Effects of water, depth and temperature on partial melting of mantle-wedge fluxed by hydrous sediment-melt in subduction zones

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Abstract

This study investigates the partial melting of variable bulk H₂O-bearing parcels of mantle-wedge hybridized by partial melt derived from subducted metapelites, at pressure-temperature (*P-T*) conditions applicable to hotter core of the mantle beneath volcanic arcs. Experiments are performed on mixtures of 25% sediment-melt with 75% fertile peridotite, from 1200-1300 °C, at 2 and 3 GPa, with bulk H₂O concentrations of 4 and 6 wt.%. Combining the results from these experiments with previous

23 experiments containing 2 wt.% bulk H₂O (Mallik et al., 2015), it is observed that all melt compositions,
24 except those produced in the lowest bulk H₂O experiments at 3 GPa, are saturated with olivine and
25 orthopyroxene. Also, higher bulk H₂O concentration increases melt fraction at the same *P-T* condition,
26 and causes exhaustion of garnet, phlogopite and clinopyroxene at lower temperatures, for a given
27 pressure. The activity coefficient of silica (γ_{SiO_2}) for olivine-orthopyroxene saturated melt compositions
28 (where the activity of silica, a_{SiO_2} , is buffered by the reaction olivine + SiO₂ = orthopyroxene) from this
29 study and from mantle melting studies in the literature are calculated. In melt compositions generated at
30 2 GPa or shallower, with increasing H₂O concentration, γ_{SiO_2} increases from <1 to ~1, indicating a
31 transition from non-ideal mixing as OH⁻ in the melt (γ_{SiO_2} <1) to ideal mixing as molecular H₂O (γ_{SiO_2}
32 ~1). At pressures >2 GPa, γ_{SiO_2} >1 at higher H₂O concentrations in the melt, indicate requirement of
33 excess energy to incorporate molecular H₂O in the silicate melt structure, along with a preference for
34 bridging species and polyhedral edge decorations. With vapor saturation in the presence of melt, γ_{SiO_2}
35 decreases indicating approach towards ideal mixing of H₂O in silicate melt. For similar H₂O
36 concentrations in the melt, γ_{SiO_2} for olivine-orthopyroxene saturated melts at 3 GPa is higher than melts
37 at 2 GPa or shallower. This results in melts generated at 3 GPa being more silica-poor than melts at 2
38 GPa. Thus, variable bulk H₂O and pressure of melt generation results in the partial melts from this study
39 varying in composition from phonotephrite to basaltic andesite at 2 GPa and foidite/phonotephrite to
40 basalt at 3 GPa, thus forming a spectrum of arc lavas. Modeling suggests that the trace element patterns
41 of sediment-melt are unaffected by the process of hybridization within the hotter core of the mantle-
42 wedge. K₂O/H₂O and H₂O/Ce ratios of the sediment-melts are unaffected, within error, by the process of
43 hybridization of the mantle-wedge. This implies that thermometers based on K₂O/H₂O and H₂O/Ce
44 ratios of arc lavas may be used to estimate slab-top temperatures when a) sediment-melt from the slab
45 reaches the hotter core of the mantle-wedge by focused flow b) sediment-melt freezes in the overlying

mantle at the slab-mantle interface and the hybridized package rises as a mélange diapir and partially melts at the hotter core of the mantle-wedge. Based on the results from this study and previous studies, both channelized and porous flow of sediment-melt/fluid through the sub-arc mantle can explain geochemical signatures of arc lavas under specific geodynamic scenarios of fluid/melt fluxing, hybridization, and subsequent mantle melting.

1. Introduction

Arc lavas have typical geochemical signatures such as enrichment in Large Ion Lithophile Elements, H₂O, U and Th due to input from subducted slab in their source. The slab component involved maybe aqueous fluid and/or melt from subducted sediments and altered oceanic crust. The involvement of subducted sediments in the formation of arc lavas has been demonstrated by the presence of the cosmogenic isotope ¹⁰Be (e.g. Morris and Terra, 1989) in arc lavas (that suggests addition of recent surface material), and with similarity in trace element patterns and ratios between arc lavas and corresponding subducted sediments. Although geochemical evidence does indicate sediments being involved in the formation of arc lavas, the process by which their geochemical signature is transferred to arc magmas is not well understood. The sedimentary contribution in cold subduction zones is likely in the form of aqueous fluids, whereas, in intermediate to hot subduction zones, it is likely in the form of hydrous melts (Hermann and Rubatto, 2009). The melt compositions of metapelitic sediments are siliceous and may contain variable water contents in subduction zone conditions (e.g. Auzanneau, et al, 2006, Hermann and Spandler, 2008, Johnson and Plank, 2000, Thomsen and Schmidt, 2008, Tsuno and Dasgupta, 2012); hence, they metasomatize the mantle-wedge to form orthopyroxene at the expense of olivine ± hydrous minerals such as phlogopite and amphibole. Although the process of hybridization of sediment-melt with mantle-wedge peridotite has been studied, there remain the following outstanding questions on melting of the arc source, which require further investigation.

- a) How do pressure (P), temperature (T), and importantly bulk H_2O content of the zone of production of primitive arc magma as well as the flux of sediment-melt to peridotite affect the phase equilibria of hybridization? In the study by Mallik et al. (2015), the authors had studied the phase equilibria of hybridization of sediment-melt with mantle-wedge peridotite containing a fixed H_2O content of 2 wt.% in the bulk sediment-melt+peridotite mixture. They had found that such a process of hybridization produced ultrapotassic leucitites, similar to ultrapotassic lavas from several post collisional settings and active arcs. However, what effect does bulk H_2O content have on the magmas derived from sediment-melt fluxed mantle-wedge and can it, along with P and T , explain wider range of alkalic to tholeiitic arc lavas remain unknown.
- b) H_2O being the most abundant volatile species in arc magmas, what effect does it have on the melting systematics of the hybridized arc source? Hydrous partial melts of spinel peridotite (pressure ≤ 2 GPa) are andesitic due to expansion of olivine stability with respect to dry systems (Kushiro, 1972; Nicholls and Ringwood, 1973), whereas, hydrous partial melts of garnet peridotite are more silica-poor than their anhydrous partial melt counterparts . Why is there a difference of the influence of H_2O on mantle melting with pressure?
- c) The presence of slab-input in the source of primary arc lavas are inferred based on geochemical tracers. In fact, K_2O/H_2O and H_2O/Ce ratios of arc lavas have been used as thermometers to determine slab-top temperatures for various arcs (Cooper et al., 2012; Hermann and Spandler, 2008; Plank et al., 2009). If the primary arc lavas are generated by reactive infiltration of slab-derived component through the mantle-wedge, or, by hybridization of the slab-component with the mantle within a mélange, the geochemical tracers can be tied back to slab-derived components or used to estimate slab-top temperatures either, if they are unaffected by the process of hybridization, or, if the effect of the process of hybridization on the geochemical

tracers are known. Hence, it is important to investigate the effect of trace element systematics of slab components on the process of metasomatism/hybridization of the mantle-wedge.

- d) A recent study by Pirard and Hermann (2015) investigated the hybridization of sediment-melt with the mantle at the slab-mantle interface via porous and channelized flow. However, it needs to be investigated how the melt compositions produced by hybridization at the slab-mantle interface continue to affect the composition of equilibrated magma at the hotter mantle-wedge core.

In this study, we investigate the phase equilibria of a mixture of sediment-melt and sub-arc fertile mantle, at a P - T range and variable bulk H_2O concentrations, but with fixed sediment-melt:mantle ratio. We simulate the reactive infiltration of a mass of sediment-melt through the mantle-wedge, and explore the closed system partial melting behavior of the parcel of mantle hybridized by the sediment-melt, similar to that explored by Mallik and Dasgupta (2015) and Pirard and Hermann (2015). Experiments of Mallik and Dasgupta (2015) were done with a fixed water content. Here we explore the phase equilibria of similar reactive process but focus on the effect of variable H_2O on the partial melt chemistry and melting reactions. In particular, we explore how variable water content affects the silica activity and other major element composition of magmas generated from sediment-melt infiltrated peridotite. We also model the trace element concentration of the partial melts using the available mineral-melt partition coefficients and the mineral modes from our experiments. We demonstrate that the trace element signatures of slab-derived sediment-melt is not significantly affected by a) focused flow of sediment-melt through the mantle-wedge followed by metasomatism at the hotter core of the wedge, b) porous flow hybridization and complete freezing-in of sediment-melt at the slab-mantle interface, followed by diapiric rise of the hybridized package and subsequent partial

melting at the hotter core of the mantle-wedge. We also show that a range of partial melt compositions with similar major and trace element signatures to those of near-primary arc magmas can be produced by hydrous partial melting of sediment-melt fluxed fertile mantle-wedge.

2. Methods

2.1 Starting materials

The sediment-melt, in this study, is similar in major elements to the average composition of partial melts of Global Subducting Sediment or GLOSS (Plank and Langmuir, 1998) generated at 3.5 GPa, 900-950 °C in the study by Hermann and Spandler (2008) . Given the diversity of sediment compositions at trenches worldwide (Plank and Langmuir, 1998) and sediment partial melt compositions generated at sub-arc *P-T* conditions (e.g., Grassi and Schmidt, 2011; Hermann and Green, 2001; Hermann and Spandler, 2008; Mann and Schmidt, 2015; Schmidt, 2015; Schmidt et al., 2004; Thomsen and Schmidt, 2008; Tsuno and Dasgupta, 2011, 2012) , the sediment-melt composition chosen in this study is one of the several likely compositions to be applicable for subduction zones. The peridotite, in this study, is similar in composition to the lherzolite KLB-1 reported by Takahashi (1986) and Davis et al. (2009) . The starting materials are mixtures of 25% sediment-melt and 75% peridotite, and on a volatile-free basis, are similar to the starting material used by Mallik et al. (2015) . The bulk water contents of the sediment-melt + peridotite mixture used in this study are 4 wt.% and 6 wt.% (Table 1). In other words, the corresponding sediment-melt water contents are 16 and 24 wt.%. Experiments with 1.8 wt.% bulk water (7.2 wt.% H₂O in sediment-melt) have been reported in Mallik et al. (2015) . The sediment-melt and peridotite mixtures with 4 wt.% and 6 wt.% H₂O were synthesized following mixing, drying, and reduction procedures as outlined in Mallik et al. (2015).

138

139 2.2 *Experimental technique*

140 The starting materials (4 wt.% and 6 wt.% bulk water) were packed in Au₇₅-Pd₂₅ capsules with
141 the inner capsule of diameter 1.5 mm and the outer capsule of diameter 3 mm. The inner capsule and the
142 space between the two capsules were packed with the same starting material, in order to minimize
143 hydrogen loss from the starting material. This double capsule geometry has been modified from Kagi et
144 al. (2005). The inner and outer capsules were sealed by coning. Welding by a graphite arc welder was
145 avoided in order to minimize carbon contamination in the starting material (Mallik et al., 2015).

146 The experiments were performed using piston-cylinder apparatus in the Experimental Petrology
147 Laboratory at Rice University at pressures of 2 and 3 GPa, with temperatures ranging from 1200 to 1300
148 °C, which correspond to *P-T* conditions applicable for sub-arc mantle-wedge (England and Katz, 2010;
149 Syracuse et al., 2010) (Table 2). The experimental assembly, procedure, and relevant calibration are
150 reported in Tsuno and Dasgupta (2011) . Experiments were run for 38-93 hours, with lower H₂O-
151 bearing experiments towards the lower end of the temperature range run for longer durations. After the
152 assembly was extracted from the piston-cylinder, the capsules were cut in half longitudinally using a
153 wire-saw (with a wire of diameter 60 micrometer) and impregnated with epoxy under vacuum to
154 preserve the delicate hydrous quench from the melt pool as well as fragile phlogopite. The capsules were
155 polished under water, with the sample being exposed first under 1200 grit silicon carbide paper,
156 followed by polishing on lapping film impregnated with 3 micrometer alumina and velvet cloth with 0.3
157 micrometer alumina slurry. It was not possible to preserve the quench matter in the melt pool while
158 ensuring a good polish for the minerals simultaneously. Therefore one half of the capsule was polished
159 minimally to expose the delicate quench matter for compositional analysis, whereas, the minerals were
160 polished well in the second half.

161

162 2.3 *Analysis of run products*

163 The polished samples were imaged using the Scanning Electron Microscope FEI Quanta 400
164 FEG ESEM at Rice University and compositional analyses for all the phases in the inner and outer
165 capsules were performed using the Cameca SX 100 Electron Microprobe Analyzer (EMPA) at Johnson
166 Space Center, Houston and Inductively Coupled Plasma Mass Spectrometer (ICPMS) ThermoFinnigan
167 Element 2 coupled with a 213 nm laser ablation system (New-Wave) at Rice University. There is no
168 compositional difference observed between phases in the inner and outer capsules, within error.

169 Major and minor elements for all the phases were measured at 15 kV using EMPA. Olivine,
170 orthopyroxene and garnet were measured using 20 nA electron beam of diameter 1 micrometer,
171 phlogopite was measured at 5 nA using a 5 micrometer beam, whereas, the quenched melt pool was
172 measured using defocused 20-50 micrometer beam at 10 nA. The quenched pool, being coarse grained
173 and very heterogeneous in composition and texture, was analyzed using wide defocused beam to analyze
174 as large a spot as possible. Also, >60 points were analyzed for melt pools in each sample to obtain
175 robust average melt compositions. Counting times for all the elements were 20 seconds at the peak and
176 10 seconds on each background. The standards used were NIST glass K411 (Si, Ca, Fe, Mg), diopside
177 (Si, Ca, Mg), fayalite (Si, Fe), almandine (Si, Al, Fe), pyrope (Si, Al, Mg), andradite (Ca), oligoclase
178 (Al, Na), orthoclase (K), rutile (Ti), chromite (Cr), apatite (P) and rhodochrosite (Mn).

179 Concentrations of major and minor elements of quenched pool for all the samples, except G358
180 and G381, were also analyzed using LA-ICPMS to crosscheck between the compositions determined by
181 the two methods. EMPA is a non-destructive method and LA-ICPMS is destructive. The principal
182 advantage of using LA-ICPMS over EMPA is the higher depth of sampling of the former (~10 μm ,
183 Gonzalez et al., 2002) as compared to the latter (~1 μm , Goldstein et al., 2012). The instrument was

184 tuned using USGS glass standard BHVO2g on La (15 µg/g) with a beam size of 55 µm, repetition rate
185 of 10 Hz and fluence of 12-16 J/cm², in order to reach a sensitivity of 250,000 – 350,000 cps. The
186 USGS glass standards BHVO2g, BCR2g and BIR1g were analyzed as external standards before and at
187 the end of the sample analyses session. The external standards were reproduced within 1-9%. The data
188 acquisition time for each spot was 2 minutes 38 seconds, with ~30 seconds for background and the
189 remaining time for ablation. Individual spots were analyzed in the quenched pool, with diameter varying
190 from 80 – 110 µm, 10 Hz repetition rate and ~11 J/cm² fluence. The isotope ⁴⁴Ca was used as internal
191 standard for instrumental drift correction. The measurements were performed in medium resolution ($m/\Delta m = 3000$) and the oxide concentrations of the following isotopes were reported: ³⁰Si, ⁴⁹Ti, ²⁷Al, ⁵²Cr,
192 ⁵⁷Fe, ⁵⁵Mn, ²⁵Mg, ⁴⁴Ca, ²³Na, ³⁹K, ³¹P. Data reduction was performed using the Excel-Visual Basic
193 program from <http://www.cintylee.org/#/facilities/>.
194
195

196 3. Results

197 3.1 Texture and phase assemblage

198 Melt (represented by quench) in the experiments pooled at the top of the inner capsule and on
199 either sides of the outer capsule (Figure 1). The heterogeneous quench aggregate consists of coarse
200 (~150 µm long) and finer (~ ≤ 50 µm long) phlogopite-like radiating, acicular and dendritic grains with
201 darker interstitial matter as well as bright Fe-rich pyroxene-like grains. Olivine grains are coarser (~ ≥
202 50 µm) than orthopyroxene (~ ≤ 50 µm), with the olivine grains crystallized closest to the melt.

203 Phlogopite and garnet, when present in the sample, were located towards the bottom of the capsules.

204 Stability of phases as a function of pressure and bulk water content is shown in Figure 2 and
205 Table 2. Garnet, phlogopite and clinopyroxene are exhausted from the experiments at a lower
206 temperature with increasing bulk water concentrations at both 2 and 3 GPa. The distinct differences in

207 phase stability between 2 and 3 GPa are: (a) Absence of olivine in the lowest bulk water samples at 3
208 GPa whereas olivine was present at all bulk water concentrations at 2 GPa, (b) Absence of garnet at 2
209 GPa whereas garnet was present at 3 GPa, (c) Phlogopite and clinopyroxene are absent in the sample at
210 lower temperatures at 2 GPa as compared to 3 GPa, for a given bulk water concentration.

211 Phase proportions (including amount of melt) for each experiment were estimated by mass
212 balance using melt compositions from electron microprobe and LA-ICPMS, and mineral compositions
213 from electron microprobe (Table 2). The amount of melt produced in the experiments increases with
214 temperature at a given pressure and decreases with pressure at a given temperature (in agreement with
215 Mallik et al., 2015), the amount of melt also increases with increasing bulk H₂O concentration at a
216 certain temperature and pressure (Figure 3, Supplementary Figure 1). For example, at 2 GPa and 1200
217 °C, the melt fraction increases from 20 ± 3 to 47 ± 1 wt.% for bulk H₂O concentration of 2 to 6 wt.%,
218 whereas, at 3 GPa and 1250 °C, the melt fraction increases from 18 ± 3 to 43 ± 3 wt.% for a similar
219 increase in bulk H₂O concentration. A similar trend is also observed for melt fractions estimated using
220 melt compositions obtained from LA-ICPMS (Supplementary Figure 1).

221

222 3.2 Approach to chemical equilibrium, Fe-loss to capsule and determination of intrinsic oxygen 223 fugacity in the experiments

224 The experiments in this study have not been reversed but an approach to chemical equilibrium
225 can be demonstrated by: (a) homogeneity in composition of minerals from core to the rim (including
226 garnets), (b) systematic change in composition of melt and minerals as a function of temperature (Table
227 1, Supplementary Table 1), (c) olivine-melt and orthopyroxene-melt Fe²⁺-Mg K_D varies from 0.29-0.41
228 (for melt compositions determined by electron microprobe; Supplementary Table 1) and 0.19-0.39 (for

melt compositions determined by LA-ICPMS; Supplementary Table 1), which lie within the range of olivine/orthopyroxene-melt Fe^{2+} -Mg K_D of previous studies based on hydrous mantle melting .

The relative amount of Fe-loss to the $\text{Au}_{75}\text{Pd}_{25}$ capsule was estimated by mass balance and varied from 6 ± 4 to 33 ± 19 . Except for run G375, the relative Fe-loss to the capsules in this study is lower than the experiments performed using single $\text{Au}_{75}\text{Pd}_{25}$ or $\text{Au}_{80}\text{Pd}_{20}$ capsule in Mallik et al. (2015) . The intrinsic $f\text{O}_2$ of the experiments was measured using the oxybarometer of Barr and Grove (2010) using the pressure and temperature of the experiments, Fe concentration of the capsule closest to the sample and FeO^* concentration of melt. $f\text{O}_2$ varied from 0.0 ± 0.2 to -1.5 ± 0.2 log units with respect to the FMQ buffer. The $f\text{O}_2$ of the experiments agree with Mallik et al. (2015) and are similar to those estimated for supra-subduction zone peridotites (Foley, 2011).

3.3 Melt composition

The melt compositions measured by both EMPA and LA-ICPMS are reported in Table 1 and are compared in Supplementary Figure 2. Overall, the melt compositions measured by the two methods are in agreement; the most distinct difference being up to 70% (relative) higher K_2O concentration measured by LA-ICPMS over that measured by EMPA. This may be explained by the greater sampling volume of LA-ICPMS as compared to that of EMPA. Some of the K-rich phlogopite-like quench crystals were likely plucked off the top surface of the samples during polishing, resulting in lower K_2O concentrations measured by EMPA. We have presented the major results from this study using both data from electron microprobe and LA-ICPMS and we find that there is no significant difference observed in terms of discussion between the two.

The melt compositional data obtained from LA-ICPMS (for experiments B307, B309, G376, G377, G379, G380, G378, G374 and G375) and electron microprobe (for the remaining experiments)

are discussed here (Figure 4). In case of partial melts generated from bulk compositions with 4 wt.% bulk H₂O, the melt compositions are more siliceous at 2 GPa than at 3 GPa. At 2 GPa, with increasing melt fraction from 0.30 to 0.53, SiO₂ and MgO concentrations increase from 47 ± 1 to 51 ± 1 wt.% and 14.5 ± 0.7 to 19 ± 1 wt.%, respectively, whereas Al₂O₃, CaO, Na₂O and K₂O concentrations decrease. FeO* concentration varies from 5.2 ± 0.2 to 7.0 ± 0.6 wt.% with increasing melting degree. At 3 GPa, with increasing melt fraction from 0.24 to 0.35, SiO₂ and MgO concentrations increase from 41 ± 2 to 42.7 ± 0.6 wt.% and 21 ± 1 to 22.4 ± 0.4 wt.%, respectively. Al₂O₃, CaO, Na₂O and K₂O concentrations decrease, whereas FeO* varies from 7.9 ± 0.2 to 8.1 ± 0.2 wt.%.

In case of partial melts generated from bulk compositions with 6 wt.% bulk H₂O, the melt compositions are also more siliceous at 2 GPa than at 3 GPa. At 2 GPa, with increasing melt fraction from 47 to 58 wt.%, SiO₂ and FeO* concentrations vary around 50 ± 2 wt.% and 7.5 ± 0.4 to 7.2 ± 0.4 wt.%, respectively. Al₂O₃, CaO, Na₂O and K₂O display a decrease, whereas, MgO concentration increases with increasing degree of partial melting. At 3 GPa, with melt fraction increasing from 21 to 46 wt.%, SiO₂ and MgO concentrations increase from 44 ± 4 to 46.3 ± 0.8 wt.%, whereas, Al₂O₃, CaO, Na₂O and K₂O concentrations decrease. FeO* varies from 6 ± 1 to 8.0 ± 0.8 wt.%.

4. Discussion

4.1 Effect of variable water on melting systematics of sediment-melt+peridotite and peridotite

The partial melts from this study and Mallik et al. (2015) range in composition from phonotephrite to basaltic andesite at 2 GPa and foidite/phonotephrite to basalt at 3 GPa (Figure 5). The melts generated at 2 GPa are more siliceous as compared to those at 3 GPa, for the same bulk H₂O concentration. The reason why melt compositions produced at 3 GPa are more silica-poor than melt compositions produced at 2 GPa is discussed in the next section. Total alkali concentration of the melt

275 compositions depend on the degree of partial melting, bulk H₂O concentration and depth of melting. At
276 lower degrees of partial melting (hence, lower temperature), presence of phlogopite in the residue
277 buffers the K₂O content of the melt from 5-8 wt.%, subsequently decreasing as phlogopite is exhausted
278 from the residue. Phlogopite is exhausted at a lower temperature with higher bulk H₂O concentration;
279 hence, at the same *P-T* of melting, lower H₂O-bearing assemblages produce higher K₂O concentrations.
280 In addition, higher pressure stabilizes phlogopite to a higher temperature due to lower degree of melting
281 at higher pressure at a constant temperature. Na₂O is incompatible in the residual minerals; hence, Na₂O
282 concentration decreases with higher degree of melting. Therefore, a variety of primary arc lavas from
283 siliceous to more silica-undersaturated compositions, with a variety of alkali concentrations can be
284 produced by hybridization of sediment-derived melt with the mantle-wedge due to variable H₂O
285 concentration and pressure of hybridization, with higher pressure hydrous melts being more silica-
286 undersaturated than lower pressure melt compositions.

287 Certain differences in melt compositions are also observed as a function of increasing bulk H₂O
288 in the partial melting of sediment-melt fluxed peridotite (Figure 5). SiO₂ concentrations of partial melts
289 with 4 and 6 wt.% bulk H₂O are lower than those with 2 wt.% bulk water at 3 GPa, around similar
290 degrees of partial melting. At higher bulk H₂O contents, garnet, clinopyroxene and phlogopite are
291 consumed at lower degrees of partial melting. Thus, Al₂O₃, CaO, and K₂O display mostly incompatible
292 behavior, i.e., decreasing concentration with increasing degree of partial melting. This is unlike the
293 partial melt compositions with 2 wt.% bulk H₂O where compatible behavior is observed for these oxides
294 at lower degrees of partial melting due to the presence of garnet, clinopyroxene and phlogopite. It may
295 appear that partial melt compositions produced in experiments with higher bulk H₂O are more enriched
296 in FeO* (especially at 3 GPa), whereas, it is possible that the higher FeO* concentration is the result of
297 lower Fe-loss in the experiments in this study as compared to those from Mallik et al. (2015). MgO

298 content increases with increasing temperature and bulk H₂O concentration (Figure 6). This is related to
299 the degree of partial melting (F), i.e., at constant bulk H₂O content, F increases with temperature, and at
300 constant temperature, F increases with higher bulk H₂O contents. Higher F increases MgO content of
301 the melt (Figure 5), MgO being a compatible component.

302 Isobaric melt productivity (which determines how fluxing agents control the process of melting
303 in subduction zones) in peridotites is reduced by the presence of H₂O, because H₂O initiates melting at
304 lower temperatures and minerals have reduced participation in melting processes at lower temperatures.
305 Approximating isobaric melt productivity as a linear function at melt fractions ≥ 10 wt.%, Mallik et al.
306 (2015) found that at 2 and 3 GPa, 2 wt.% H₂O- bearing sediment-melt+peridotite lithology has a similar
307 melt productivity (0.14 – 0.15 wt.%/ °C at melt fraction of 7 to 35 wt.%) as peridotite+H₂O at 3.5 GPa
308 (0.10-0.13 wt.%/ °C at melt fraction of 18 to 31 wt.%). This indicates that the nature of fluxing agent
309 (hydrous sediment-melt versus aqueous fluid) has minor effect on the melt productivity of the
310 assemblage. Combining results from this study with the study by Mallik et al. (2015), it is observed that
311 4 wt.% H₂O-bearing sediment-melt+peridotite assemblage has an isobaric melt productivity of 0.2 wt.%/
312 °C at 3 GPa (melt fractions from 24 to 35 wt.%) and 6 wt.% H₂O-bearing sediment-melt+peridotite
313 lithology has an isobaric melt productivity of 0.18 wt.%/ °C at 3 GPa (melt fractions from 21 to 46
314 wt.%) (Figure 3). This corroborates the fact that at 3 GPa, irrespective of H₂O content, peridotite
315 hybridized by hydrous sediment-melt has a melt productivity that is similar to peridotite fluxed by H₂O
316 only. At 2 GPa, however, 4 and 6 wt.% sediment-melt+peridotite mixtures have isobaric melt
317 productivities of 0.24 wt.%/ °C (at melt fractions of 24 to 53 wt.%) and 0.32 wt.%/ °C (at melt fractions
318 of 45 to 61 wt.%), which are higher than the melt productivities of the same lithology at 3 GPa,
319 irrespective of bulk H₂O content. Thus, it appears that lower pressure of melting increases melt
320 productivity for the same amount of bulk H₂O present, especially at higher bulk H₂O contents.

321 Mallik et al. (2015) demonstrated that the difference in composition of partial melts of peridotite
322 fluxed by hydrous sediment-melt versus only H₂O is due to a difference in bulk composition of the two
323 lithologies, such as, higher SiO₂, Al₂O₃, Na₂O and K₂O and lower FeO*, MgO and CaO. A similar
324 observation is made when partial melt compositions of 4 and 6 wt.% H₂O-bearing sediment-
325 melt+peridotite mixtures are compared with partial melt compositions of hydrous peridotite at similar
326 pressure conditions as this study (Figure 4). This reinforces the argument that bulk compositional
327 control or the nature of fluxing agent has a strong effect on partial melt compositions of peridotite, even
328 though isobaric melt productivity remains similar.

329

330 4.2 *Effect of water and pressure on silica activity of mantle-derived partial melts*

331 Several studies have investigated the effect of H₂O on partial melting of mantle lithologies, and it
332 has been observed that hydrous partial melts of spinel peridotite (pressure of magma generation < 2.5
333 GPa) are more siliceous than hydrous partial melts of garnet peridotite (pressure of magma generation ≥
334 2.5 GPa) (e.g. Hirose, 1997; Hirose and Kawamoto, 1995; Irving and Green, 2008; Tenner et al., 2012).
335 Also, whereas a trend is not conspicuous for melts generated at 2 GPa and shallower, for deeper hydrous
336 mantle melts, the SiO₂ concentration of the melt decreases with increasing melt H₂O concentration, the
337 exception being that vapor-saturated mantle melts are very siliceous (Figure 7). In order to explain how
338 H₂O affects partial melting and the difference in the effect of H₂O at the two pressure ranges, we
339 estimated the activity coefficient of silica (γ_{SiO_2}) of mantle-derived partial melts (both anhydrous and
340 hydrous) saturated with olivine and orthopyroxene, as a function of melt H₂O content for spinel and
341 garnet-bearing mantle lithologies.

342 In a mantle-derived melt saturated with olivine and orthopyroxene at the liquidus, the activity of
343 silica (a_{SiO_2}) is buffered by the following reaction:

345 (Olivine) (Melt) (Orthopyroxene)

346 From equation 1, the following relation can be derived from equilibrium thermodynamics:

347 $\ln a_{\text{SiO}_2} = \frac{\Delta G^0}{RT} + \frac{1}{RT} \int P \Delta V dP + \ln a_{\text{EnOpx}} a_{\text{FoOl}}$ (2)

348 where, a_{SiO_2} = activity of SiO_2 in the melt

349 ΔG^0 = Gibbs free energy of the end-members in the reaction at T of reaction and P of 1 bar

350 ΔV = Molar volume change of the reaction at T of the reaction and P of 1 bar

351 a_{EnOpx} = Activity of enstatite in orthopyroxene at P - T of the reaction

352 a_{FoOl} = Activity of forsterite in olivine at P - T of the reaction

353 R = Molar gas constant

354 The methods of calculation of a_{SiO_2} and Y_{SiO_2} of the melt compositions using Equation 2 are
355 explained in Supplementary Information.

356 Y_{SiO_2} increases with increasing melt H_2O concentration, with Y_{SiO_2} increasing to 1 for melts
357 generated at 2 GPa and shallower, and Y_{SiO_2} increasing to >1 for melts generated deeper than 2 GPa.
358 H_2O mixing in silicate liquids is non-ideal and H_2O interacts with silica components during mixing,
359 which has also been demonstrated by Gaetani and Grove (1998) and Médard and Grove (2008). At
360 pressures of 2 GPa and shallower, melt compositions having low H_2O contents with $Y_{\text{SiO}_2} < 1$ likely have
361 H_2O mostly dissolved as OH^- in the silicate melt structure. As Y_{SiO_2} approaches 1, H_2O in the silicate
362 melt likely dissolves as molecular H_2O , placed in the ionic porosity of the silicate melt structure. At
363 higher pressures (> 2 GPa), Y_{SiO_2} exceeding 1 implies a positive deviation from ideal solution, implying

364 that excess energy is required to place the molecular H₂O in the ionic porosity of the silicate structure,
365 probably due to a combination of saturation of ionic porosity and due to reduction of ionic porosity due
366 to compressibility of the silicate melt at higher pressures. Also, a recent study investigating mechanism
367 of H₂O incorporation in silicate melt structures using first principles molecular dynamic simulations
368 (Bajgain et al., 2015) has demonstrated the preferred existence of bridging species and polyhedral edge
369 decorations (hydroxyls connected by hydrogen bonding) at high pressures at the expense of hydroxyls
370 (not connected by hydrogen bonding) as well as molecular water. Such high values of Y_{SiO_2} at values of
371 a_{SiO_2} buffered by equation 1 indicate approach towards immiscibility or aqueous fluid saturation.
372 Interestingly, with aqueous fluid saturation in the presence of melt, Y_{SiO_2} decreases indicating approach
373 towards ideal mixing of H₂O in the silicate melt. 2 GPa melts have lower Y_{SiO_2} than 3 GPa melts for
374 similar H₂O concentrations, the difference in Y_{SiO_2} increasing at higher melt H₂O concentrations. Melt
375 compositions with higher Y_{SiO_2} likely display higher structural connectivity due to the presence of
376 connected H₂O species. This enhances orthopyroxene to olivine formation, thus melts generated at >2
377 GPa are more silica deficient than melts generated below 2 GPa, as long as no aqueous fluid coexists
378 with the melt. Hence, it appears that at higher pressure, the effect of H₂O on partial melting is similar to
379 that of CO₂ i.e. higher orthopyroxene to olivine ratio resulting in lowering of silica concentration of the
380 melt compositions (Dasgupta et al., 2007; Dasgupta et al., 2013). Aqueous fluid co-existing with the
381 melt causes decrease in Y_{SiO_2} , resulting in suppressed orthopyroxene to olivine formation. This also
382 results in aqueous fluid-present melt compositions being more siliceous than aqueous fluid-
383 undersaturated melt compositions. This is a potential explanation for aqueous fluid-present near-solidus
384 partial melts of hydrous metasomatised lherzolites produced at 3.2 GPa being granitic (Grove and Till,
385 2014). Future studies employing *in-situ* techniques (in order to avoid quench modification) that would

386 complement first principles studies, are required to verify the differential speciation of dissolved H in
387 silicate melts as a function of pressure.

388

389 *4.3 Trace element signature of residual melts formed by metasomatism of the mantle-wedge by*
390 *sediment-derived melt*

391 Trace element signature of arc lavas have been used to infer the nature of slab input to the arc
392 source. However, it is important to understand if the slab-derived fluids undergo significant change in
393 trace element signatures during their reactive transport through the mantle-wedge (Pirard and Hermann,
394 2015).

395 The starting materials used in this study have not been doped with trace elements; hence, trace element
396 concentrations in the partial melts could not be measured. Instead, the trace element concentrations of
397 partial melts have been calculated by the method described in Supplementary Information.

398

399 *4.3.1 Trace element patterns of partial melt compositions derived by hybridization of hot mantle-*
400 *wedge by sediment-melt*

401 Trace element signatures of infiltrating sediment-melt and natural arc lavas, such as high
402 LILE/HREE, are preserved in the residual melt formed after metasomatism of the mantle-wedge
403 (Supplementary Figure 4). This implies that the trace element signature of the sediment-melt is carried
404 over to the primary arc lava, without modification by the process of metasomatism. The overall trace
405 element pattern of infiltrating sediment-melt is preserved after metasomatism and subsequent melting of
406 the metasomatised lithology irrespective of the mode of sediment-melt flow through the mantle-wedge,
407 i.e. for porous transport at the hotter core of the mantle-wedge (this study, Mallik et al., 2015) and
408 channelized transport from slab-top to hotter core of the mantle-wedge (Pirard and Hermann, 2015).

409 This is because the dominant mineralogy of the residue of metasomatism of peridotite by hydrous
410 sediment-melt consists of olivine and orthopyroxene, both minerals that do not fractionate trace
411 elements from one another. There is approximately 14 wt.% or less phlogopite present in the
412 experiments from Mallik et al. (2015) and this study; however, it appears that the modal proportions of
413 phlogopite (that could potentially deplete the coexisting melt in LILEs such as Cs, Rb and Ba) are not
414 high enough to alter the overall trace element pattern of the residual melts post metasomatism.

415 The fact that our experiments simulate natural subduction zone processes is further corroborated
416 by the modeled trace element signatures of the orthopyroxenes and the bulk residue of partial melting of
417 the hybridized mantle-wedge from our studies matching those of orthopyroxenes and
418 peridotites/pyroxenites from supra-subduction zones (representing crust-mantle interaction) in terms of
419 positive U and Pb anomalies and negative Nb anomaly (Supplementary Figure 5).

420

421 4.3.2 *Effect of hybridization of mantle-wedge on K_2O/H_2O and H_2O/Ce ratio of the sediment-melt*

422

423 K_2O/H_2O (Hermann and Spandler, 2008) and H_2O/Ce ratios have been used as thermometers to
424 estimate the slab-top temperatures in arcs. According to Hermann and Spandler (2008), K_2O/H_2O ratios
425 in sediment-derived fluids/melts depend on whether the slab-derived input to the arc source is a fluid or
426 melt. A fluid would result in very low K_2O/H_2O ratio ($\ll 0.002$). Melting of phengite (in sediments) in
427 fluid-present conditions will result in K_2O/H_2O varying from 0.1-1, and fluid-absent melting of phengite
428 will result in K_2O/H_2O ratios of ~ 2 , (given by the stoichiometry of K_2O and H_2O in phengite). Fluid-
429 absent sediment melting resulting in exhaustion of phengite will cause the K_2O/H_2O ratio of the slab-
430 derived component to be between 1 and 2. The H_2O/Ce ratio of sediment-melts, on the other hand, is
431 determined by the presence of allanite and monazite during sediment melting.

432 Pirard and Hermann (2015) investigated porous and channelized flow of sediment-melt through
433 the mantle-wedge at the slab-mantle interface. They reported that the K_2O/H_2O ratio of the residual
434 melt, during metasomatism of peridotite by porous flow of sediment-melt, is reduced by the formation
435 of phlogopite to the extent that the K_2O/H_2O ratio of the residual melts may exit the field of natural arc
436 lavas (Figure 8). Channelized flow of sediment-melt through the mantle-wedge at the slab-mantle
437 interface preserves the K_2O/H_2O and H_2O/Ce ratios of the residual melts because such a mode of
438 metasomatism leaves olivine and orthopyroxene in the residue, and neither of these minerals incorporate
439 H_2O , K and Ce in their structure; hence, does not alter such ratios in melts. On the basis of such an
440 argument, Pirard and Hermann (2015) concluded that arc lavas with sediment signatures are formed by
441 channelized flow of sediment-melt at the slab-mantle interface rather than porous flow. In this study, we
442 investigate the hybridization of the mantle-wedge at P - T conditions existing in its hotter core (Syracuse
443 et al., 2010)-by sediment-melt and subsequent partial melting of the hybridized lithology. We find:

444 a) The H_2O/Ce and K_2O/H_2O ratios of the infiltrating sediment-melt are very close to that of the
445 partial melts of the hybridized lithology (Figure 8, Table 3), i.e., the ratios in the infiltrating sediment-
446 melt are preserved in the subsequent partial melts. The only exception appears to be the partial melt
447 composition (estimated by electron microprobe) generated at 3 GPa with 4 wt.% bulk H_2O , where the
448 K_2O/H_2O ratio of this melt composition (0.07 ± 0.01) is much lower than the K_2O/H_2O of its infiltrating
449 sediment-melt with 16 wt.% H_2O (0.34). This difference may be attributed to the lower estimation of
450 K_2O concentrations in melt by electron microprobe due to loss of K-bearing quench phases from the
451 surface during polishing. When the melt composition was estimated by LA-ICPMS for the same
452 experiment, the difference between K_2O/H_2O ratios of the partial melt (0.21 ± 0.05) and the infiltrating
453 sediment-melt (0.34) are greatly reduced. The reason why K_2O/H_2O and H_2O/Ce ratios of infiltrating
454 sediment-melts and reacted partial melts do not differ significantly is because the primary residual

minerals formed in the experiments from this study and Mallik et al. (2015) are olivine and orthopyroxene, neither of which incorporates K, H₂O or Ce in their structure. Some experiments do contain minor to trace amount of phlogopite, which would affect the K₂O/H₂O ratio, if only slightly so. If one were to calculate slab-top temperatures using the K₂O/H₂O and H₂O/Ce ratios of infiltrating sediment-melts and reacted partial melts using the thermometers of Plank et al. (2009) and Cooper et al. (2012), it is observed that the temperatures determined using ratios of sediment-melts and reacted melts converge well, within error (Table 3). This demonstrates that slab-derived components could retain the memory of geochemical fingerprints and this may be utilized for slab-top temperature estimates, provided the following conditions are met: (1) only minor to trace amount of hydrous phases are formed during reactive crystallization of the slab-component through the mantle-wedge, (2) no accessory phases crystallize during the process of metasomatism. The geodynamic scenarios (i) and (ii) proposed to be applicable for the experiments in this study (see the following section) support the former condition. Accessory phases being present in mantle-wedge peridotitic samples is very rare (Tumiati et al., 2005), and no accessory phases crystallized in the hybridization experiments of Pirard and Hermann (2015), which were doped with trace elements. This indicates that the latter condition, i.e., no accessory phases crystallize during metasomatism, is likely fulfilled.

b) The K₂O/H₂O and H₂O/Ce ratios of the partial melts from this study and Mallik et al. (2015) lie mostly within the field of natural arc lavas (Cooper et al., 2012) (Figure 8), although towards the high K₂O/H₂O ratio extremity, which could simply be because the sediment-melts used in our study were relatively H₂O-poor compared to those generated by fluid-present melting of subducting sediments. Also, diapiric rise of sediments from the slab-top and subsequent melting of these sediments in the hotter core of the mantle-wedge also appear to produce sediment-melt compositions that would lie

outside the field of natural arc lavas in terms of K_2O/H_2O and H_2O/Ce ratios (Pirard and Hermann, 2015).

4.4 *Geodynamic scenarios applicable for this study with respect to hybridization of mantle-wedge by sediment-melt*

The following geodynamic scenarios are proposed in order to showcase how the experiments in this study and those in Mallik et al. (2015) are applicable to natural subduction zone processes (Figure 9):

- i) The residual melts due to focused flow of sediment-melt through the mantle-wedge at the slab-mantle interface from Pirard and Hermann (2015) are rhyolitic on a volatile-free basis and very close in composition to the infiltrating sediment-melt used in this study and Mallik et al., (2015). Therefore, the focused flow residual melts from Pirard and Hermann (2015), after their formation at the slab-mantle-wedge interface can continue to the core of the mantle-wedge where they can hybridize it. Subsequent partial melting of the hybridized lithology can take place, as simulated in this study and Mallik et al. (2015).
- ii) Sediment-melt generated at the slab-top can completely freeze in the mantle-wedge at the slab-mantle interface, i.e., if H_2O in the sediment-melt can be incorporated completely by hydrous minerals, which may be possible if slab melting occurs at fluid-absent or fluid-poor conditions in relatively hot subduction zones (Tsuno and Dasgupta, 2012; Tsuno et al., 2012). The hybridized lithology can rise to the core of the mantle-wedge as part of a *mélange* diapir, where it can undergo subsequent partial melting.
- iii) Porous flow of sediment-melt through the mantle-wedge at the slab-mantle interface creates a phlogopite-rich high K_2O/H_2O residue and a coexisting residual low K_2O/H_2O melt (step 1).

The residual low K_2O/H_2O melt rises through the mantle-wedge via channelized mode of transfer, and interacts with a preexisting phlogopite-rich, high K_2O/H_2O residue from a previous episode of metasomatism in the hotter core of the mantle-wedge (step 2). If the melt-rock ratio during step 1 is the same as that during step 2, the K_2O/H_2O ratio of the original sediment-melt will be reproduced in the final primary arc lava produced by the two-step hybridization. However, the melt-rock ratios being the same at the two steps would be fortuitous. Hence, this scenario, although applicable for the experiments performed in this study, is not favorable for preservation of K_2O/H_2O ratio of the original sediment-melt in the primary arc lava. In fact, if this fractionation and two-step process sets the K_2O/H_2O ratio of primary arc magmas, such ratio may not have any bearing on the slab surface temperatures. However, the H_2O/Ce ratio of the sediment-melt will be preserved through the two-step hybridization process and transferred to the primary arc lava. This is due to the fact that no phases involved in the hybridization processes fractionate H_2O from Ce . Subsequent partial melting of this hybridized lithology will create partial melt compositions akin to those produced in this study and Mallik et al. (2015).

All of the above geodynamic scenarios require that the infiltrating sediment-melt compositions with variable H_2O concentrations, used in this study and Mallik et al. (2015), are produced at slab-top conditions in subduction zones. According to Table 3, the slab-top temperature produced by the lowest H_2O -bearing sediment-melt (8 wt.% H_2O , 850 - 960 °C) may be produced at the slab-top at 3 GPa only in the hottest subduction zones such as Cascadia and Mexico (Syracuse et al., 2010). However, the geodynamic scenarios described above are valid even for hydrous silica-rich partial melts that are generated by lower degree partial melting

of wet sediments, formed at lower slab-top temperatures found in subduction zones colder than Cascadia and Mexico.

5. Conclusions

Partial melting of a hydrous sediment-fluxed peridotite at variable pressure, temperature and bulk water contents produces hydrous melt with olivine + orthopyroxene $\pm$ garnet $\pm$ phlogopite in the residue. A variety of primary arc lavas (from foidites to basaltic andesites) can be formed by hybridization of sediment-derived melt with the mantle-wedge, with the more silica-undersaturated, and MgO-rich melts being formed at higher concentrations of H₂O in the melt and higher pressure. Fluxing peridotite with hydrous sediment-melt versus H₂O does not affect the isobaric melt productivity at higher pressures, although the melt productivity increases with increasing bulk H₂O concentration at lower pressures. Also, the differential effect of H₂O with pressure on silica composition of the partial melts of mantle lithologies can be explained by changing Y_{SiO_2} of the melt with pressure, which, in turn, is a function of H speciation in the melt. This study also demonstrates that the process of hydrous metasomatism of mantle-wedge by slab-derived sediment-melts does not affect trace element characteristics of the slab/sediment-melt as long as minor to trace hydrous minerals and no accessory minerals remain in the residue during the course of hybridization; hence, primary arc lavas formed by such a process will retain the geochemical signature of the slab input. This is particularly the case if mantle-wedge hybridization occurs with relatively H₂O-rich sediment partial melt, which would destabilize phlogopite at much lower temperatures thereby preserving the K₂O/H₂O ratio of the original slab melt.

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683 **Figure captions:**

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685 Figure 1. (a) and (b) Backscattered electron image of experiment B309 (2 GPa, 1250 °C, 4 wt.% bulk
686 H₂O). (a) Image of the whole capsule showing quenched melt pool in the inner and outer capsules along
687 with a solid residue. (b) Magnification of the white box in a). Quenched melt pool in the outer capsule
688 co-existing with coarser grained olivine and finer grained orthopyroxene. The coarse nature of olivine
689 grains and their location next to the melt indicates that these are likely formed from dissolution and re-
690 precipitation of olivine grains in the original peridotitic composition. (c) Backscattered electron image of
691 experiment G376 (3 GPa, 1225 °C, 4 wt.% bulk H₂O) showing orthopyroxene, garnet and phlogopite at
692 the bottom of the outer capsule. Abbreviations used are: *Ol* - olivine; *Opx* - orthopyroxene; *Phl* –
693 phlogopite.

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695 Figure 2. Stability of phases in the pressure-temperature-bulk H₂O concentration space explored in this
696 study and Mallik and Dasgupta (2015). The results from experiments with 2 wt.% H₂O are from Mallik
697 and Dasgupta (2015). Dark field indicates presence of a phase. Abbreviations used are: *Ol* - olivine; *Opx*
698 - orthopyroxene; *Cpx* - clinopyroxene; *Phl* - phlogopite; *Gt* - garnet. Orthopyroxene and quenched melt
699 are present in all the experiments.

700 Figure 3. Amount of melt in weight fraction (estimated using melt compositions measured by LA-
701 ICPMS for experiments B307, B309, G376, G377, G379, G380, G378, G374 and G375) versus
702 temperature for this study compared with previous experimental studies of H₂O-bearing (vapor-
703 undersaturated) peridotite partial melting at 3.5 GPa (Tenner et al. 2012), H₂O vapor-saturated peridotite
704 partial melting at 3.2 GPa (Till et al. 2012) and anhydrous peridotite partial melting at 3 GPa (Walter
705 1998). Regression lines have been fitted through the melt fractions as a function of temperature. The
706 slopes of the lines are isobaric dF/dT. This figure is modified from Mallik et al., (2015). The equations
707 of the best-fit lines are as follows: $F = 0.13T - 160.09$ (1.5 wt% H₂O bearing peridotite), $F = 0.13T -$
708 140.2 (2.5 wt% H₂O-bearing peridotite), $F = 0.10T - 103.2$ (5 wt% H₂O-bearing peridotite), $F = 0.15T -$
709 146.77 (14.5 wt% H₂O-bearing peridotite), $F = 0.15T - 162.91$ (2 GPa, 2 wt.% H₂O), $F = 0.24T - 265$ (2
710 GPa, 4 wt.% H₂O, LA-ICPMS, this study), $F = 0.32T - 343$ (2 GPa, 6 wt.% H₂O, LA-ICPMS, this
711 study), $F = 0.14T - 161.33$ (3 GPa, 2 wt.% H₂O), $F = 0.22T - 245$ (3 GPa, 4 wt.% H₂O, LA-ICPMS, this
712 study), $F = 0.18T - 181$ (3 GPa, 6 wt.% H₂O, LA-ICPMS, this study).

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Figure 4. Partial melt compositions from this study and Mallik et al. (2015) reported on a volatile-free basis (experiments B307, B309, G376, G377, G379, G380, G378, G374 and G375 estimated by LA-ICPMS, the rest by electron microprobe) compared with partial melts of peridotite+H₂O, and sediment-melt+olivine. Vapor-absent partial melt compositions of peridotite+H₂O generated at 1.6-2.0 GPa and 3.5 GPa (pressures ranges closest to 2 and 3 GPa, i.e., pressures of melting explored in this study) are taken from Tenner et al. (2011), Mitchell and Grove (2015) and Pirard and Hermann (2015). Vapor-present partial melt compositions of peridotite+H₂O generated at 1.6 GPa and 3.2 GPa are taken from Till et al. (2012). The porous flow partial melt compositions of sediment-melt+olivine mixture are taken from Pirard and Hermann (2014, 2015).

Figure 5. SiO₂ versus total alkali (Na₂O+K₂O) concentration of partial melts from this study and Mallik et al. (2015). The classification of volcanic rocks used in the total alkali-silica figure is from Le Bas et al. (1986). The dashed line for dividing alkalic and tholeiitic lavas is from Irvine and Baragar (1971). All the partial melt compositions, except those from experiments with 2 wt.% H₂O formed at 3 GPa are olivine and orthopyroxene saturated, and have been reported on a volatile-free basis. The partial melt compositions have been determined by electron microprobe in 'a' and LA-ICPMS in 'b'. Natural arc lavas are compiled from GEOROC (Aegean, Aeolian, Cascades, Central American Volcanic Province, Izu-Bonin, Mariana, Sunda and Tonga) and Mallik et al. (2015) for ultrapotassics. They are filtered for compositions with MgO > 6 wt.%, and olivine is added until the composition is in equilibrium with Fo₈₉, in order to compare the experimental melt compositions with primary arc lavas.

Figure 6. MgO content on a water-free basis versus temperature of experiments for partial melt compositions from this study (melt compositions from experiments B307, B309, G376, G377, G379, G380, G378, G374 and G375 determined by LA-ICPMS and the rest by electron microprobe) and Mallik et al., (2015). Line 1 indicates MgO versus temperature relationship from Ulmer (2001) for anhydrous mantle melt compositions. Lines 2, 3 and 4 are hand drawn to guide the readers to note the MgO versus temperature relationships for 2, 4 and 6 wt.% bulk H₂O systems, respectively from this study. Our data clearly show that partial melts with higher MgO contents are derived from peridotite at a fixed temperature when fluxed with higher water content. Olivine-orthopyroxene saturated hydrous partial melt compositions from literature (Gaetani and Grove, 1998; Grove et al., 2006; Hirose, 1997; Hirose and Kawamoto, 1995; Kinzler and Grove, 1992; Kogiso et al., 1998; Mitchell and Grove, 2015; Parman and Grove, 2004; Pirard and Hermann, 2014; Pirard and Hermann, 2015; Tenner et al., 2012; Till et al., 2012) are also plotted for comparison.

Figure 7. SiO₂ versus H₂O concentration of melt from this study and Mallik et al. (2015) compared with peridotite+H₂O melt compositions. (a) Melt compositions from this study are determined by electron microprobe. (b) Melt compositions from experiments B307, B309, G376, G377, G379, G380, G378, G374 (this study) are determined by LA-ICPMS. Silica activity coefficient (γ_{SiO_2}) of olivine-orthopyroxene saturated melt as a function of H₂O concentration in melt, where melt compositions from this study and Mallik et al. (2015) were determined by electron microprobe. (c) γ_{SiO_2} calculated by the first method (see Supplementary Information for further details). (d) γ_{SiO_2} calculated using by the

second method (see Supplementary Information for further details), i.e., using the thermodynamic database and equation of state of Holland and Powell, (2011). Peridotite±H₂O partial melt compositions generated at 2 GPa and shallower, without coexisting with vapor (Gaetani and Grove, 1998; Hirose, 1997; Hirose and Kawamoto, 1995; Kinzler and Grove, 1992; Kogiso et al., 1998; Mitchell and Grove, 2015; Parman and Grove, 2004), and coexisting with vapor/fluid (Grove et al., 2006; Till et al., 2012) are plotted along with peridotite±H₂O partial melt compositions generated deeper than 2 GPa, without coexisting vapor/fluid (Falloo and Green, 1988; Falloo et al., 1988; Hirose and Kushiro, 1993; Parman and Grove, 2004; Takahashi, 1986; Tenner et al., 2012; Walter, 1998), and coexisting with vapor/fluid (Till et al., 2012). The hydrous sediment-melt+olivine partial melt compositions are taken from Pirard and Hermann (2014, 2015). Y_{SiO_2} of melt compositions generated deeper than 2 GPa, without coexisting with vapor display an increasing trend with melt H₂O contents represented by the black curve. Melt composition generated deeper than 2 GPa, coexisting with vapor/fluid has a lower Y_{SiO_2} as compared to melt compositions not coexisting with vapor or fluid. The melt compositions from Pirard and Hermann (2014, 2015) with H₂O concentrations greater than 20 wt.% are most likely vapor/fluid-present, which would explain their low Y_{SiO_2} as compared to H₂O-rich melt compositions generated deeper than 2 GPa. Y_{SiO_2} calculated by both methods display nearly identical trends of Y_{SiO_2} as a function of H₂O content of melt.

Figure 8. K₂O/H₂O and H₂O/Ce ratios of natural arc lavas (Cooper et al., 2012), obtained from least-degassed melt inclusions, and sediment-melts formed in diapirs (Hermann and Spandler, 2008; Stepanov et al., 2012) compared with infiltrating sediment-melts and partial melts of peridotite hybridized with sediment-melt (used in this study and Mallik et al., 2015) as well as low-K and high-K infiltrating sediment-melts and their corresponding reacted melts from porous flow and focused flow of sediment-melts through dunite (Pirard and Hermann, 2015). (a) Partial melt compositions from this study and Mallik et al. (2015) determined using electron microprobe (b) Partial melt compositions from this study (experiments B307, B309, G376, G377, G379, G380, G378, G374 and G375) determined using LA-ICPMS. This figure is modified from Figure 3 of Pirard and Hermann (2015).

Figure 9. Geodynamic scenarios applicable for experiments performed in this study and Mallik et al. (2015). The yellow blobs represent final residues of melting that have minor to trace phlogopite or are devoid of phlogopite. a) Sediment-melt from the subducted slab undergoes channelized flow through the mantle to hybridize the mantle-wedge at its hotter core. b) Sediment-melt from the subducted slab completely freezes into the mantle at the slab-mantle interface. The hybridized lithology rises to the hotter core of the mantle-wedge via mélange diapirs. c) Step 1 – sediment-melt from the slab hybridizes the mantle at the slab-mantle interface via porous flow and produces a low K₂O/H₂O melt and phlogopite-rich high K₂O/H₂O residue. Step 2 - the low K₂O/H₂O melt undergoes channelized flow through the mantle-wedge and reacts with a pre-existing high K₂O/H₂O, potentially phlogopite-bearing residue. The hybridized lithology formed in this second stage undergoes partial melting. For details, refer to the text. If scenario (c) is applicable to nature then the resulting K₂O/H₂O ratio of the mantle partial melt may not provide any insight on the slab surface temperatures.

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Table 1

Table 1. Compositions of starting materials and melt compositions in the experiments

Exp. No.	Bulk H ₂ O (wt.%)	P (GPa)	T (°C)	n	Name of starting material / Phase	SiO ₂	TiO ₂	Al ₂ O ₃	Cr ₂ O ₃	FeO*	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	Sum	H ₂ O
					SPMH*	72.69	0.43	14.93	0	0.94	0.03	0.26	0.95	3.19	6.38	0.18	100	
					KLB-1ox [†]	44.82	0.15	3.51	0.32	8.19	0.12	39.5	3.07	0.3	0.02	0	100	
					SPMHKLB4	51.81	0.22	6.37	0.24	6.38	0.1	29.7	2.54	1.02	1.61	0.05	100.05	4
					SPMHKLB6	50.44	0.21	5.81	0.26	6.73	0.12	31.56	2.64	0.89	1.31	0.04	100	6
B307	4	2	1200	134	Melt ¹	50.9(8)	0.63(9)	13.5(5)	0.2(1)	5.1(6)	0.13(3)	14.5(7)	8.3(4)	2.6(3)	4.1(4)	0.03(6)	100.0	13.2(4)
				5	Melt ²	47(1)	0.79(4)	16.2(7)	0.091(3)	5.2(2)	0.156(5)	12.5(6)	10.0(4)	3.2(1)	4.8(2)	0.164(7)	100.0	16.6 (1)
B309	4	2	1250	108	Melt ¹	50.3(9)	0.57(7)	12.4(5)	0.3(2)	7.4(9)	0.15(4)	15(2)	8(2)	2.7(5)	3.8(1)	0.13(4)	100.1	10.4(3)
				6	Melt ²	48.9(8)	0.61(2)	13.22(1)	0.108(3)	6.7(2)	0.127(8)	15(1)	7.6(6)	3.0(4)	4.3(6)	0.15(1)	100.0	10.9(3)
G358	4	2	1300	69	Melt ¹	51(1)	0.44(6)	11.0(8)	0.28(9)	7.0(6)	0.13(3)	19(1)	5.9(6)	1.9(4)	2.7(4)	0.09(3)	100.0	7.6(4)
G376	4	3	1225	82	Melt ¹	46(3)	0.6(1)	13(1)	0.14(6)	7.0(8)	0.14(6)	18(2)	11(2)	3(1)	1.1(7)	0.2(1)	100.0	16(4)
				8	Melt ²	41(2)	0.69(3)	13.9(6)	0.149(8)	7.9(2)	0.15(1)	21(1)	8.8(8)	2.6(4)	3.4(4)	0.12(1)	100.0	17(1)
G377	4	3	1275	61	Melt ¹	46(2)	0.5(1)	14(2)	0.13(5)	7.1(7)	0.18(7)	20(2)	9(2)	2.2(7)	1.0(5)	0.1(1)	100.0	12(2)
				5	Melt ²	42.7(6)	0.60(2)	13.19(8)	0.163(3)	8.1(2)	0.15(2)	22.4(4)	7(1)	2.20(9)	3.2(4)	0.116(8)	100.0	11.5 (1)
G379	6	2	1200	102	Melt ¹	52(2)	0.43(5)	13(2)	0.18(4)	6.2(6)	0.12(4)	16(2)	8(1)	1.7(5)	1.5(4)	0.07(4)	100.0	13(3)
				6	Melt ²	50(2)	0.50(3)	12.0(3)	0.191(8)	7.5(4)	0.144(5)	17(1)	7.5(4)	1.81(8)	2.7(6)	0.074(7)	100.0	13.4(6)
G380	6	2	1250	106	Melt ¹	52(2)	0.36(5)	12(2)	0.24(5)	7.0(7)	0.13(4)	20(2)	5.1(7)	1.4(4)	2.2(7)	0.08(4)	100.0	10(2)
				8	Melt ²	50(2)	0.40(3)	10(1)	0.27(2)	7.2(4)	0.154(7)	24(1)	5.3(6)	1.3(1)	2.1(1)	0.078(3)	100.0	9.8(5)
G381	6	3	1150	32	Melt ¹	44(4)	0.7(2)	17(5)	0.10(6)	6(1)	0.14(6)	14(3)	10(2)	5(3)	2.2(9)	0.14(7)	100.0	26(2)
G378	6	3	1200	141	Melt ¹	47(3)	0.5(1)	16(4)	0.15(6)	6.4(7)	0.14(5)	17(3)	9(2)	2.3(7)	2.0(9)	0.12(8)	100.1	15.8(3)
				6	Melt ²	45(2)	0.66(3)	12(1)	0.17(1)	7.8(5)	0.17(1)	20(1)	8.5(7)	1.7(1)	3.6(4)	0.078(6)	100.0	15.8
G374	6	3	1250	134	Melt ¹	49(2)	0.5(1)	12(2)	0.17(7)	7.5(9)	0.16(6)	18(3)	10(2)	1.8(5)	1.1(7)	0.08(8)	100.0	15(1)
				7	Melt ²	44(2)	0.59(3)	12.9(2)	0.19(1)	10.2(3)	0.201(1)	19.2(9)	7.5(8)	1.7(5)	3.2(4)	0.061(6)	100.0	15.7(2)
G375	6	3	1300	67	Melt ¹	49(2)	0.5(1)	13(4)	0.17(7)	4.8(6)	0.16(5)	21(3)	8(3)	1.4(6)	3(1)	0.03(5)	100.0	13.1(5)
				6	Melt ²	46.3(8)	0.58(2)	11.6(3)	0.195(7)	8.0(8)	0.16(2)	21(2)	7.9(6)	1.74(1)	3.0(6)	0.07(1)	100.0	13.8(4)

All major element compositions are reported on a volatile-free basis. Al(OH)₃ and Mg(OH)₂ in case of SPMHKLB4 and SPMHKLB6, mixed in the starting compositions.

* = Volatile-free composition as reported in Mallik et al (2015); † = Composition as reported in Mallik and Dasgupta (2013,2014).

All oxide and H₂O concentrations are reported in weight percent. H₂O concentration is estimated by mass balance.

¹Melt compositions estimated by electron microprobe; ²Melt compositions estimated by LA-ICPMS

Table 2

Table 2. Conditions, phase proportions and intrinsic oxygen fugacity estimates for experiments used in this study.

Exp. No.	P (GPa)	T (°C)	Duration (h)		Melt	Ol	Opx	Cpx	Gt	Phl	Σr ²	Fe-loss	Δ fO ₂
4 wt.% bulk water													
B307	2	1200	93	EMPA	0.304(9)	0.058(7)	0.58(1)			0.056(8)	0.5(6)	22.0(4)	-1.5(2)
				LA-ICPMS	0.241(1)	0.005(5)	0.70(1)			0.055(5)	0.5(5)	22.8(6)	
B309	2	1250	91	EMPA	0.38(2)	0.113(1)	0.50(2)				1.2(6)	6.2(3)	-0.2(3)
				LA-ICPMS	0.37(1)	0.082(2)	0.55(1)				0.7(2)	12(2)	
G358	2	1300	93	EMPA	0.53(3)	0.150(6)	0.32(3)				0.6(7)	15	-0.4(3)
G376	3	1225	90	EMPA	0.25(7)	0.02(4)	0.60(5)		+	0.13(6)	0.3(1)	15(7)	-0.9(3)
				LA-ICPMS	0.24(2)	+	0.67(1)		+	0.065(5)	0.4(1)	8.3(1)	
G377	3	1275	52	EMPA	0.35(5)	+	0.65(2)				2.8(9)	13(1)	-0.4(3)
				LA-ICPMS	0.348(2)	+	0.652(2)				3.0(2)		
6 wt.% bulk water													
G379	2	1200	70	EMPA	0.47(8)	0.20(6)	0.33(9)				2.7(5)	9(8)	0.0(2)
				LA-ICPMS	0.45(3)	0.15(2)	0.40(6)				1.4(6)	3(5)	
G380	2	1250	66	EMPA	0.58(7)	0.16(6)	0.26(9)				1.1(3)	5(3)	-0.4(3)
				LA-ICPMS	0.61(4)	0.06(3)	0.33(7)				1(2)	4(7)	
G381	3	1150	77	EMPA	0.21(1)	0.04(1)	0.64(2)			0.11(2)	0(1)	6(1)	-0.5(2)
G378	3	1200	66	EMPA	0.38(2)	0.10 (1)	0.52(2)				1.3(8)	9.0(1)	-0.6(2)
				LA-ICPMS	0.38	0.02	0.60				1.4(6)	4(3)	
G374	3	1250	91	EMPA	0.43(3)	0.10(4)	0.48(5)				4(4)	7(5)	-1.9(3)
				LA-ICPMS	0.375(9)	0.011(5)	0.61(1)				1.0(3)	-4(4)	
G375	3	1300	45	EMPA	0.460(8)	+	0.54(1)				4.6(9)	40(16)	-0.7(3)
				LA-ICPMS	0.46(2)	+	0.54(2)				2(1)	20(7)	

Abbreviations used: Ol - Olivine; Opx - Orthopyroxene; Cpx - Clinopyroxene; Gt - Garnet; Phl - Phlogopite.. Phase proportions are reported in weight fraction. $\pm 1\sigma$ errors are mentioned in brackets and reported as least digits cited, e.g. 0.304(9) to be read as 0.304 $\pm$ 0.009 wt. fraction Σr^2 - Sum of residual square. Fe-loss indicates the relative Fe-loss to the Au-Pd capsules w.r.t the bulk composition, as estimated by mass balance. ΔfO_2 - Difference between intrinsic oxygen fugacity of experiments estimated by the oxy-barometer of Barr and Grove (2010) and FMQ (fayalite–magnetite–quartz) buffer of O'Neill (1987) and Ballhaus et al. (1991). '+' - Indicates trace presence of a phase which could not be detected by mass balance

Table 3. K₂O/H₂O and H₂O/Ce ratios, as well as slab-top temperatures estimated using K₂O/H₂O and H₂O/Ce ratios (Plank et al., 2009, Cooper et al., 2012)

H ₂ O in sediment melt (wt.%) / Bulk H ₂ O (wt.%) [#]	P (GPa)	K ₂ O/H ₂ O	T _{K2O/H2O}	H ₂ O/Ce	T _{H2O/Ce}
Starting sediment melt compositions					
8		0.73	960	1818	850
16		0.34	880	3636	790
24		0.20	830	5454	750
Experiments with sediment melt and peridotite mixtures*					
2	2	0.8(2)	960(32)	1,700(117)	861(7)
	3	0.6(3)	930(68)	1,800(456)	860(22)
4	2	0.31(4)	870(12)	3,809(4)	785.9(1)
	3	0.07(1)	720(22)	3,760(77)	787(2)
6	2	0.2(1)	840(46)	3,804(1)	786.00(3)
	3	0.16(8)	800(46)	3,600(461)	790(12)
Experiments with sediment melt and peridotite mixtures ^s					
4	2	0.31(6)	870(21)	4,200(542)	780(12)
	3	0.21(5)	830(25)	3,860(79)	785(2)
6	2	0.29(2)	865(5)	3,805(2)	785.98(4)
	3	0.24(8)	840(45)	3,700(560)	790(14)

- Bulk H₂O refers to H₂O concentration in the 25% sediment-melt and 75% peridotite mixture used in experiments

Thus, 8 wt.% H₂O in the sediment-melt indicates 2 wt.% bulk H₂O; 16 wt.% H₂O in the sediment-melt indicates 4 wt.% bulk H₂O; 24 wt.% H₂O in the sediment-melt indicates 6 wt.% bulk H₂O

* - Melt compositions determined using electron microprobe

\$ - Melt compositions determined using LA-ICPMS

Figure 1

Figure 2

Figure 3

Figure 7

Figure 9

a)

b)

c)

Hybridization of low $\text{K}_2\text{O}/\text{H}_2\text{O}$ melt and high $\text{K}_2\text{O}/\text{H}_2\text{O}$ residue. Partial melting of hybridized lithology