{\circ}\text{C}$ .

**Fig. 3** Measured conductivity of (a) omphacite and (b) garnet at 1-2.5 GPa and 250-850  $^{\circ}\text{C}$ . The run at 2 GPa was marked, and lines are linear fittings to measured data of each sample (with  $\text{H}_2\text{O}$  contents labeled) by Eq. (1). Symbols around each line are from the same sample (different symbols for different cycles). Samples omp2 and grt2 were from Liu et al. (2019). Uncertainty is usually smaller than/comparable to symbol sizes. Data sources: assembly background, Yang et al. (2011); Z19, synthetic omphacites in Zhang et al. (2019) at 3 GPa (0% FeO and 280 ppm  $\text{H}_2\text{O}$  and 5.81% FeO and 420 ppm  $\text{H}_2\text{O}$ , with  $\text{H}_2\text{O}$  contents recalculated (see text)); Y12, augite in Yang and McCammon (2012) at 1 GPa (8.31% FeO and 75 ppm  $\text{H}_2\text{O}$ ); D09, pyrope in Dai and Karato (2009) at 8 GPa (7.42% FeO and 160 ppm  $\text{H}_2\text{O}$ , with  $\text{H}_2\text{O}$  content not recalculated owing to the different OH bands).

**Fig. 4** Modeled conductivity of (a) omphacite and (b) garnet at various FeO and  $\text{H}_2\text{O}$

contents. Colored solid lines were made using Eq. (2), and dashed and dotted grey lines were plotted using Eq. (3) for the bottom and top boundaries of FeO content (omp1 and omp3: see text), respectively. H<sub>2</sub>O contents of 100, 200 and 400 ppm in omphacite and 20, 40 and 80 ppm in garnet at equilibrium OH partitioning were assumed (see text).

**Fig. 5** Electrical conductivity of eclogites with different modal composition and Fe and H<sub>2</sub>O contents. (a) omp-rich (65 vol.% omp + 35 vol.% grt) eclogite, (b) grt-rich (35 vol.% omp + 65 vol.% grt) eclogite, and (c) a typical conductivity-temperature structure along subduction zones (modified after McGary et al. (2014): note that some reference resistivities were imposed in their starting inversion model: see text). Modeling in (a, b) was conducted with the mineral data in Fig. 4 and Eq. (4): blue, red and green colors highlight the different FeO contents, omp1 + grt1, omp2 + grt2 and omp3 + grt3 (with labels on the right side), and star symbols denote different OH contents in the minerals. Horizontal/vertical shadows in (a, b) show the conductivity/temperature range of the subducting crust at ~70-120 km depth in (c).

Table 1 Composition of the starting omphacite and garnet (by wt.%)

	SiO <sub>2</sub>	TiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	FeO	MnO	MgO	CaO	Na <sub>2</sub> O	K <sub>2</sub> O	Total
omp1	56.84	0.03	9.59	1.64	0.02	10.57	15.39	5.72	0.01	99.81
	<i>0.42</i>	<i>0.02</i>	<i>0.07</i>	<i>0.03</i>	<i>0.01</i>	<i>0.30</i>	<i>0.09</i>	<i>0.03</i>	<i>0.01</i>	
omp2*	55.64	0.07	11.38	4.27	0.02	8.44	12.98	7.31	<0.01	100.11
	<i>0.16</i>	<i>0.03</i>	<i>0.35</i>	<i>0.28</i>	<i>0.02</i>	<i>0.17</i>	<i>0.40</i>	<i>0.24</i>	<i>&lt;0.01</i>	
omp3	55.37	0.05	7.87	7.92	0.04	8.39	13.54	6.64	<0.01	99.82
	<i>0.26</i>	<i>0.01</i>	<i>0.08</i>	<i>0.15</i>	<i>0.04</i>	<i>0.03</i>	<i>0.08</i>	<i>0.16</i>	<i>&lt;0.01</i>	
grt1	39.48	0.05	22.22	15.40	0.29	9.33	12.24	<0.01	<0.01	99.01
	<i>0.15</i>	<i>0.03</i>	<i>0.07</i>	<i>0.17</i>	<i>0.01</i>	<i>0.18</i>	<i>0.19</i>	<i>&lt;0.01</i>	<i>&lt;0.01</i>	
grt2*	37.64	0.03	22.73	26.55	0.74	6.81	5.63	0.03	<0.01	100.16
	<i>0.20</i>	<i>0.02</i>	<i>0.12</i>	<i>0.36</i>	<i>0.08</i>	<i>0.17</i>	<i>0.32</i>	<i>0.02</i>	<i>&lt;0.01</i>	
grt3	37.49	0.07	21.63	26.58	0.14	5.02	8.79	0.04	<0.01	99.76
	<i>0.15</i>	<i>0.04</i>	<i>0.10</i>	<i>0.07</i>	<i>0.04</i>	<i>0.10</i>	<i>0.03</i>	<i>0.02</i>	<i>&lt;0.01</i>	

Assuming all Fe as FeO. Data are the average and standard deviation (italic) by electron

microprobe analyses. omp, omphacite; grt, garnet.

\*: data were reproduced from Liu et al. (2019) (see text).



570 Table 2 Summary of samples and fit parameters

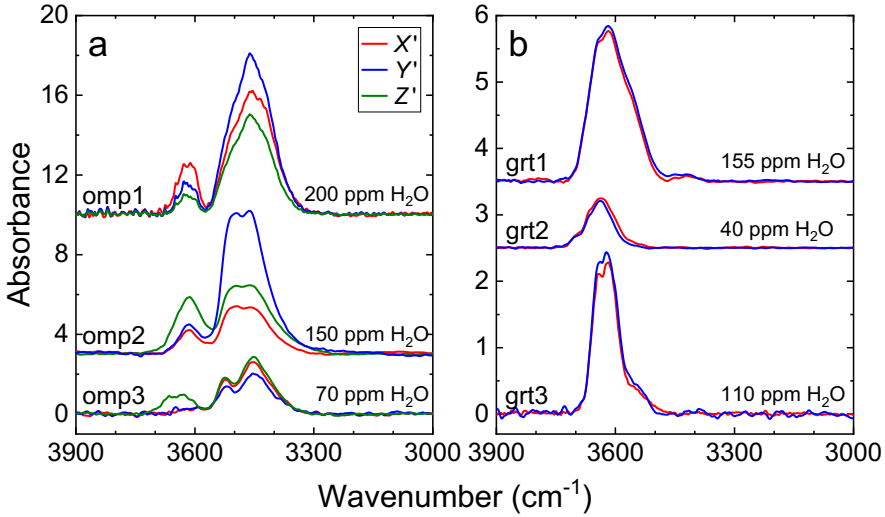
Sample	FeO (wt.%)	<i>P</i> (GPa)	<i>T</i> (°C)	ppm H <sub>2</sub> O (initial)	ppm H <sub>2</sub> O (final)	<i>L/S</i> (m <sup>-1</sup> )	log <sub>10</sub> ( <i>A</i> <sub>0</sub> (S/m))	Δ <i>H</i> (kJ/mol)
B255 (omp1)	1.64	1	350-700	200	190	288	1.97±0.01	87±2
B185 (omp2)*	4.27	1	350-600	85	80	290	1.38±0.24	84±4
B149 (omp2)*	4.27	1	350-600	150	160	216	1.52±0.14	82±2
B145 (omp2)*	4.27	1	350-650	290	300	269	1.75±0.12	81±2
B260 (omp3)	7.92	1	250-700	70	70	280	1.21±0.07	64±1
B261 (omp3)	7.92	2.5	250-700	70	65	285	1.45±0.04	65±1
B256 (grt1)	15.40	1	350-800	155	150	340	1.89±0.18	93±3
B167 (grt2)*	26.55	1	350-800	40	45	226	1.80±0.17	90±3
B174 (grt2)*	26.55	1	350-800	100	95	245	2.26±0.12	91±2
B267 (grt3)	26.58	1	350-850	110	110	246	2.15±0.13	88±2

571 Water contents are the values prior to (initial) and after (final) conductivity runs, and are rounded to the nearest 5 ppm. *L/S* is the ratio of  
572 sample length to cross-section area (by considering the effective contact between electrodes and sample). FeO contents of the samples are  
573 shown (from Table 1).

574 \*: samples were used in Liu et al. (2019) for documenting the water content exponent of 1 in Eq. (2).

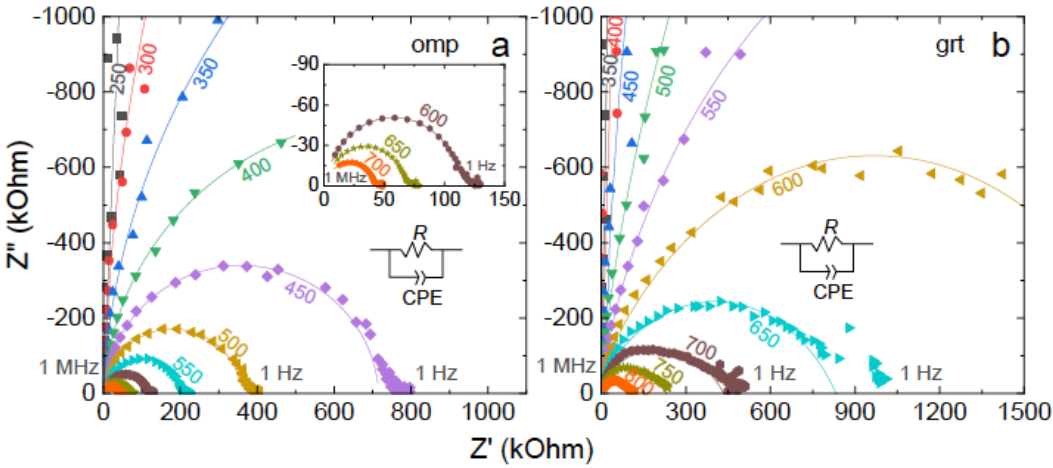
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Fig. 1



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Fig. 2



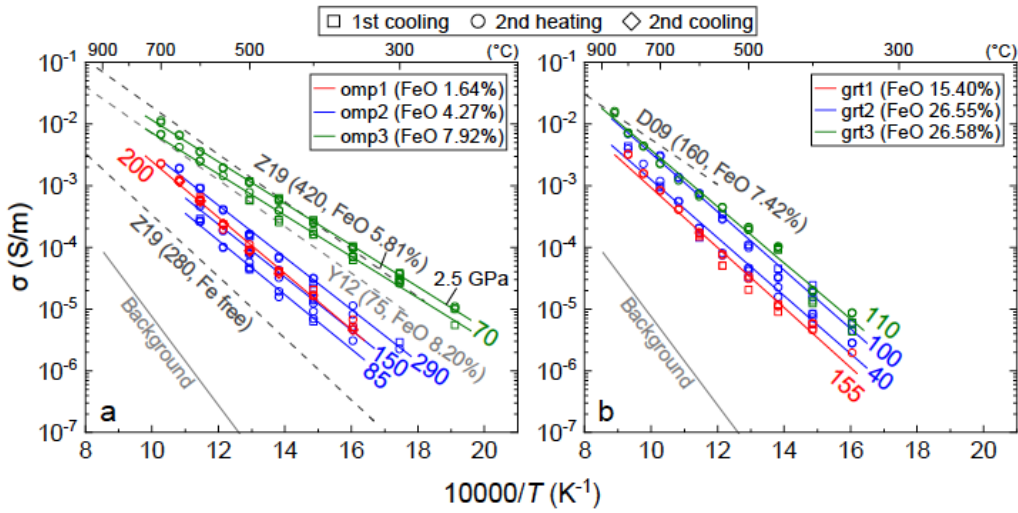
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Fig. 3



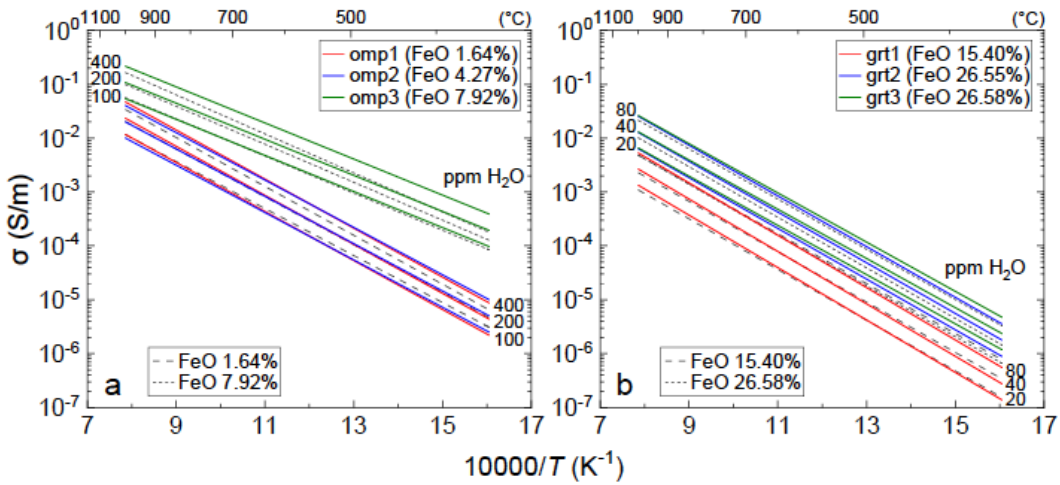
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Fig. 4



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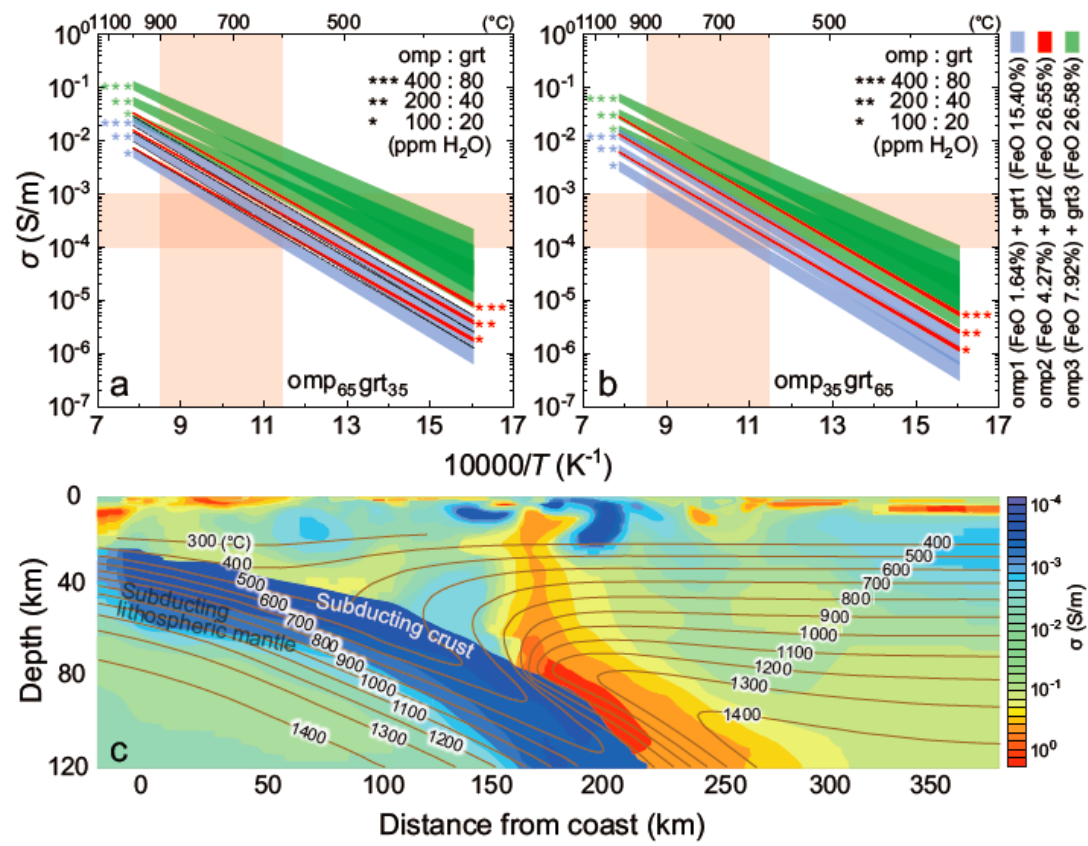
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Fig. 5



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