

1 Revision 1

2 Experimental constraints on the partial melting of sediment-metasomatized

3 lithospheric mantle in subduction zones

4

5 Yanfei Zhang^{1,3,*}, Xuran Liang^{2,3}, Chao Wang³, Zhenmin Jin³, Lüyun Zhu⁴, Wei Gan⁵

6

7 ¹ College of Oceanography, Hohai University, Nanjing 210098, China

8 ² Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou

9 510640, China

10 ³ State Key Laboratory of Geological Processes and Mineral Resources, China

11 University of Geosciences, Wuhan 430074, China

12 ⁴ School of Earth Resources, China University of Geosciences, Wuhan 430074, China

13 ⁵ School of Earth Sciences and Resources, China University of Geosciences, Beijing

14 100083, China

15 * Corresponding author: Y. Zhang

16 Email: yanfzhang@126.com

17 No. 1, Xikang road, Nanjing 210098, Jiangsu, China

18 No. 388, Lumo road, Wuhan 430074, Hubei, China

19

20 **Abstract**

21 Sedimentary diapirs can be relaminated to the base of the lithosphere during slab
22 subduction, where they can interact with the ambient lithospheric mantle to form
23 variably metasomatized zones. Here, high-pressure experiments in
24 sediment-harzburgite systems were conducted at 1.5–2.5 GPa and 800–1300 °C to
25 investigate the interaction between relaminated sediment diapirs and lithospheric
26 mantle. Two end-member processes of mixed experiments and layered (reaction)
27 experiments were explored. In the first end-member, sediment and harzburgite
28 powders were mixed to a homogeneous proportion (1:3) whereas in the second, the
29 two powders were juxtaposed as separate layers. In the first series of experiments, the
30 run products were mainly composed of olivine + orthopyroxene + clinopyroxene +
31 phlogopite in subsolidus experiments, while the phase assemblages were then
32 replaced by olivine + orthopyroxene + melt (or trace phlogopite) in supersolidus
33 experiments. Basaltic and foiditic melts were observed in all supersolidus mixed
34 experiments (~44–52 wt% SiO₂ at 1.5 GPa, ~35–43 wt% SiO₂ at 2.5 GPa). In the
35 phlogopite-rich experiment (PC431, 1.5 GPa and 1100 °C), the formed melts had low
36 alkali contents (~<2 wt%) and K₂O/Na₂O ratios (~0.4–1.1). In contrast, the quenched
37 melt in phlogopite-free/poor experiments showed relatively higher alkali contents
38 (~4–8 wt%) and K₂O/Na₂O ratios (~2–5). Therefore, the stability of phlogopite could
39 control the bulk K₂O and K₂O/Na₂O ratios of magmas derived from
40 sediment-metasomatized lithospheric mantle. In layered experiments, a reaction zone

41 dominated by clinopyroxene + amphibole (or orthopyroxene) was formed because of
42 the reaction between harzburgite and bottom sediment-derived melts (~62.5–67 wt%
43 SiO₂). The total alkali contents and K₂O/Na₂O ratios of the formed melts were about
44 6–8 wt% and 1.5–3, respectively. Experimentally formed melts from both mixed and
45 reaction experiments were rich in large ion lithophile elements and displayed similar
46 patterns with natural potassium-rich arc lavas from oceanic subduction zones (i.e.,
47 Mexian, Sunda, Central American and Aleutian). The experimental results
48 demonstrated that bulk sediment diapirs, in addition to sediment melt, may be another
49 possible mechanism to transfer material from a subducting slab to an upper mantle
50 wedge or lithospheric mantle. On the other hand, the breakdown of phlogopite may
51 play an important role in the mantle source that produces potassium-rich arc lavas in
52 subduction zones.

53

54 Keywords: bulk sediment, lithosphere, partial melting, potassium-rich arc lava

55

56

57 **1. Introduction**

58 Subducted sediment is thought to have played an important role in creating
59 mantle heterogeneity and the origin of potassium (K)-rich magmas in subduction
60 zones (e.g., [Tatsumi et al. 1986](#); [Blundy and Sparks 1992](#); [Prouteau et al. 2001](#)).
61 High-pressure experimental studies have demonstrated that subducted sediment may
62 experience partial melting or dehydration because of heating from the surrounding
63 mantle (e.g., [Hermann and Spandler 2008](#); [Hermann and Rubatto 2009](#); [Castro et al.](#)
64 [2010](#); [Mann and Schmidt 2015](#); [Schmidt and Poli 2014](#); [Schmidt 2015](#)), which could
65 release trace elements to the upper mantle wedge and contribute to the origin of arc
66 lavas (e.g., [Peacock 1990](#); [Ulmer and Trommsdorff 1995](#); [Shimoda et al. 1998](#);
67 [Bindeman et al. 2005](#); [Duggen et al. 2007](#); [Turner et al. 2012](#); [Spandler and Pirard](#)
68 [2013](#); [Kendrick et al. 2014](#); [Harvey et al. 2014](#); [Scambelluri et al. 2015](#); [Shu et al.](#)
69 [2017](#)). The recycling of sediment melt or fluid back into a mantle wedge has been
70 demonstrated by matching the isotopic and trace element properties of sediment input
71 and volcanic magma output (e.g., [Plank and Langmuir 1993](#); [Peate et al. 1997](#); [Porter](#)
72 [and White 2009](#); [Turner et al. 2012](#)).

73 Recent numerical simulations and geochemical analyses have revealed that
74 subducted sediments could detach from a downgoing slab at temperatures of 500–850
75 °C to form buoyancy diapirs ([Behn et al. 2011](#)). Then, a portion of the cold diapir
76 would rise buoyantly from the surface of the subducting slab and relaminate into the
77 mantle beneath arc lavas ([Hacker et al. 2011](#); [Marschall and Schumacher 2012](#);

78 [Maunder et al. 2016](#)), where they would form a variety of mélanges through physical
79 mixing before arc magma generation ([Marschall and Schumacher 2012](#); [Nielsen and](#)
80 [Marschall 2017](#)). On the other hand, geochemical analyses of alkaline arc lavas (very
81 low Nd/Sr at low $^{143}\text{Nd}/^{144}\text{Nd}$ values, $^{87}\text{Sr}/^{86}\text{Sr}$ ratios close to the depleted mantle)
82 have suggested that the composition of these arc lavas plot on the mixing line between
83 bulk sediment and mantle peridotite, rather than sediment melt and mantle peridotite
84 in Sr-Nd isotope space ([Cruz-Uribe et al. 2018](#)). Recently, [Shu et al. \(2017\)](#)
85 investigated the variation of arc lava Tl, Sr, Nd and Pb isotopes from the Ryukyu Arc
86 and Okinawa Trough; their results demonstrated that a mixing model between bulk
87 sediment (without trace element fractionation) and lithospheric mantle should be the
88 principle source of these arc lavas. Therefore, it was suggested that melting of
89 bulk-mixed mélange between sediment and lithospheric mantle/peridotite should be
90 an important transfer mechanism for the recycling of sediment components from a
91 subducting slab to arc lavas ([Marschall and Schumacher 2012](#)). On one hand,
92 sediment diapirs may react with mantle peridotite (lherzolite) at sub-arc depths (~80–
93 120 km); on the other hand, some sediment diapirs may be relaminated into the bottom
94 of lithospheric mantle because of buoyancy and react with lithospheric mantle
95 (harzburgite). There are two key issues concerning the partial melting phase relations
96 of sediment-harzburgite mélange and partial melt chemical compositions. Many
97 high-pressure experiments have been conducted to constrain the partial melting phase
98 relations in sediment + mid-oceanic ridge basalt (MORB) ([Castro et al. 2010](#)),

99 eclogite-derived melt + lherzolite ([Mallik and Dasgupta 2013, 2014](#); [Gervasoni et al.](#)
100 [2017](#)), hydrous rhyolite + lherzolite ([Mallik et al. 2015](#)), sediment melt + lherzolite
101 ([Mallik et al. 2016](#)), carbonate melt + lherzolite ([Gervasoni et al. 2017](#)), glimmerite +
102 harzburgite ([Förster et al. 2017](#)), phyllite + dunite ([Wang et al. 2017](#); [Wang and Foley](#)
103 [2018](#)), adakite + lherzolite ([Corgne et al. 2018](#)) and sediment-dominated mélange +
104 lherzolite ([Codillo et al., 2018](#)). Those experimental results were mainly used to
105 constrain melt-rock reaction processes at subarc depths and the origin of arc lavas in
106 oceanic subduction zones. However, the metasomatism of lithospheric mantle by bulk
107 sediment diapirs or the partial melting phase relations of sediment +
108 harzburgite/dunite mélange is not completely understood because few experiments
109 have been done ([Förster et al. 2019a, b](#)).

110 In this study, we conducted a series of experiments to examine the partial melting
111 phase relations of sediment-harzburgite mélanges (mixed system vs. layered (reaction)
112 system] at 1.5–2.5 GPa and 800–1300 °C, which corresponds to depths of ~45–75 km
113 below the surface. The explored models assumed that a portion of sediment diapirs
114 have been relaminated to the bottom of the lithospheric mantle during slab subduction
115 and formed a variety of mélanges. The main purpose of this study was to investigate
116 the effect of bulk sediment diapirs on the partial melting phase relations of mantle,
117 and its implications for the origin of potassium-rich arc lavas.

118

119 **2. Experimental details**

120 **2.1 Starting materials**

121 The natural sediment sample (MSCS-3) used in this study was collected from the
122 East China Sea continental shelf and is composed mainly of quartz, illite, chlorite and
123 trace amounts of calcite ([Wang et al. 2009](#)). The volatile components in the sediment
124 sample were mainly of H₂O (~4.9 wt%) and a small amount of CO₂ (~2.1 wt%). Both
125 the major and trace element compositions of MSCS-3 were similar to global
126 subducting sediment (GLOSS) ([Plank and Langmuir 1998](#)), except that it contained
127 relatively higher concentrations of Al₂O₃ and lower concentrations of SiO₂ and CaO.
128 The MSCS-3 powder was finely ground (~< 15 μm) in alcohol with an agate mortar.
129 The harzburgite sample (Hz13) was synthesized by mixing approximately 82 wt%
130 olivine with 18 wt% enstatite. The olivine and enstatite crystals were first
131 hand-picked from a natural peridotite sample (DMP-018), then separately crushed
132 into fine powder (~5–15 μm) and ground together in alcohol with an agate mortar to
133 produce a homogeneous mixture. The mixed powder of the starting material (HS1A)
134 was ground by mixing 25 wt% MSCS-3 with 75 wt% harzburgite in alcohol with an
135 agate mortar to form a homogeneous fine powder. For reaction experiments, MSCS-3
136 was juxtaposed at the bottom of Hz13; the proportions of the two powders were
137 roughly 1:1. Details of the starting material chemical compositions in this study and
138 previous experiments are listed in [Table 1](#).

139

140 **2.2 Experimental techniques**

141 High-pressure and high-temperature experiments were conducted at the
142 laboratory for the Study of the Earth's Deep Interior (SEDI-Lab) at China University
143 of Geosciences (Wuhan, China). Experiments at 1.5 GPa were carried out using a 150
144 ton non-end-loaded piston cylinder apparatus; experiments at 2.5 GPa were conducted
145 in a 1000 ton multi-anvil press using 18/12 ((octahedral edge length/truncated edge
146 length) cell assembly. For experiments conducted in the piston cylinder apparatus, the
147 assembly consisted of a sample capsule placed between two MgO rods in a graphite
148 heater, enclosed by a Pyrex and salt sleeve; the sample capsule was separated by a
149 short MgO tube from the graphite heater (Wang et al. 2010). In terms of the 18/12
150 assembly, a ZrO₂ sleeve was placed inside of the spinel octahedron for thermal
151 insulation and a graphite furnace was placed inside the ZrO₂ sleeve for heating; the
152 sample capsule was placed inside a short MgO tube and sandwiched between two
153 MgO rods. Type C thermocouples were used to measure the temperature at the center
154 of the furnace without correcting for the effect of pressure on electromotive force.
155 Au₈₀-Pd₂₀ tubes were used as sample capsules. The capsules were first laser-sealed at
156 one end, then the sample powders were loaded into the capsules, which were welded
157 shut to avoid volatile release during the experiments. Experiments were terminated by
158 switching off power to the furnace, quenching to below 200 °C within 10 s and then
159 automatic decompression to atmospheric pressure.

160

161 **2.3 Analytical techniques**

162 The recovered samples were first polished for microstructure analysis using a
163 scanning electron microscope (FE-SEM). Quantitative major element analyses of
164 minerals and melts in the recovered samples were made on a JXA-8100 electron
165 probe micro-analyzer (EPMA) using an accelerating voltage of 15 kV, a beam current
166 of 20 nA and a beam size of 1–2 μm for minerals, and a beam current of 10 nA and a
167 beam size of 5–30 μm for quench melts. The count times for measured elements were
168 10 s on peaks and 5 s on the background on each side of the peaks. The standard
169 materials were diopside (Si, Mg and Ca), jadeite (Na), K-feldspar (K), pyrope (Al),
170 $\text{Ca}_5\text{P}_3\text{F}$ (P), hematite (Fe), MnSiO_3 (Mn), rutile (Ti), NiO (Ni) and eskolaite (Cr). The
171 measurement errors were generally about 5% for elements of no more than 1 wt%, <1%
172 for element concentrations of 1 wt% to 5 wt%, and <0.5% for elements contents of >5
173 wt%. Phase proportions in mixed experiments were estimated based on anhydrous
174 mass balance calculations; minor oxides, such as Cr_2O_3 , NiO and P_2O_5 , were excluded.
175 A spreadsheet package provided by [Herrmann and Berry \(2002\)](#) was used to calculate
176 the mineral and melt weight proportions from major element compositions of bulk
177 rock and each phase in the recovered samples. The amount of Fe-loss to the sample
178 capsule was also estimated based on mass balance calculations. The calculated weight
179 proportions of phases, uncertainties and residual errors are shown in [Table 2](#).

180 Trace element contents of melt pools were measured by laser ablation inductively
181 coupled plasma-mass spectrometry (LA-ICP-MS). The laser ablation system is
182 GeoLas 2005, which is combining with an Agilent 7500a ICP-MS instrument at the

183 State Key Laboratory of Geological Processes and Mineral Resources (China
184 University of Geosciences). All samples were measured in a time-resolved analysis
185 mode and ablated by 193 nm laser under conditions of 5 J/cm² (energy density), 6 Hz
186 (repetition rate) and 32 μm (spot size). More detailed information about operating
187 conditions of the LA-ICP-MS are described in [Zhu et al. \(2013\)](#). Each analysis
188 includes a background acquisition section of approximately 20–30 s (gas blank) and a
189 data acquisition section of 50 s (sample). In this study, the combination data strategy
190 of external standard calibration and internal standard is adopted for data calibration, in
191 order to obtain accurate and precise element contents ([Longerich et al. 1996](#);
192 [Mukherjee et al. 2014](#)). In details, the international reference material NIST610 and
193 U.S. Geological Survey reference glasses BIR-1G, BCR-2G, and BHVO-2G were
194 used for data calibration of LA-ICP-MS analysis. The time-drift of the signal is
195 corrected by linear interpolation (with time) according to the variations in NIST 610.
196 Moreover, BCR-2G, BHVO-2G and BIR-1G were adopted as the external standards
197 for the quantitative calibration. In general, off-line selection and integration of
198 background and analytical signals along with time-drift correction and quantitative
199 calibration were conducted using ICPMSDataCal ([Liu et al. 2008](#)).

200

201 **3. Results**

202 Back-scattered electron images of representative run products are shown in
203 [Figure 1](#). The stability range of mixed experiment phases in *P-T* space is shown in

204 [Figure 2](#). Details of the experimental conditions and run products are summarized in
205 [Table 2](#) and the mineral and quench melt chemical compositions in all experiments
206 are listed in [Tables S1–S3](#) (supplementary data). In the reaction experiments, the
207 analyzed melt pools were greater than $\sim 200\text{--}300\ \mu\text{m}$ in diameter; the quench melt
208 chemical compositions were homogeneous because no quench crystals were found in
209 the recovered charges. Therefore, representative melt compositions of both major and
210 trace elements can be analyzed in large melt pools far away from the solid minerals.
211 In most of the mixed experiments ([Figure 1](#)), re-crystallization textures were observed
212 in quench melt pools. For EPMA analyses, large beam sizes ($5\text{--}30\ \mu\text{m}$) were adopted
213 to mitigate compositional errors associated with the quench textures, and
214 representative values for each composition were obtained by averaging analytical
215 points ($n=15\text{--}21$) in different regions of the sample capsules. In these experiments, the
216 largest melt pools were usually no more than $\sim 200\ \mu\text{m}$ in diameter, which limited the
217 LA-ICP-MS analyses as the laser spot had to be at least $32\ \mu\text{m}$ in diameter. Reliable
218 trace element compositions were obtained for melt pools greater than $\sim 50\text{--}100\ \mu\text{m}$ in
219 diameter. Melt pools that were slightly smaller than $\sim 50\ \mu\text{m}$ ($\sim 30\text{--}50\ \mu\text{m}$) were also
220 selected for LA-ICP-MS analyses, but no reliable data were obtained because the
221 trace element compositions were easily contaminated by the surrounding solid phases.
222 Each time-resolved spectrum was carefully checked for possible contamination by
223 trace element-rich minerals. The analytical results in different domains showed

224 similar trace elements patterns, indicating that re-crystallization of quench melts had a
225 small effect on trace element migration during experiment quenching.

226

227 **3.1 Chemical equilibrium and the loss of iron in mixed experiments**

228 In this study, reversal experiments were not conducted. In subsolidus
229 experiments, variable Mg# [$100 \cdot \text{Mg}/(\text{Mg}+\text{Fe})$] of olivine and orthopyroxene may
230 indicate residual olivine crystals from the starting harzburgite that have not yet
231 achieved Fe-Mg exchange equilibrium with the rest of the minerals in the sample.
232 This is because a relatively lower temperature is adopted in those experiments, and it
233 is very difficult to achieve equilibrium without a liquid diffusive medium. In
234 supersolidus experiments, an approach to chemical equilibrium can be demonstrated
235 by: (1) limited heterogeneity in mineral composition from core to rim and in different
236 zones of the recovered sample capsule; (2) systematic changes in phlogopite
237 composition as a function of temperature ([Figure 3c–f](#)); (3) systematic changes in
238 olivine and orthopyroxene composition as a function of temperature (≥ 1100 °C)
239 ([Figure 3a, b](#)); (4) variations in olivine-melt and orthopyroxene-melt Fe^{2+} -Mg K_D
240 from 0.16 to 0.33, which is roughly consistent with previous partial melting
241 experiments in a sediment melt + lherzolite system ($K_D^{\text{Fe-Mg}}_{(\text{Ol}/\text{Opx-melt})} = 0.2\text{--}0.4$) (e.g.,
242 [Mallik et al. 2016](#)); (5) the sum of residual squares ranges from 0.1 to 2.0, which is
243 low when considering the uncertainties in melt composition from average

244 heterogeneous quench phases. These criteria may imply that the experiments have
245 approached chemical equilibrium.

246 The relative amount of Fe-loss to the sample capsule was calculated based on
247 mass balance and varied from 17(1) to 32(1) % (Table 2). The relative Fe-loss in this
248 study is consistent with previous studies in sediment melt + lherzolite systems
249 (~10%–40%) that used Au₈₀Pd₂₀ or Au₇₅Pd₂₅ capsules (e.g., Mallik et al. 2015, 2016).
250 Iron loss resulted in FeO deficiency in both the quench melt and solid phases. By
251 assuming that the partition coefficient of FeO between the melt and other solid phases
252 is not affected by Fe-loss, the deficiency of FeO in the quench melt can be calculated
253 using the following equation: $FeO_{Def} = FeO_{Blost} / [D_{FeO}*(1-F)+F]$

254 where FeO_{Def} represents the deficiency of FeO from the melt composition, which
255 can be added to measured melt FeO to obtain the FeO concentration of the melt prior
256 to Fe-loss; FeO_{Blost} is the lost FeO from the bulk composition; D_{FeO} represents the
257 bulk partition coefficient of FeO between the solid phases and melt, which can be
258 calculated through the weight percent of each mineral and the mineral/melt partition
259 coefficient of FeO; F is the weight proportion of melt.

260

261 **3.2 Mixed experiments**

262 In mixed experiments, the starting material was first transformed into an
263 assemblage consisting of olivine + orthopyroxene + clinopyroxene + phlogopite and
264 trace accessory minerals (i.e., magnesite, hematite, rutile, apatite and zircon) within a

265 temperature range of 800–1000 °C at 1.5 GPa (Table 2, Figure 2). When the
266 temperature was increased to 1100 °C, phlogopite and traces of melt were observed in
267 the run products (Figure 1a, c), indicating that the stability field of phlogopite is
268 greater than ~1100 °C at pressures of 1.5–2.5 GPa (Figure 2). For experiments
269 conducted at 1200–1300 °C, a fraction of quench melt was observed within
270 interparticles or along the sample capsules, but clinopyroxene and phlogopite were
271 not present in the run products (Figure 1b, d; Figure 2). The observation of magnesite
272 in the run products is consistent with the presence of trace amounts of carbonate in the
273 starting material; therefore, the quench melts may also contain traces of CO₂ because
274 no magnesite (or other carbonate phase) was detected in all supersolidus experiments.

275 Microprobe analyses provided compositional data for the coexisting phases in
276 the recovered experiments. Figure 3 shows the compositional variations of olivine,
277 orthopyroxene and phlogopite versus temperature at 1.5 GPa and 2.5 GPa. The Mg#
278 for olivine and orthopyroxene were corrected for Fe-loss by adding Fe to the resulting
279 experimental phases. The average Mg# of olivine and orthopyroxene in subsolidus
280 experiments was in the range of 88–90 and 87–89, respectively, while the Mg# of
281 olivine and orthopyroxene increased slightly with temperature in supersolidus
282 experiments (Figure 3a, b). As an important alkali-bearing phase, the concentrations
283 of K₂O and TiO₂ in phlogopite ranged from ~6–9 wt% and 0.4–1.6 wt%, respectively,
284 with temperatures from 800 °C to 1200 °C; while the Na₂O and NiO contents were
285 reduced from ~2 wt% to 1 wt% and ~0.4 wt% to 0.2 wt%, respectively, with

286 temperatures from 800 °C to 1100 °C (Figure 3c–f). Both alkali contents and
287 K₂O/Na₂O ratios for quench melts were lower at 1100 °C compared with 1200–1300
288 °C (Figure 3g, h), because a significant portion of K was contained by phlogopite
289 (PC431, ~8 wt%) that coexisted with melts in the run products.

290

291 **3.3 Reaction experiments**

292 In the reaction experiments, the sediment powder was juxtaposed at the bottom
293 of the harzburgite layer (Figure 1e). Three experiments were carried out at 1.5–2.5
294 GPa and 950–1000 °C to simulate the reaction between sediment-derived melt and
295 lithospheric mantle. In the run products, the bottom sediment powder was composed
296 mainly of melt + garnet and a small fraction of plagioclase or clinopyroxene. The
297 upper harzburgite layer mainly consisted of olivine + orthopyroxene, and trace
298 amounts of spinel. For experiments conducted at 950 °C and 1.5 GPa, a reaction zone
299 composed mainly of clinopyroxene and amphibole was observed between the
300 sediment-derived melt and harzburgite (Figure 1f), which was replaced by
301 orthopyroxene at 1000 °C and pressures of 1.5–2.5 GPa.

302

303 **3.4 Melt composition**

304 Melt compositions obtained using EPMA are shown in Figures 4–5. The FeO
305 concentrations in experimental melts were also corrected for Fe-loss by adding FeO to
306 melts, based on the partitioning of Fe between melt and other phases. Figure 4 shows

307 the SiO_2 -($\text{K}_2\text{O}+\text{Na}_2\text{O}$) diagram of melt compositions from both mixed and reaction
308 experiments. In mixed experiments, melts formed at 1.5 GPa were mainly located
309 within basalt and tephrite-trachybasalt regions in phlogopite-rich and
310 phlogopite-free/poor experiments, respectively. In phlogopite-free/poor experiments,
311 the experimental melts had total alkali contents and $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratios of ~4–8 wt%
312 and ~2–5, respectively. In phlogopite-rich experiment (i.e., PC431), the quench melt
313 displayed SiO_2 concentrations of ~44–52 wt% with total alkali contents and
314 $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratios of ~0.7–3.7 wt% and ~0.4–1.1, respectively, because a significant
315 portion of the bulk K_2O was contained by phlogopite in the residue phases. With a
316 pressure increase to 2.5 GPa, the experimental melts were located in the
317 foidite-tephrite zone, which displayed similar alkali concentrations and $\text{K}_2\text{O}/\text{Na}_2\text{O}$
318 ratios to experimental melts from phlogopite-free/poor experiments at 1.5 GPa, but
319 had relatively lower SiO_2 concentrations (~35–43 wt%). In reaction experiments, the
320 resultant melts were mainly located within the dacite-trachydacite region, which
321 showed SiO_2 concentrations of ~62.5–67 wt% and total alkalis and $\text{K}_2\text{O}/\text{Na}_2\text{O}$ ratios
322 of ~6–8 wt% and ~1.5–3, respectively. The quench melt compositions in
323 phlogopite-rich (1.5 GPa) and reaction experiments belong to the subalkaline series,
324 while experimental melts formed at 2.5 GPa are in the alkaline series.

325 Harker diagrams (Figure 5) were used to show the variations in major element
326 compositions in this study. The Al_2O_3 concentrations in the quench melts showed
327 weak correlations with increased SiO_2 (Figure 5a). In melts derived from mixed

328 experiments, the Al₂O₃ contents varied from ~16 wt% to 20 wt% at 1.5 GPa and
329 1100–1300 °C, which then decreased to ~12–17 wt% when pressure was increased to
330 2.5 GPa. In melts formed in reaction experiments, the average Al₂O₃ concentrations
331 were about 14.6–15.8 wt% at 1.5 GPa and 950–1000 °C, which then decreased to
332 about 13.4–13.8 wt% at 2.5 GPa and 1000 °C. Both MgO and FeO concentrations
333 decreased with an increase of SiO₂ (Figure 5b, c). In mixed experiments, the MgO
334 and FeO contents varied from ~10–19 wt% and ~6.5–9 wt%, respectively, at 2.5 GPa,
335 then decreased to ~1–10 wt% and ~2–7 wt%, respectively, at 1.5 GPa. In reaction
336 experiments, the experimental melts had the lowest MgO (~0.5–0.8 wt%) and FeO
337 (~1.2–1.8 wt%) concentrations (Figure 5b, c). The CaO, Na₂O and K₂O
338 concentrations in experimental melts showed weak correlations with increased SiO₂
339 and narrower variations of CaO and K₂O in reaction experiments compared with
340 mixed experiments (Figure 5d–f). In reaction experiments, the resultant melts had
341 relatively lower concentrations of MgO and FeO, indicating that they did not achieve
342 chemical equilibrium with the upper harzburgite layer, because the bottom sediment is
343 depleted in MgO and FeO with respect to harzburgite. The hybrid melts in mixed
344 experiments were rich in MgO and FeO, which may represent well-equilibrated
345 products in the residual solid phases.

346

347 **4. Discussion**

348 **4.1 Phlogopite stability in sediment-metasomatized lithospheric mantle**

349 Experimental results in phlogopite + diopside system (Sudo and Tatsumi 1990;
350 Luth 1997), phlogopite-doped peridotite system (Fumagalli et al. 2009; Tumiati et al.
351 2013; Condamine and Médard 2014; Condamine et al. 2016), sediment melt +
352 dunite/lherzolite system (Pirard and Hermann 2015; Mallik et al. 2015, 2016),
353 sediment + dunite system (Förster et al. 2019a, b) and glimmerite + harzburgite
354 system (Förster et al. 2017) have shown that phlogopite is an important K-bearing
355 phase in metasomatized lithospheric mantle or mantle wedge. The stability field of
356 phlogopite ranges from ~950 °C to >1300 °C at pressures of 1–3 GPa. For example,
357 phlogopite was observed as a stable phase at temperatures of 800–1050 °C and 3.5
358 GPa in a sediment melt (25 wt%) + dunite (75 wt%) system (Pirard and Hermann
359 2015); while in a sediment melt (25 wt%) + lherzolite (75 wt%) system, phlogopite
360 stability was thought to be ≤1250–1300 °C at 2–3 GPa (Mallik et al. 2015). In this
361 study, phlogopite was observed at temperatures of no more than 1100–1200 °C (1.5–
362 2.5 GPa) in mixed experiments. The stability field of phlogopite obtained in this study
363 is roughly consistent with experiments conducted by Förster et al. (2017) and
364 Condamine and Médard (2014) in glimmerite + harzburgite and phlogopite-doped
365 peridotite systems, respectively. Figure 6 shows the variations of phase proportions in
366 mixed sediment + harzburgite system as a function of temperature at 1.5 GPa. The
367 proportion of olivine increased with temperature, accompanied by reductions in the
368 orthopyroxene and phlogopite fractions. The disappearance of phlogopite and
369 clinopyroxene was accompanied by the formation of quench melts, indicating that the

370 formation of potassium-rich melts should have a close relationship with the
371 breakdown of phlogopite.

372 In oceanic subduction zones, potassium can be introduced into mantle wedges
373 and lithospheric mantle through slab-derived fluid/melt, continental-derived sediment
374 and phlogopite-bearing peridotite and form a variety of metasomatized zones (e.g.,
375 [Foley 1992, 1993](#); [Prelević et al. 2008](#); [Wang and Foley 2018](#)). Then, partial melting
376 of the metasomatized peridotite may contribute to the origin of potassium-rich
377 magmas in these regions. Therefore, it has been suggested that the breakdown or
378 melting of phlogopite with increased temperature is essential for the generation of
379 potassium-rich magmas in subduction zones and post-collisional settings ([Wyllie and](#)
380 [Sekine 1982](#); [Foley et al. 1996](#); [Prelević et al. 2012](#); [Fritschle et al. 2013](#); [Condamine](#)
381 [and Médard 2014](#); [Mallik et al. 2015](#)). However, phlogopite was also thought to be
382 unnecessary in a mantle source to produce potassium-rich magmas based on mixed
383 experiments in phyllite + dunite system ([Wang et al. 2017](#); [Wang and Foley 2018](#)).
384 They suggested that potassium was introduced by the addition of continental-derived
385 sediment via phengite; the breakdown of phengite could then release a significant
386 amount of potassium into the resulting partial melt. Therefore, this phlogopite-free
387 mantle source may be an alternative explanation for the generation of potassium-rich
388 magmas in post-collisional settings ([Wang et al. 2017](#); [Wang and Foley 2018](#)).
389 Phengite is an important potassium-bearing phase in subducted sediment, oceanic
390 crust and upper continental crust at depths \sim <300 km below surface (e.g., [Schmidt](#)

391 [and Poli 1998](#); [Ono 1998](#); [Hermann and Green 2001](#); [Hermann 2002](#); [Hermann and](#)
392 [Spandler 2008](#); [Wu et al. 2009](#)). However, phengite has never been observed in mixed
393 experiments, such as andesite + lherzolite, sediment + MORB, sediment melt +
394 dunite/lherzolite, metasediment + dunite and sediment-dominated mélange +
395 lherzolite systems (*e.g.*, [Castro et al. 2010](#); [Mallik and Dasgupta 2014](#), [Pirard and](#)
396 [Hermann 2015](#); [Mallik et al. 2015, 2016](#); [Codillo et al., 2018](#)), because they have
397 compositions that are too high in Mg to produce phengite. In fact, neither phengite nor
398 phlogopite has been observed in all experiments conducted by [Wang and Foley \(2018\)](#)
399 in phyllite + dunite systems. We suggest that the absence of phlogopite or phengite in
400 those experiments may indicate a relationship with lower H₂O in the starting material
401 (0.88 wt%), which is not conducive to the formation of these K-bearing minerals
402 because they are hydrous phases. In addition, the limited temperature range (1000–
403 1100 °C) in [Wang and Foley \(2018\)](#) may also be related to the absence of phlogopite
404 or phengite in the run products, because phlogopite has been mostly observed as a
405 stable phase in subsolidus conditions and the stable field of phengite is usually less
406 than ~900 °C at <5 GPa ([Schmidt and Poli 1998](#)). After combining our experimental
407 results with previous studies (*e.g.*, [Wyllie and Sekine 1982](#); [Foley et al. 1996](#); [Prelević](#)
408 [et al. 2012](#); [Fritschle et al. 2013](#); [Condamine and Médard 2014](#); [Mallik et al. 2016](#)),
409 we suggest that phlogopite is an important potassium-bearing phase in
410 sediment/sediment melt-metasomatized mantle at shallow depths because there is
411 little experimental evidence for the presence of phengite, although its presence is

412 unable to be ruled out completely by our experiments. With an increase in
413 temperature (or upwelling), the breakdown or melting of phlogopite may result some
414 potassium-rich melts. These melts could transport sediment signatures and potassium
415 into arc lavas in subduction zones or potassic magmas in post-collisional settings.

416

417 **4.2 Comparison of experimentally formed melts**

418 Experimental melts formed in sediment melt + lherzolite (Mallik et al. 2016),
419 sediment melt + dunite (Pirard and Hermann 2015), mélange + lherzolite (Codillo et
420 al. 2018), phyllite/sediment + dunite (Wang and Foley 2018; Förster et al. 2019a, b)
421 and mélange (Cruz-Uribe et al., 2018) systems were selected for comparison (Figure
422 5). If the mixed and reaction experiments are assumed to be two end-member
423 processes of slab material reacting with lithospheric mantle, a mixing of the resultant
424 melts in our experiments may show similar Al₂O₃ and FeO variations with melts
425 formed in mélange + lherzolite (Codillo et al. 2018) and sediment melt + dunite
426 (Pirard and Hermann 2015) systems (Figure 5a, c). The mixing line of MgO and CaO
427 in experimental melts from this study was roughly consistent with mélange-derived
428 melts (Figure 5b, d; Cruz-Uribe et al. 2018). The CaO concentrations in experimental
429 melts derived from mélange + lherzolite (Codillo et al. 2018) and sediment melt +
430 lherzolite (Mallik et al. 2016) systems were slightly higher than other experiments.
431 Experimental melts derived from mélange (Cruz-Uribe et al. 2018) and sediment melt
432 + dunite (Pirard and Hermann 2015) had slightly higher Na₂O compared with melts

433 from other experiments (Figure 5e). In terms of a phyllite + dunite system (Wang and
434 Foley 2018), the experimental melts exhibited relatively higher SiO₂ because the
435 starting material they use is rich in SiO₂ (~59 wt%). The composition of mélange +
436 lherzolite-derived melts (Codillo et al. 2018) displayed higher MgO and CaO and
437 lower Na₂O and K₂O than melts formed in a mélange system (Cruz-Uribe et al. 2018).
438 In a sediment + dunite system (Förster et al. 2019a, b), the experimental melts
439 exhibited lower concentrations of MgO and FeO (Figure 5b, c), because only layered
440 (reaction) experiments were conducted and the resulting melts may not have
441 equilibrated with the upper dunite layer (rich in MgO and FeO).

442 The N-MORB normalized trace element patterns of experimental melts in this
443 study are shown in Figure 7. Partial melts produced in mixed and reaction
444 experiments exhibited similar patterns for rare earth elements (REEs), large ion
445 lithophile elements (LILEs) and high field strength elements (HFSEs), except for
446 slightly positive Zr-Hf anomalies in melts derived from reaction experiments (Figure
447 7). Recently, Wang and Foley (2018) found that melts formed in mixed experiments
448 are rich in heavy rare earth elements (HREEs) with respect to melts formed in
449 reaction experiments. This may be caused by the presence of higher proportions of
450 residual garnet in reaction experiments, which can preserve a significant portion of
451 the bulk rock HREE; therefore, the corresponding melts are poor in HREEs. The
452 mélange-derived melts exhibited similar trace element patterns with experimental
453 melts from this study (Figure 8a; Castro et al. 2010). Experimental melts formed in a

454 mélange + lherzolite system (Cruz-Urbe et al. 2018) displayed positive Zr-Hf
455 anomalies with respect to mélange-derived melts (Castro et al. 2010); this result may
456 indicate a relationship with relatively higher temperatures (Cruz-Urbe et al. 2018),
457 because it favors the breakdown of zircon and release of Zr-Hf to experimental melts.
458 In a sediment melt + lherzolite system (Mallik et al. 2016), the resultant melts
459 exhibited positive Sr and negative Th anomalies compared with melts in this study
460 (Figure 8b). Experimental melts formed in a phyllite + dunite system also showed
461 similar trace element patterns with melts derived from sediment + harzburgite, except
462 for relatively lower concentrations of HREEs and weak Zr-Hf anomalies (Figure 8c;
463 Wang and Foley 2018). The partial melts derived from reaction experiments between
464 sediment and dunite exhibited positive U and Ta anomalies compared with the melts
465 formed in this study (Figure 8c; Förster et al. 2019a). The melt/fluid from Pirard and
466 Hermann (2015) exhibited higher trace element concentrations and positive U, Ta, Sm
467 and Eu anomalies (Figure 8d); this result may be related to the relatively lower
468 temperatures they used, so that the proportions of the resulting melt/fluid were low
469 and the trace element concentrations in the melt/fluid were high.

470

471 **4.3 Recycling of sediment diapirs in subduction zones**

472 Major element compositions of natural potassium-rich arc lavas from the
473 Aleutian, Sunda, Central American and Mexian subduction zones were selected for
474 comparison with experimental melts (Figure 5). The potassium-rich arc data

475 (K_2O/Na_2O ratios of ≥ 1) were compiled from the GEOROC database
476 (<http://georoc.mpch-mainz.gwdg.de>). The mixing lines of melt composition between
477 mixed and reaction experiments are consistent with natural potassium-rich arc lavas in
478 SiO_2 , Al_2O_3 , MgO , K_2O and a portion of CaO and Na_2O (Figure 5a, b, d–f). At SiO_2
479 concentrations lower than ~51 wt% (Figure 5a), the Al_2O_3 in potassium-rich arc lavas
480 was slightly lower than experimentally formed melt in a sediment + harzburgite
481 system, but consistent with melt derived from sediment melt + lherzolite (Mallik et al.
482 2016). In addition, experimentally formed melts exhibited lower FeO concentrations
483 than potassium-rich arc lavas (Figure 5c); this result may be caused by crystal
484 fractionation in natural arc lavas during ascent to near-surface, because early
485 differentiation could simultaneously increase the total FeO content of the residue
486 magmas and reduce Al_2O_3 (e.g., Stakes et al. 1984). Therefore, it suggests that early
487 crystal fractionation of arc magmas could make variations in FeO and Al_2O_3 approach
488 the compositions of experimental-derived melts. The variations of CaO and Na_2O in
489 potassium-rich arc lavas are slightly higher than experimental melts obtained in this
490 study (Figure 5d, e), but consistent with melts derived from mélange (sediment +
491 oceanic crust) + lherzolite (Codillo et al. 2018) and sediment melt (CaO - Na_2O -rich) +
492 lherzolite (Mallik et al. 2016) systems. This may indicate that a variety of CaO and
493 Na_2O -rich components, such as oceanic crust, may be involved in the origins of such
494 arc lavas. Therefore, we suggest that recycled bulk sediment alone is insufficient to
495 explain the major element compositions of potassium-rich arc lavas; a variety of

496 basaltic crust (or melt derived from basaltic crust) components may also be involved
497 in the mantle source of these arc lavas.

498 To further constrain the involvement of bulk sediment diapirs in the genesis of
499 potassium-rich arc lavas, the trace element compositions of mixed and reaction melts
500 in this study were compared with natural potassium-rich arc lavas from subduction
501 zones (Figure 9). The trace element pattern of potassium-rich lavas from the Mexican
502 and Sunda subduction zones are consistent with the melts derived from sediment +
503 harzburgite experiments in this study (Figure 9a, b). The slightly positive Zr-Hf
504 anomalies in some of the arc lavas from the Mexican subduction zone can be
505 interpreted as melts from reaction experiments in sediment + harzburgite or
506 sediment-dominated mélange systems (Figure 8a). For potassium-rich arc lavas from
507 the Central American and Aleutian subduction zones, variations of Rb and Ba (or Th)
508 are inconsistent with experimental melts from both mixed and reaction experiments
509 (Figure 9c, d). This discrepancy can be explained by the involvement of mélange or
510 sediment melt in the mantle source of these magmas (Figure 8a–b). The similar trace
511 element patterns of melts derived from sediment + harzburgite experiments and
512 natural potassium-rich arc lavas provide evidence for the involvement of bulk
513 sediment diapirs in the mantle source of these arc lavas.

514

515 **5. Implications**

516 The transfer mechanisms of continental-derived sediments from a subducting
517 slab to the overlying mantle wedge or lithospheric mantle include
518 metamorphism-induced melts or fluids ([Plank and Langmuir 1993](#); [Peate et al. 1997](#);
519 [Porter and White 2009](#); [Turner et al. 2012](#); [Pirard and Hermann 2015](#)), bulk sediment
520 diapirs ([Behn et al., 2011](#); [Nielsen and Marschall 2017](#)), and sediment-dominated
521 mélanges ([Codillo et al. 2018](#); [Cruz-Uribe et al. 2018](#)); however, the dominant
522 transfer mechanism for this material is still debated. Previous studies have suggested
523 that bulk sediment diapirs can be introduced into the bottom of lithospheric mantle
524 during slab subduction and form variable mélanges. Relaminated sediment
525 components may significantly affect the physical and chemical properties of
526 lithosphere (harzburgite), because they have different major and trace element
527 compositions. Partial melting of these mélanges may result in magmas with different
528 potassium concentrations, depending on the stability or breakdown of phlogopite. The
529 new data from this study have revealed similar trace element patterns for the partial
530 melts derived from sediment + harzburgite mélanges and natural potassium-rich arc
531 lavas from subduction zones, indicating that bulk sediment diapirs may be a possible
532 transfer mechanism for slab material from a subducting plate to the upper mantle
533 wedge/lithospheric mantle. Therefore, these experimental data may help to understand
534 the partial melting phase relations of sediment-modified lithospheric mantle and
535 provide constraints on the evolution of lithosphere and the generation of
536 potassium-rich arc lavas in oceanic subduction zones.

537

538 **Acknowledgments**

539 We thank Jihao Zhu and Wei Li for technical support during EPMA analyses. We
540 acknowledge the use of EPMA in the Key Laboratory of Submarine Geosciences,
541 State Oceanic Administration. We thank Junlong Yang, Zhenjiang Wang, Xiangfa
542 Wang, Juan Wang, Xingdong Zhou and Zhonghang Wang for technical support
543 during high-pressure experiments and SEM analyses. We are grateful to Fred Davis,
544 Ananya Mallik and two anonymous reviewers for their constructive suggestions and
545 comments. Editorial handling by Don R. Baker and Sylvie Demouchy is greatly
546 appreciated. This research was supported by the National Programme on Global
547 Change and Air-Sea Interaction (GASI-GEOGE-02), National Natural Science
548 Foundation of China (41772040, 91858104).

549

550 **References**

- 551 Behn, M.D., Kelemen, P.B., Hirth, G., Hacker, B.R., and Massonne, H.J. (2011)
552 Diapirs as the source of the sediment signature in arc lavas. *Nature Geoscience*,
553 4, 641–646.
- 554 Bindeman, I.N., Eiler, J.M., Yogodzinski, G.M., Tatsumi, Y., Stern, C.R., Grove, T.L.,
555 Portnyagin, M., Hoernle, K., and Danyushevsky, L.V. (2005) Oxygen isotope
556 evidence for slab melting in modern and ancient subduction zones. *Earth and*
557 *Planetary Science Letters*, 235, 480–496.

- 558 Blundy, J.D., and Sparks, R.S.J. (1992) Petrogenesis of mafic inclusions in granitoids
559 of the Adamello Massif, Italy. *Journal of Petrology*, 33, 1039–1104.
- 560 Castro, A., Gerya, T., Garcia-Casco, A., Fernandez, C., Diaz-Alvarado, J.,
561 Moreno-Ventas, I., and Low, I. (2010) Melting relations of MORB-sediment
562 mélanges in underplated mantle wedge plumes: Implications for the origin of
563 cordilleran-type batholiths. *Journal of Petrology*, 51, 1267–1295.
- 564 Codillo, E.A., Le Roux, V., and Marschall, H.R. (2018) Arc-like magmas generated
565 by mélange-peridotite interaction in the mantle wedge. *Nature Communications*,
566 9(1), 2864.
- 567 Condamine, P., and Médard, E. (2014) Experimental melting of phlogopite-bearing
568 mantle at 1 GPa: Implications for potassic magmatism. *Earth and Planetary
569 Science Letters*, 397, 80–92.
- 570 Condamine, P., Médard, E., and Devidal, J.L. (2016) Experimental melting of
571 phlogopite-peridotite in the garnet stability field. *Contributions to Mineralogy
572 and Petrology*, 171(11), 95.
- 573 Corgne, A., Schilling, M.E., Grégoire, M., and Langlade, J. (2018) Experimental
574 constraints on metasomatism of mantle wedge peridotites by hybridized adakitic
575 melts. *Lithos*, 308-309, 213–226.
- 576 Duggen, S., Portnyagin, M., Baker, J., Ulfbeck, D., Hoernle, K., Garbe-SchoÅNnberg,
577 D., and Grassineau, N. (2007) Drastic shift in lava geochemistry in the
578 volcanic-front to near-arc region of the Southern Kamchatkan subduction zone:

- 579 Evidence for the transition from slab surface dehydration to sediment melting.
580 *Geochimica et Cosmochimica Acta*, 71, 452–480.
- 581 Foley, S.F. (1992) Vein-plus-wall-rock melting mechanisms in the lithosphere and the
582 origin of potassic alkaline magmas. *Lithos*, 28, 435–453.
- 583 Foley, S.F. (1993) An experimental study of olivine lamproite—first results from the
584 diamond stability field. *Geochimica et Cosmochimica Acta*, 57, 483–489.
- 585 Foley, S.F., Jackson, S.E., Fryer, B.J., Greenough, J.D., and Jenner, G.A. (1996)
586 Trace element partition coefficients for clinopyroxene and phlogopite in an
587 alkaline lamprophyre from Newfoundland by LAM-ICP-MS. *Geochimica et*
588 *Cosmochimica Acta*, 604, 629–638.
- 589 Förster, M.W., Prelević, D., Buhre, S., Mertz-Kraus, R., and Foley, S.F. (2019a) An
590 experimental study of the role of partial melts of sediments versus mantle melts
591 in the sources of potassic magmatism. *Journal of Asian Earth Sciences*, 177, 76–
592 88.
- 593 Förster, M.W., Foley, S.F., Alard, O., and Buhre, S. (2019b) Partitioning of nitrogen
594 during melting and recycling in subduction zones and the evolution of
595 atmospheric nitrogen. *Chemical Geology*, 525, 334–342.
- 596 Förster, M.W., Prelević, D., Schmück, H.R., Buhre, S., Veter, M., Mertz-Kraus, R.,
597 Foley, S.F., and Jacob, D.E. (2017) Melting and dynamic metasomatism of
598 mixed harzburgite + glimmerite mantle source: Implications for the genesis of
599 orogenic potassic magmas. *Chemical Geology*, 455, 182–191.

- 600 Fritschle, T., Prelevic, D., Foley, S.F., and Jacob, D.E. (2013) Petrological
601 characterization of the mantle source of Mediterranean lamproites: Indications
602 from major and trace elements of phlogopite. *Chemical Geology*, 353, 267–279.
- 603 Fumagalli, P., Zanchetta, S., and Poli, S. (2009) Alkali in phlogopite and amphibole
604 and their effects on phase relations in metasomatized peridotites: a high-pressure
605 study. *Contributions to Mineralogy and Petrology*, 158, 723–737.
- 606 Gervasoni, F., Klemme, S., Rohrbach, A., Grützner, T., and Berndt, J. (2017)
607 Experimental constraints on mantle metasomatism caused by silicate and
608 carbonate melts. *Lithos*, 282-283, 173–186.
- 609 Hacker, B.R., Kelemen, P.B., and Behn, M.D. (2011) Differentiation of the
610 continental crust by relamination. *Earth and Planetary Science Letters*, 307,
611 501–516.
- 612 Harvey, J., Garrido, C.J., Savov, I., Agostini, S., Padro, J., Navata, J.A., Marchesi,
613 C., Sánchez-Vizcaíno, V.L., and Gómez-Pugnaire, M.T. (2014)
614 ¹¹B-rich fluids in subduction zones: The role of antigorite dehydration in
615 subducting slabs and boron isotope heterogeneity in the mantle. *Chemical*
616 *Geology*, 376, 20–30.
- 617 Hermann, J. (2002) Experimental constraints on phase relations in subducted
618 continental crust. *Contributions to Mineralogy and Petrology*, 143, 219–235.

- 619 Herrmann, W., and Berry, R.F. (2002) MINSQ—a least squares spreadsheet method
620 for calculating mineral proportions from whole rock major element analyses.
621 Geochemistry: Exploration, Environment, Analysis, 2, 361–368.
- 622 Hermann, J., and Green, D.H. (2001) Experimental constraints on high pressure
623 melting in subducted crust. Earth and Planetary Science Letters, 188, 149–168.
- 624 Hermann, J., and Rubatto, D. (2009) Accessory phase control on the trace element
625 signature of sediment melts in subduction zones. Chemical Geology, 265, 512–
626 526.
- 627 Hermann, J., and Spandler, C.J. (2008) Sediment melts at sub-arc depths: An
628 experimental study. Journal of Petrology, 49, 717–740.
- 629 Kendrick, M.A., Arculus, R.J., and Danyushevsky, L.V. (2014) Subduction-related
630 halogens (Cl, Br, and I) and H₂O in magmatic glasses from Southwest Pacific
631 Backarc Basins. Earth and Planetary Science Letters, 400, 165–176.
- 632 Liu, Y., Hu, Z., Gao, S., Günther, D., Xu, J., Gao, C., and Chen, H. (2008) In situ
633 analysis of major and trace elements of anhydrous minerals by LA-ICP-MS
634 without applying an internal standard. Chemical Geology, 257, 34–43.
- 635 Longerich, H.P., Jackson, S.E., and Günther, D. (1996) Laser ablation inductively
636 coupled plasma mass spectrometric transient signal data acquisition and analyte
637 concentration calculation. Journal of Analytical Atomic Spectrometry, 11(9),
638 899–904.

- 639 Luth, R.W. (1997) Experimental study of the system phlogopite diopside from 3.5 to
640 17 GPa. *American Mineralogist*, 82, 1198–1209.
- 641 Mallik, A., and Dasgupta, R. (2013) Reactive infiltration of MORB-eclogite derived
642 carbonated silicate melt into fertile peridotite at 3 GPa and genesis of alkalic
643 magmas. *Journal of Petrology*, 54, 2267–2300.
- 644 Mallik, A., and Dasgupta, R. (2014) Effect of variable CO₂ on eclogite derived
645 andesite and lherzolite reaction at 3 GPa: Implications for mantle source
646 characteristics of alkalic ocean island basalts. *Geochemistry Geophysics
647 Geosystems*, 15, 1533–1557.
- 648 Mallik, A., Dasgupta, R., Tsuno, K., and Nelson, J. (2016) Effects of water, depth and
649 temperature on partial melting of mantle-wedge fluxed by hydrous
650 sediment-melt in subduction zones. *Geochimica et Cosmochimica Acta*, 195,
651 226–243.
- 652 Mallik, A., Nelson, J., and Dasgupta, R. (2015) Partial melting of fertile peridotite
653 fluxed by hydrous rhyolitic melt at 2–3 GPa: Implications for mantle wedge
654 hybridization by sediment melt and generation of ultrapotassic magmas in
655 convergent margins. *Contributions to Mineralogy and Petrology*, 169, 48.
- 656 Marschall, H.R., and Schumacher, J.C. (2012) Arc magmas sourced from mélange
657 diapirs in subduction zones. *Nature Geoscience*, 5, 862–867.
- 658 Maunder, B., and van Hunen, J. (2016) Relamination of mafic subducting crust
659 throughout Earth's history. *Earth and Planetary Science Letters*, 449, 206–216.

- 660 Mukherjee, P.K., Khanna, P.P., and Saini, N.K. (2014) Rapid determination of trace
661 and ultra trace level elements in diverse silicate rocks in pressed powder pellet
662 targets by LA-ICP-MS using a matrix-independent protocol. *Geostandards and*
663 *Geoanalytical Research*, 38 (3), 363–379.
- 664 Nielsen, S.G., and Marschall, H.R. (2017) Geochemical evidence for mélange melting
665 in global arcs. *Science Advance*, 3, e1602402.
- 666 Ono, S. (1998) Stability limits of hydrous minerals in sediment and mid-ocean ridge
667 basalt compositions: Implications for water transport in subduction zones.
668 *Journal of Geophysical Research*, 103, 18253–18267.
- 669 Peate, D.W., Pearce, J.A., Hawkesworth, C.J., Colley, H., Edwards, C.M.H., and
670 Hirose, K. (1997) Geochemical variations in Vanuatu arc lavas: The role of
671 subducted material and a variable mantle wedge composition. *Journal of*
672 *Petrology*, 38, 1331–1358.
- 673 Pirard, C., and Hermann, J. (2015) Experimentally determined stability of alkali
674 amphibole in metasomatised dunite at sub-arc pressures. *Contributions to*
675 *Mineralogy and Petrology*, 169, 1–26.
- 676 Plank, T., and Langmuir, C.H. (1993) Tracing trace elements from sediment input to
677 volcanic output at subduction zones. *Nature*, 362, 739–743.
- 678 Plank, T., and Langmuir, C.H. (1998) The chemical composition of subducting
679 sediment: implications for the crust and mantle. *Chemical Geology*, 145, 325–
680 394.

- 681 Porter, K.A., and White, W.M. (2009) Deep mantle subduction flux. *Geochemistry*
682 *Geophysics Geosystems*, 10, 1–23.
- 683 Prelević, D., Foley, S.F., Romer, R., and Conticelli, S. (2008) Mediterranean tertiary
684 lamproites derived from multiple source components in post-collisional
685 geodynamics. *Geochimica et Cosmochimica Acta*, 72, 2125–2156.
- 686 Prelević, D., Akal, C., Foley, S.F., Romer, R.L., Stracke, A., and van den Bogaard, P.
687 (2012) Ultrapotassic mafic rocks as geochemical proxies for post-collisional
688 dynamics of orogenic lithospheric mantle: The case of southwestern Anatolia,
689 Turkey. *Journal of Petrology*, 53, 1019–1055.
- 690 Prouteau, G., Scaillet, B., Pichavant, M., and Maury, R. (2001) Evidence for mantle
691 metasomatism by hydrous silicic melts derived from subducted oceanic crust.
692 *Nature*, 410, 197–200.
- 693 Scambelluri, M., Pettke, T., and Cannàò, E. (2015) Fluid-related inclusions in Alpine
694 high-pressure peridotite reveal trace element recycling during subduction-zone
695 dehydration of serpentinized mantle (Cima di Gagnone, Swiss Alps). *Earth and*
696 *Planetary Science Letters*, 429, 45–59.
- 697 Schmidt, M.W. (2015) Melting of pelitic sediments at subarc depths: 2. Melt
698 chemistry, viscosities and a parameterization of melt composition. *Chemical*
699 *geology*, 404, 168–182.

- 700 Schmidt, M.W., and Poli, P. (1998) Experimentally based water budgets for
701 dehydrating slabs and consequences for arc magma generation. *Earth and*
702 *Planetary Science Letters*, 163, 361–379.
- 703 Schmidt, M.W., and Poli, S. (2014) Devolatilization during subduction. In H.H.D.
704 Holland, K.K. Turekian, Ed., *Treatise on Geochemistry (Second Edition)*, p.
705 669–701, Pergamon, Oxford.
- 706 Shimoda, G., Tatsumi, Y., Nohda, S., Ishizaka, K., and Jahn, B.M. (1998) Setouchi
707 high-Mg andesites revisited: Geochemical evidence for melting of subducting
708 sediments. *Earth and Planetary Science Letters*, 160, 479–492.
- 709 Shu, Y., Nielsen, S.G., Zeng, Z., Shinjo, R., Blusztajn, J., Wang, X., and Chen, S.
710 (2017) Tracing subducted sediment inputs to the Ryukyu arc-Okinawa Trough
711 system: Evidence from thallium isotopes. *Geochimica et Cosmochimica Acta*,
712 217, 462–491.
- 713 Spandler, C., and Pirard, C. (2013) Element recycling from subducting slabs to arc
714 crust: A review. *Lithos*, 170-171, 208–223.
- 715 Stakes, D.S., Shervais, J.W. and Hopson, C.A. (1984) The volcanic-tectonic cycle of
716 the FAMOUS and AMAR Valleys, Mid-Atlantic Ridge (36°47'N): Evidence
717 from basalt glass and phenocryst compositional variations for a steady state
718 magma chamber beneath the valley midsections, AMAR 3. *Journal of*
719 *Geophysical Research: Solid Earth*, 89, 6995–7028.

- 720 Sudo, A., and Tatsumi, Y. (1990) Phlogopite and K-amphibole in the upper mantle:
721 Implication for magma genesis in subduction zones. *Geophysics Research Letter*,
722 17, 29–32.
- 723 Tatsumi, Y., Hamilton, D.L., and Nesbitt, R.W. (1986) Chemical characteristics of
724 fluid phase released from a subducted lithosphere and origin of arc magmas:
725 Evidence from high-pressure experiments and natural rocks. *Journal of*
726 *Volcanology and Geothermal Research*, 29, 293–309.
- 727 Tumiami, S., Fumagalli, P., Tiraboschi, C., and Poli, S. (2013) An experimental study
728 on COH-bearing peridotite up to 3·2GPa and implications for crust–mantle
729 recycling. *Journal of Petrology*, 54, 453–479.
- 730 Turner, S., Caulfield, J., Turner, M., van Keken, P., Maury, R., Sandiford, M., and
731 Prouteau, G. (2012) Recent contribution of sediments and fluids to the mantle's
732 volatile budget: *Nature Geoscience*, 5, 50–54.
- 733 Ulmer, P., and Trommsdorff, V. (1995) Serpentine stability to mantle depths and
734 subduction-related magmatism. *Science*, 268, 858–861.
- 735 Wang, C., Jin, Z., Gao, S., Zhang, J. and Zheng, S. (2010) Eclogite-melt/peridotite
736 reaction: Experimental constrains on the destruction mechanism of the North
737 China Craton. *Science China: Earth Sciences*, 53, 797–809.
- 738 Wang, X., Wang, Y., Gao, Y., Huang, Y., Wang, Z., and Shi, X. (2009) Preparation of
739 five China Sea and continental shelf sediment reference materials (MSCS-1–5)

- 740 with ultra-fineparticle size distributions. *Geostandards and Geoanalytical*
741 *Research*, 33, 357–368.
- 742 Wang, Y., Foley, S.F., and Prelević, D. (2017) Potassium-rich magmatism from a
743 phlogopite-free source. *Geology*, 45, 467–470.
- 744 Wang, Y., and Foley, S.F. (2018) Hybridization melting between continent-derived
745 sediment and depleted peridotite in subduction zones. *Journal of Geophysical*
746 *Research: Solid Earth*, 123, 3414–3429.
- 747 Wu, Y., Fei, Y., Jin, Z., and Liu, X. (2009) The fate of subducted Upper Continental
748 Crust: An experimental study. *Earth and Planetary Science Letters*, 282, 275–
749 284.
- 750 Wyllie, P.J., and Sekine, T. (1982) The formation of mantle phlogopite in subduction
751 zone hybridization. *Contributions to Mineralogy and Petrology*, 79, 375–380.
- 752 Zhu, L., Liu, Y., Hu, Z., Hu, Q., Tong, X., Zong, K., Chen, H., and Gao S. (2013)
753 Simultaneous Determination of Major and Trace Elements in Fused Volcanic
754 Rock Powders Using a Hermetic Vessel Heater and LA-ICP-MS. *Geostandards*
755 *and Geoanalytical Research*, 37(2), 207–229.
- 756
- 757

758 **Figure captions**

759 **Figure 1.** Backscattered electron images of representative run products. (a) and (b)
760 Mixed experiments conducted at 1.5 GPa and 1100 °C (PC431) and 1.5 GPa and 1300
761 °C (PC463), respectively; melt + phlogopite or melt coexisting with olivine +
762 orthopyroxene was observed in the run products. (c) Mixed experiment conducted at
763 2.5 GPa and 1100 °C (R908), showing that phlogopite and trace garnet coexisting
764 with olivine and orthopyroxene. (d) Mixed experiment at 2.5 GPa and 1200 °C (R957),
765 showing that an amount of quench melt was segregated along the sample capsule. (e)
766 and (f) Reaction experiment carried out at 1.5 GPa and 950 °C (PC370), showing a
767 reaction zone composed mainly of clinopyroxene and amphibole.

768

769 **Figure 2.** Experimental *P-T* phase diagram in the sediment + harzburgite system
770 (mixed experiments). Major phases are shown in squares. The approximate solidus
771 and stability limits of phlogopite were estimated from the experimental results.

772

773 **Figure 3.** Chemical compositions of minerals and melt in mixed experiments. (a) and
774 (b) Variations of Mg# in olivine and orthopyroxene versus temperature; Fe-loss was
775 corrected by adding Fe to olivine and orthopyroxene based on the partitioning of Fe
776 between solid phases and melt. (c) to (f) Variations of Na₂O, K₂O, TiO₂ and NiO
777 versus temperature in phlogopite. (g) and (h) Variations of K₂O+Na₂O and K₂O/Na₂O
778 versus temperature in melts.

779

780 **Figure 4.** SiO₂-(K₂O+Na₂O) diagram of melt compositions obtained in both mixed
781 and reaction experiments. *R-exp*, melt in reaction experiments; *M-1.5(Phl)*, melt in
782 mixed experiments at 1.5 GPa with phlogopite in the residue minerals (PC431); *M-1.5*,
783 melt in mixed experiments at 1.5 GPa, phlogopite was not present or only very trace
784 amounts were present in the residue phases (PC460, PC463); *M-2.5*, melt in mixed
785 experiments at 2.5 GPa, phlogopite was not present in the residue phases (R957,
786 R1010).

787

788 **Figure 5.** Major element compositions of experimental melts and potassium-rich arc
789 lavas from this study and literatures. Major element variations of (a) Al₂O₃, (b) MgO,
790 (c) FeO, (d) CaO, (e) Na₂O and (f) K₂O versus SiO₂ for experimental melts and
791 natural potassium-rich arc lavas. Mélange + lherzolite data were from [Codillo et al.](#)
792 [\(2018\)](#); mélange ([Cruz-Urbe et al. 2018](#)); sediment melt + lherzolite ([Mallik et al.](#)
793 [2016](#)); sediment melt + dunite ([Pirard and Hermann 2015](#)); phyllite + dunite ([Wang](#)
794 [and Foley 2018](#)); sediment + dunite ([Förster et al. 2019a, b](#)). Experimental melts
795 obtained in this study were corrected for Fe-loss, by adding FeO back to experimental
796 melts. The purple shadowed zone represents natural potassium-rich arc lavas with
797 K₂O/Na₂O ratios ≥ 1 from the Mexian, Sunda, Central American and Aleutian
798 subduction zones, compiled from the GEOROC database
799 (<http://georoc.mpch-mainz.gwdg.de>).

800

801 **Figure 6.** Weight proportions of the phases in mixed experiments versus temperature
802 at 1.5 GPa.

803

804 **Figure 7.** Normal mid-oceanic ridge basalt (N-MORB) normalized trace element
805 compositions of experimental melts. Data for N-MORB were from [Sun and](#)
806 [McDonough \(1989\)](#).

807

808 **Figure 8.** N-MORB normalized trace element compositions of experimental melts
809 from this study and literatures. Experimental melts in (a) mélange ([Cruz-Uribe et al.](#)
810 [2018](#)) or mélange + lherzolite ([Codillo et al. 2018](#)), (b) and (d) sediment melt +
811 lherzolite/dunite ([Mallik et al. 2016](#); [Pirard and Hermann 2015](#)), (c) phyllite + dunite
812 ([Wang and Foley 2018](#)) and sediment + dunite ([Förster et al. 2019a](#)). Melts in
813 M-experiments/ R-experiments indicate melts from mixed/reaction experiments in
814 this study.

815

816 **Figure 9.** Comparison of trace element patterns in the experimental melts of this
817 study and natural potassium-rich arc lavas. Arc lavas from the (a) Mexian, (b) Sunda,
818 (c) Central American and (d) Aleutian subduction zones.

819

820

821

822 Table 1. Compositions of starting materials used in this study and previous reports.

| | MSCS-3 | GLOSS | Hz13 | HS-1A | SPMHKLB4 | SPMHKLB6 | Wang18 | PER-SED | PH-high | PH-low |
|--------------------------------|--------|--------|-------|-------|----------|----------|--------|---------|---------|--------|
| SiO ₂ | 57.72 | 58.57 | 43.86 | 47.20 | 51.81 | 50.44 | 59.43 | 47.80 | 48.60 | 47.03 |
| TiO ₂ | 0.80 | 0.62 | 0.04 | 0.23 | 0.22 | 0.21 | 0.37 | 0.28 | 0.13 | 0.11 |
| Al ₂ O ₃ | 16.95 | 11.91 | 0.72 | 4.78 | 6.37 | 5.81 | 5.90 | 5.83 | 3.47 | 2.72 |
| Cr ₂ O ₃ | - | - | 0.06 | 0.05 | 0.24 | 0.26 | 0.19 | - | - | - |
| FeO ^T | 6.08 | 5.21 | 9.23 | 8.44 | 6.38 | 6.73 | 5.88 | 7.27 | 6.93 | 6.92 |
| MnO | 0.21 | 0.32 | 0.14 | 0.16 | 0.10 | 0.12 | 0.10 | 0.13 | 0.09 | 0.10 |
| MgO | 2.74 | 2.48 | 45.46 | 34.78 | 29.70 | 31.56 | 25.32 | 33.40 | 37.42 | 37.42 |
| CaO | 2.82 | 5.95 | 0.08 | 0.77 | 2.54 | 2.64 | 0.90 | 4.06 | 0.23 | 0.33 |
| Na ₂ O | 2.57 | 2.43 | 0.01 | 0.65 | 1.02 | 0.89 | 0.84 | 0.89 | 0.53 | 1.21 |
| K ₂ O | 3.40 | 2.04 | - | 0.85 | 1.61 | 1.31 | 0.95 | 0.33 | 1.51 | 0.73 |
| P ₂ O ₅ | 0.18 | 0.19 | - | 0.05 | 0.05 | 0.04 | - | 0.01 | - | - |
| LOI | 7.02 | 10.30 | - | 1.76 | 4.00 | 6.00 | 0.88 | 0.34 | 1.25 | 3.75 |
| Total | 100.00 | 100.02 | 99.60 | 99.70 | 104.04 | 106.01 | 100.73 | 100.35 | 100.15 | 100.31 |

823 *HS-1A*, the hybrid sample powder used in this study, synthesized by mixing 25 wt% sediment (MSCS-3) with 75

824 wt% harzburgite (Hz13).

825 *SPMHKLB4* and *SPMHKLB6*, synthesized by mixing 25 wt% sediment-melt with 75 wt% lherzolite, from Mallik

826 et al. (2016).

827 *Wang18*, synthesized by mixing 50 wt% phyllite with 50 wt% dunite, from Wang et al. (2018).828 *PER-SED*, synthesized by mixing 15 wt% mélange with 85 wt% lherzolite, from Codillo et al. (2018).829 *PH-high* and *PH-low*, synthesized by mixing 25 wt% sediment-melt with 75 wt% olivine, from Pirard and

830 Hermann (2015).

831 FeO^T, total Fe was calculated as FeO.

832

833 Table 2. Summary of experimental conditions and run products.

| Run no. | T (°C) | P (GPa) | D (hrs) | Geometry | Phase Modes (wt%) | | | | | | | | |
|---------|--------|---------|---------|----------|-------------------|-------|------|-------|------|-------|--------------|---------|--|
| | | | | | Ol | Opx | Cpx | Phl | Grt | melt | Σr^2 | Fe-loss | other minerals |
| PC307 | 800 | 1.5 | 113 | Mixture | 29(2) | 54(2) | 4(2) | 12(1) | | | 0.7±0.5 | 19(2) | Mag, Hem, Rut |
| PC322 | 900 | 1.5 | 72 | Mixture | 30(3) | 53(3) | 5(2) | 11(1) | | | 0.6±0.2 | 23(3) | Mag, Hem, Apa |
| PC368 | 950 | 1.5 | 60 | Mixture | 31(2) | 49(2) | 7(1) | 12(1) | | | 0.3±0.3 | 18(0) | Mag, Hem, Zir |
| PC365 | 1000 | 1.5 | 48 | Mixture | 32(3) | 52(4) | 6(2) | 11(2) | | | 0.3±0.1 | 17(1) | Mag, Hem, |
| PC431 | 1100 | 1.5 | 48 | Mixture | 34(3) | 51(4) | | 8(1) | | 5(1) | 0.3±0.1 | 17(1) | Mag |
| PC460 | 1200 | 1.5 | 53 | Mixture | 35(1) | 50(1) | | | | 13(1) | 0.2±0.1 | 26(1) | |
| PC463 | 1300 | 1.5 | 44 | Mixture | 46(2) | 35(5) | | | | 18(3) | 1.0±1.0 | 23(5) | |
| R908 | 1100 | 2.5 | 30 | Mixture | 27(1) | 56(1) | | 9(1) | 7(1) | ‡ | 0.4±0.1 | 21(1) | |
| R957 | 1200 | 2.5 | 26 | Mixture | 34(1) | 52(2) | | ‡ | | 14(3) | 0.3±0.1 | 18(2) | |
| R1010 | 1300 | 2.5 | 30 | Mixture | 32(3) | 46(3) | | | | 21(2) | 0.2±0.1 | 32(1) | |
| PC370 | 950 | 1.5 | 53 | Layer | | | | | | | | | Harzburgite: Ol + Opx + Sp; Sediment: Grt + Pl + melt; Reaction zone: Cpx + Amp; |
| PC478 | 1000 | 1.5 | 48 | Layer | | | | | | | | | Harzburgite: Ol + Opx; Sediment: Grt + Cpx + melt; Reaction zone: Opx |
| R1028 | 1000 | 2.5 | 30 | Layer | | | | | | | | | Harzburgite: Ol + Opx; Sediment: Grt + melt; Reaction zone: Opx |

834 Mass balance calculations were performed to estimate the phase proportions (wt%) in mixed experiments.

835 ‡, Indicates trace presence of a phase which could not be detected by mass balance.

836 Σr^2 , Sum of residual square.

837 Fe-loss, Indicates the relative Fe-loss to the Au-Pd capsules with respect to the bulk Fe concentration in the starting material, as estimated by mass balance.

839 Mineral abbreviations: *Ol*, olivine; *Opx*, orthopyroxene; *Cpx*, clinopyroxene; *Phl*, phlogopite; *Mag*, magnesite;

840 *Hem*, hematite; *Rut*, rutile; *Apa*, apatite; *Zir*, zircon; *Grt*, garnet; *Sp*, spinel; *Pl*, plagioclase; *Amp*, amphibole.

841

842

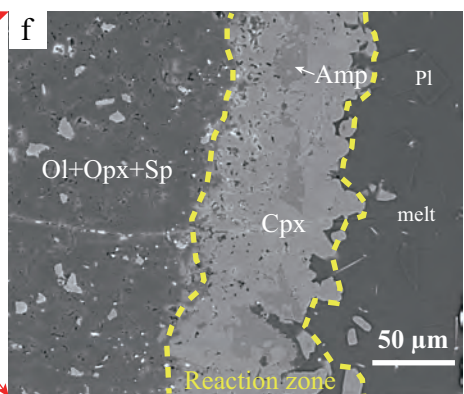
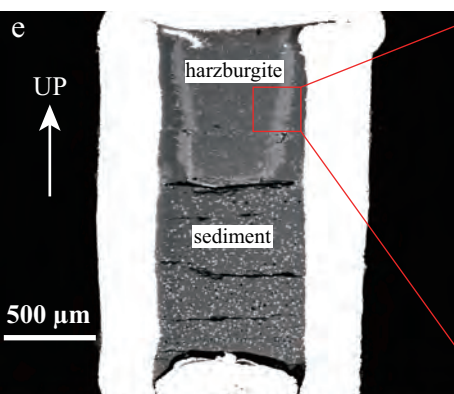
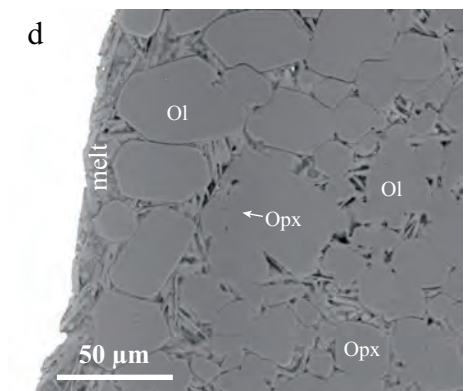
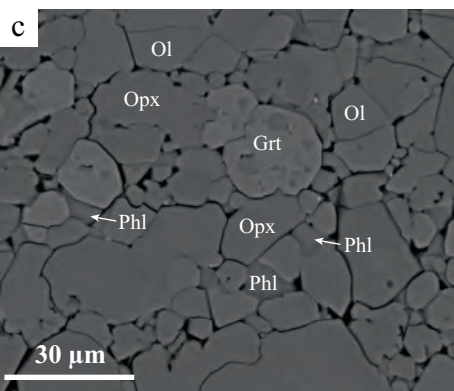
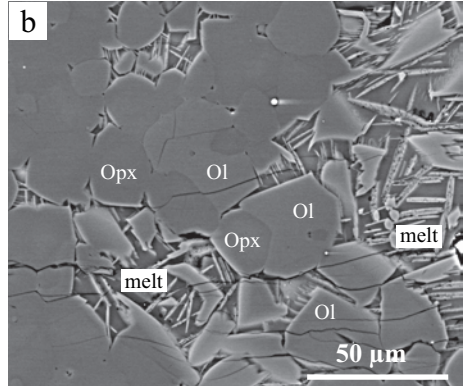
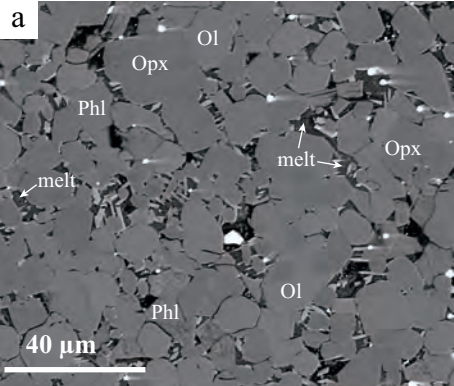


Figure 1

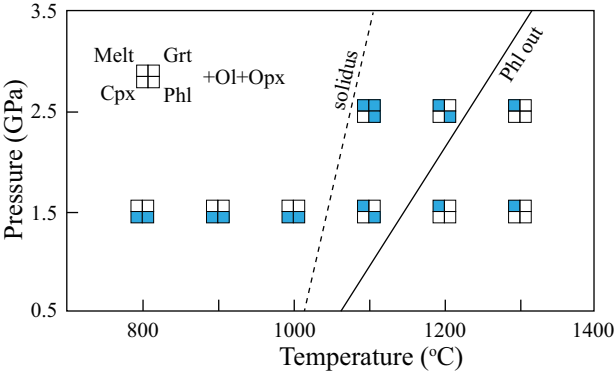


Figure 2

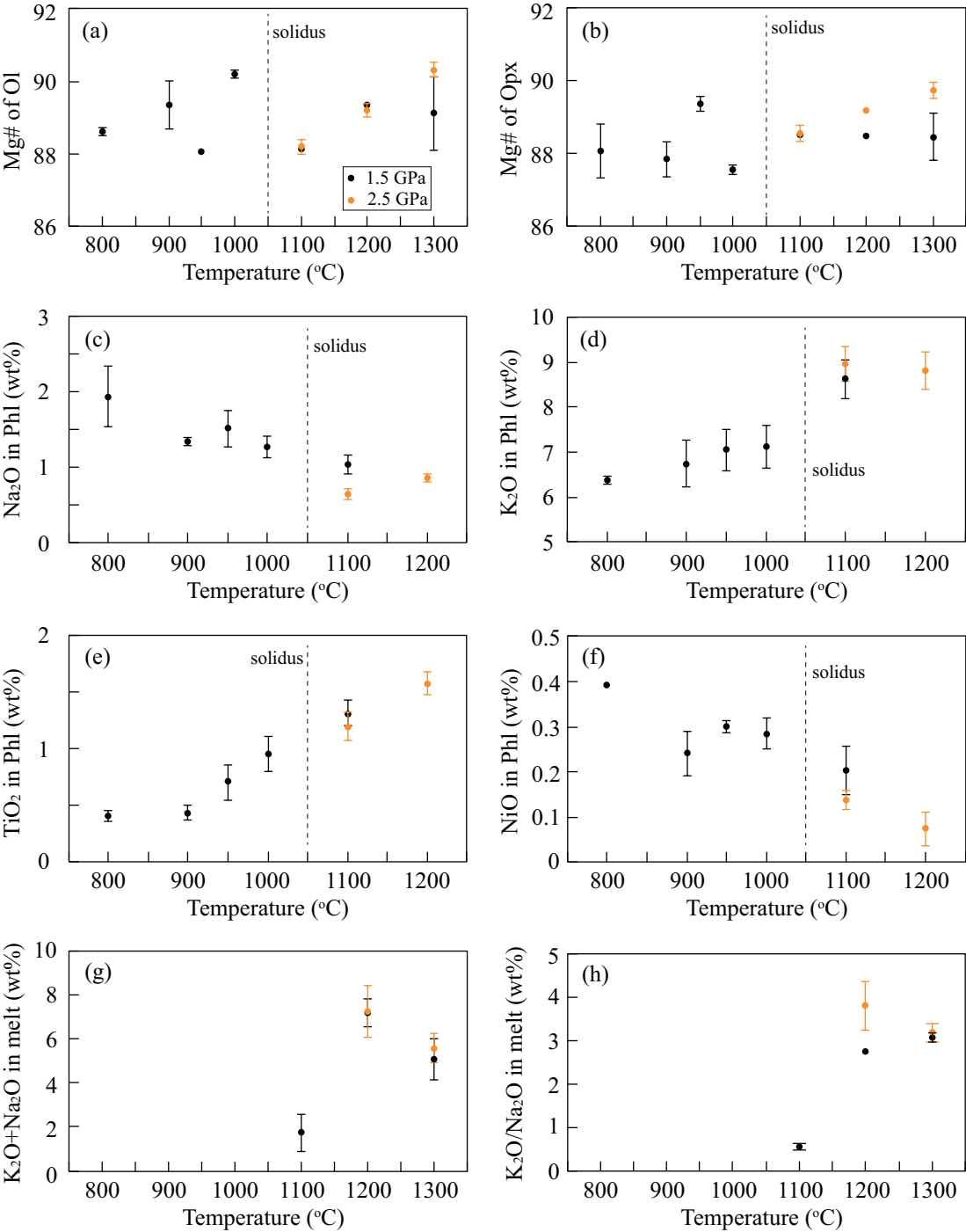


Figure 3

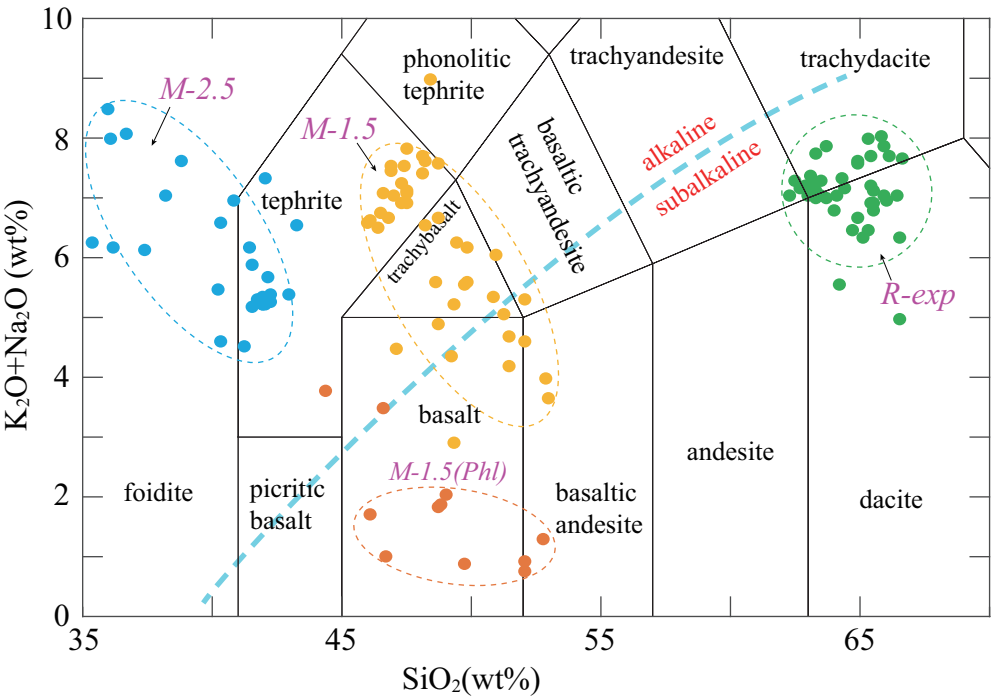


Figure 4

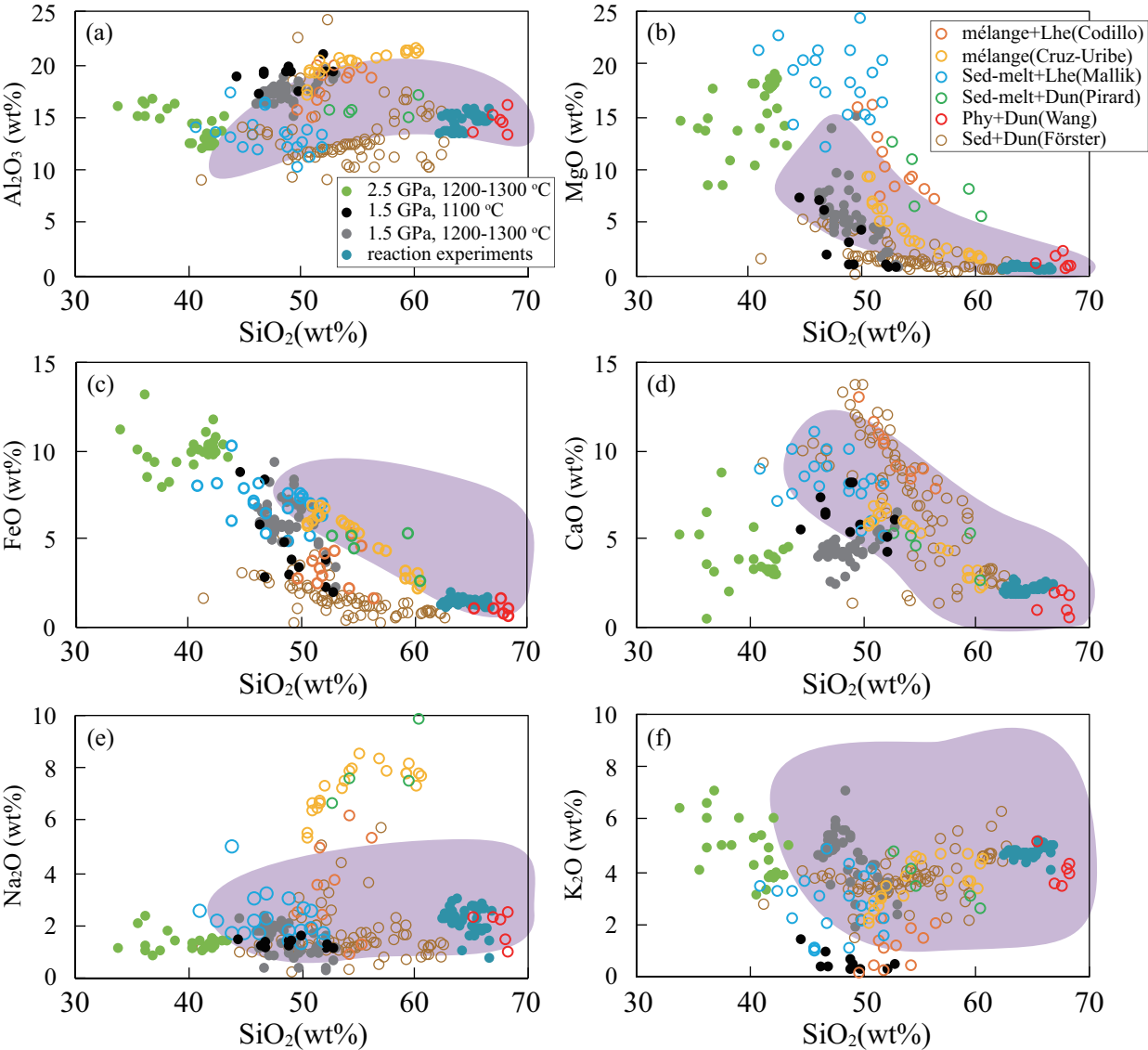


Figure 5

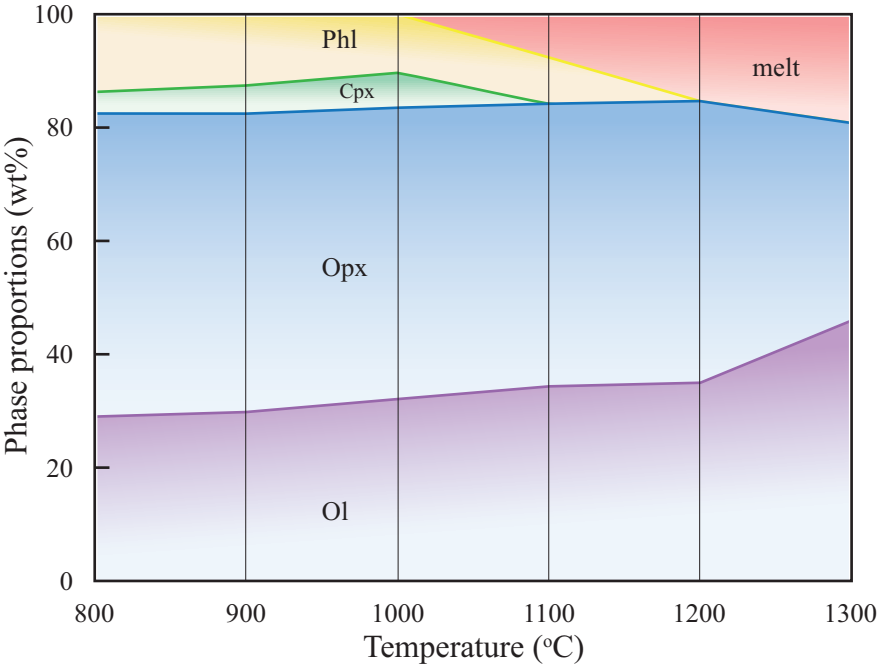


Figure 6

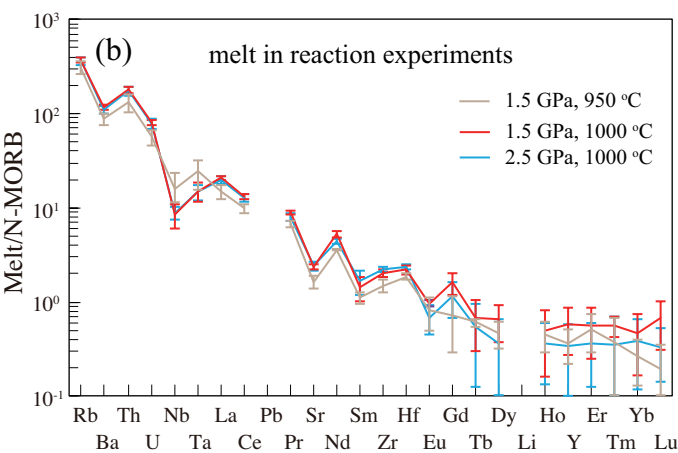
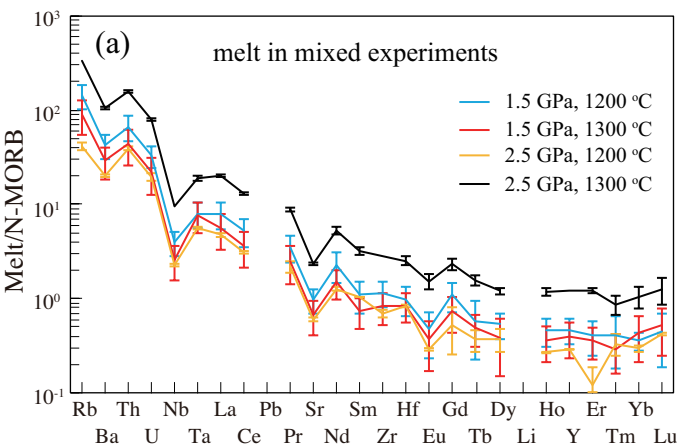


Figure 7

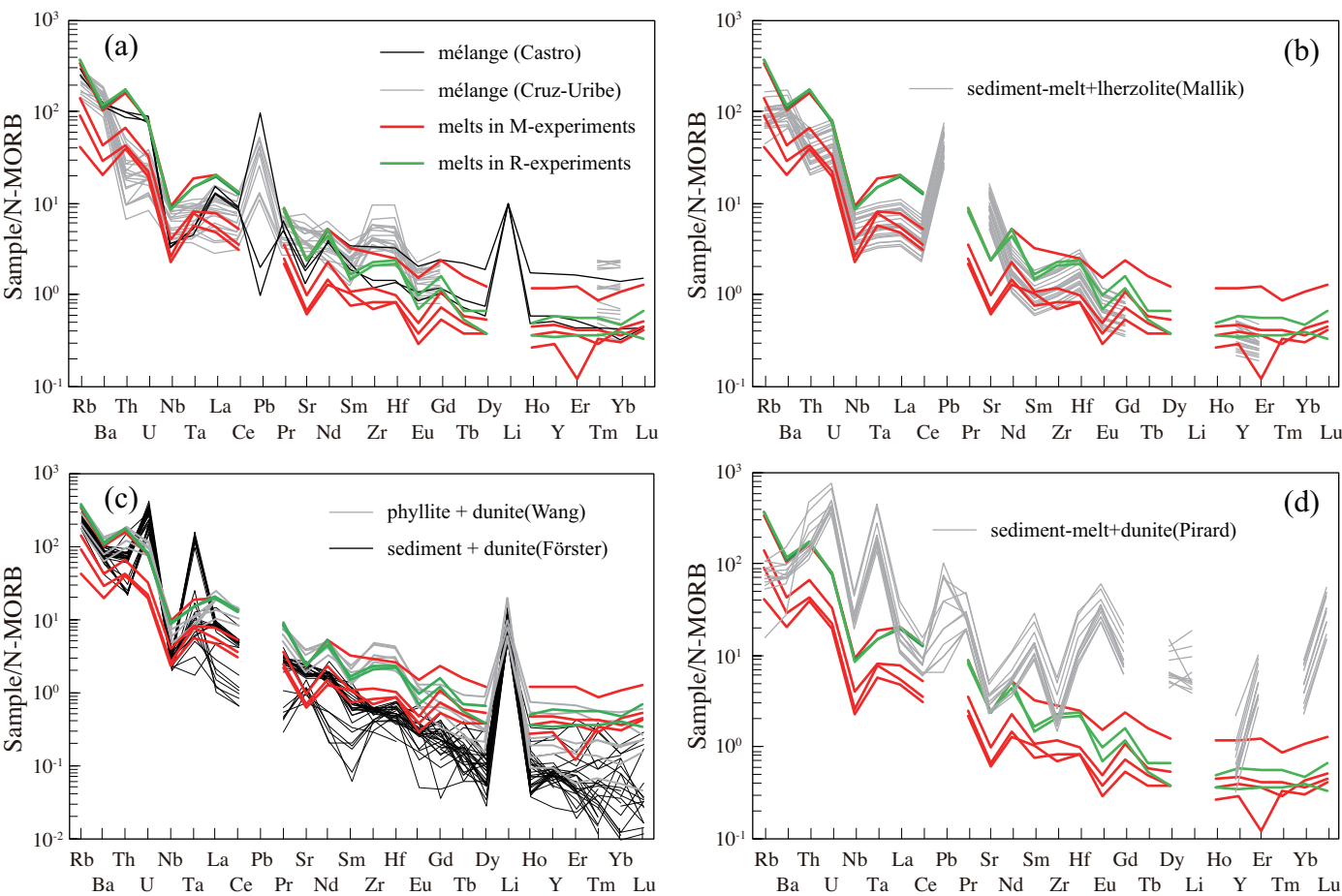


Figure 8

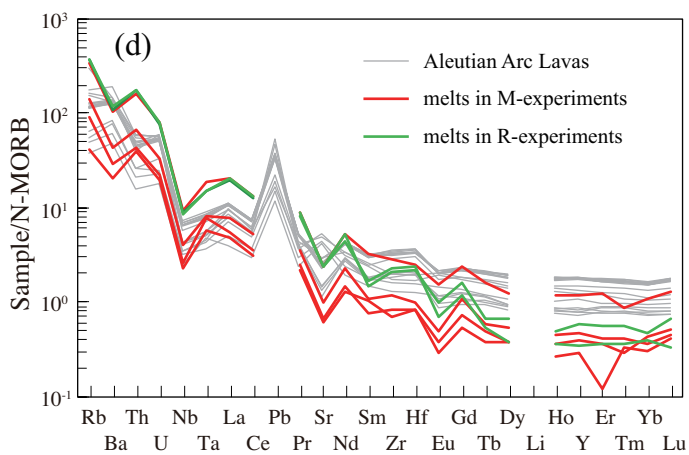
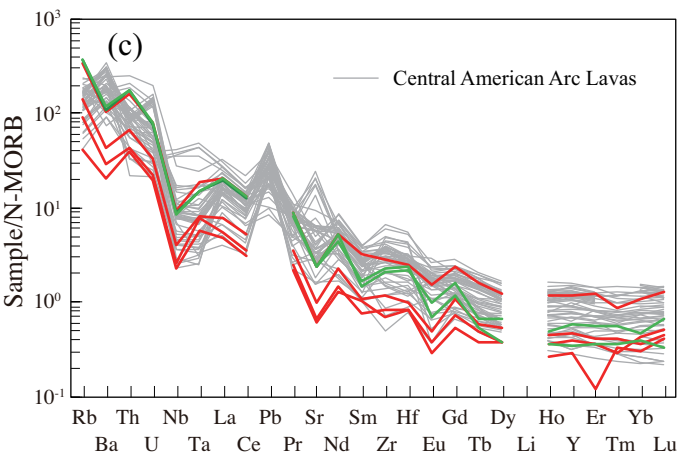
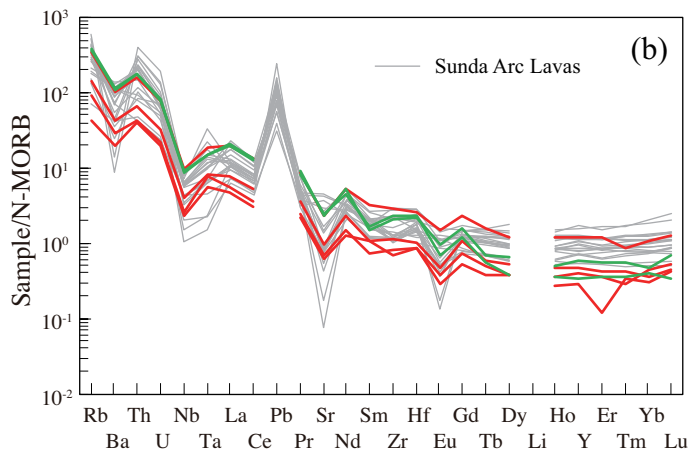
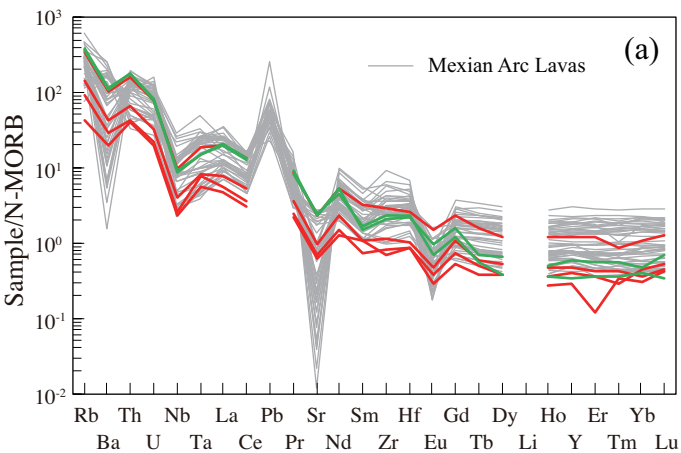


Figure 9