

1 **Mafic inputs into the rhyolitic magmatic system of the**

2 **2.08 Ma Huckleberry Ridge eruption, Yellowstone**

3 Revision 1

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ABSTRACT

25 The silicic (broadly dacitic to rhyolitic) magmatic systems that feed supereruptions show
26 great diversity, but have in common a role for mafic (broadly basaltic to andesitic) magmas
27 as drivers of the systems. Here we document the mafic component in the rhyolitic magmatic
28 system of the 2.08 Ma Huckleberry Ridge Tuff (HRT, Yellowstone), and compare it to mafic
29 materials erupted prior to and following the HRT eruption in the area within and
30 immediately around its associated caldera. The HRT eruption generated initial fall deposits,
31 then three ignimbrite members A, B and C, with further fall deposits locally separating B and
32 C. A 'scoria' component was previously known from the upper B ignimbrite, but we
33 additionally recognise juvenile mafic material as a sparse component in early A, locally
34 abundant in upper A and sparsely in lower B. It has not been found in the C ignimbrite. In
35 upper B the mafic material is vesicular, black to oxidised red-brown scoria, but at other sites
36 is overwhelmingly non-vesicular, and sparsely porphyritic to aphyric. Despite their
37 contrasting appearances and occurrences, the mafic components form a coherent
38 compositional suite from 49.3-63.3 wt % SiO₂, with high alkalis (Na₂O+K₂O = 4.5-7.3 wt %),
39 high P₂O₅ (0.52-1.80 wt %), and notably high concentrations of both high field strength and
40 large-ion lithophile elements (e.g. Zr = 790-1830 ppm; Ba = 2650-3800 ppm). Coupled with
41 the trace-element data, Sr-Nd-Pb isotopic systematics show influences from Archean age
42 lower crust and lithospheric mantle modified by metasomatism during the late Cretaceous
43 to Eocene, as previously proposed for extensive Eocene magmatism/volcanism around the
44 Yellowstone area. The HRT mafic compositions contrast markedly with the Snake River Plain
45 olivine tholeiites erupted before and after the HRT eruption, but are broadly similar in
46 several respects to the generally small-volume Craters of the Moon-type mafic to

47 intermediate lavas erupted recently just west of the HRT caldera, as well as farther west in
48 their type area. The combination of trace element and isotopic data on the HRT mafics are
49 only consistent with an origin for their parental magma as melts from mantle enriched by
50 high temperature and pressure melts, most likely from the underlying Farallon slab.
51 Subsequent interaction of the HRT mafic magmas occurred with the Archean lower crust
52 and lithospheric mantle, but not the highly radiogenic upper crust in this area. The close
53 temporal and spatial relationships of the HRT mafic compositions and the preceding Snake
54 River Plain olivine tholeiite eruptives suggest a high degree of spatial heterogeneity in the
55 mantle beneath the Yellowstone area during the early (and subsequent) development of its
56 modern magmatic system.

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58 **Keywords:** Yellowstone, Huckleberry Ridge Tuff, Craters of the Moon, mafic magmas,
59 magma genesis, mantle metasomatism

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INTRODUCTION

62 Mafic magmas (in this context basaltic to andesitic in composition) are widely
63 considered to exert a fundamental control on the generation and development of large-
64 scale silicic (dacitic to rhyolitic) magmatic systems in the crust (e.g. Hildreth 1981;
65 Bachmann and Bergantz 2008). Over the long-term, mafic magmas provide heat and mass
66 that drive the development of the silicic system (e.g. Bindeman et al. 2008; Christiansen and
67 McCurry 2008). Over short timescales, inferences on the effects of mafic magma are
68 generally focused around the associated influxes of heat and/or volatiles into the silicic
69 system that may mobilize it and trigger eruptions (e.g. Sparks et al. 1977; Bachmann et al.

70 2002; Huber et al. 2011). As silicic magmatic systems act as density traps, evidence of the
71 direct influence of mafic magmas on silicic systems is often limited to one or more of co-
72 erupted mafic enclaves or mingled magmas, and up-temperature geochemical signals in the
73 growth records of crystals in the eruption products (e.g. Sparks et al. 1977; Bacon and Metz
74 1984; Bachmann et al. 2002; Wilson et al. 2006; Pritchard et al. 2013; Barker et al. 2016;
75 Singer et al. 2016; Stelten et al. 2017). The most primitive compositions are, however, often
76 not represented in the co-erupted mafic components when compared with mafic magmas
77 erupted away from the focus of silicic volcanism suggesting that some hybridization has
78 taken place (e.g. Bacon and Metz 1984). Stalling of mafic magmas beneath a silicic system
79 due to density trapping may serve to enhance the interaction of mafic magmas with host
80 rocks and/or silicic magmas and the generation of distinct compositions not seen in adjacent
81 areas (e.g. Wilson et al. 2006). Analysis of mafic inclusions, therefore, can give valuable
82 insights into the thermal and chemical driving mechanisms beneath silicic systems. To
83 illuminate these mechanisms, we here present data on the mafic compositions of inclusions
84 and surficial lavas associated with the Yellowstone Plateau volcanic field, specifically those
85 associated with the earliest caldera-forming cycle that generated the Huckleberry Ridge Tuff.

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GEOLOGICAL SETTING

88 Yellowstone and the Snake River Plain

89 The Yellowstone Plateau volcanic field (YPVF) is the youngest (active since ~2.1 Ma)
90 volcano-magmatic system at the northeastern end of the Yellowstone-Snake River Plain
91 (YSRP) volcanic area (Christiansen 2001). The YSRP area is a 700 km long, NE-ward
92 progressing volcanic province extending from eastern Oregon and northern Nevada to

93 northwestern Wyoming (Fig. 1; Pierce and Morgan 2009). The province contains a series of
94 caldera complexes associated with voluminous silicic volcanism, followed by voluminous
95 basaltic activity. The caldera systems were initiated at ~16 Ma in northernmost Nevada,
96 broadly coincident with the eruption of the Columbia River Basalts to the north (e.g. Coble
97 and Mahood 2012, 2015) and have migrated spasmodically eastward. Although the focus of
98 silicic volcanism has migrated to the northeast (at a rate and in a direction corresponding to
99 movement of the North American Plate over a fixed point), voluminous basaltic volcanism
100 has persisted along the YSRP into Holocene time (Armstrong et al. 1975; Kuntz et al. 1992).

101 The northeastward propagating volcanism of the YSRP has been often been
102 attributed to the movement of the North American plate over a stationary mantle plume,
103 forming a 'hotspot track' (Pierce and Morgan 1992, 2009). This hypothesis is supported by
104 the imaging of a weak thermal anomaly, inferred to represent material containing small
105 degrees of partial melt, down to and across the 660 km discontinuity (Yuan and Dueker
106 2005; Smith et al. 2009; Schmandt et al. 2012). However, a high-velocity zone located in the
107 mantle beneath the Snake River Plain and Yellowstone at ~400-500 km depth has been
108 interpreted as a remnant of the subducted Farallon plate, that foundered beneath the
109 western US at ~50 Ma (Schmandt and Humphreys 2011; James et al. 2011). The presence of
110 this high-velocity zone has led to an alternative model for YSRP volcanism invoking poloidal
111 asthenospheric upwelling around the foundering slab. It has been proposed that the
112 northern and eastern edges of the slab serve to delineate the margins of the YSRP, creating
113 a low-velocity zone observed in the upper mantle along the length of the YSRP (James et al.
114 2011; Zhou et al. 2017). This low-velocity zone is inferred to contain partial melt and to be
115 hydrated, explaining the continuing basaltic volcanism along the SRP even after the
116 termination of local silicic volcanism (Armstrong et al. 1975; Schmandt and Humphreys 2010;

117 James et al. 2011). However, despite the presence of the low-velocity zone along the length
118 of the YSRP, elevated heat and mantle-derived-gas fluxes in the Yellowstone area (e.g.
119 Hurwitz and Lowenstern 2014) require there to be a deep-seated thermal anomaly distinct
120 from that extending westwards into the Snake River Plain.

121 Volcanism along the YSRP locus has typically been considered as bimodal. Olivine
122 tholeiites dominate the mafic compositions and are inferred to provide heat to partially
123 melt solidified, underplated tholeiitic intrusive forerunners which, in turn, become parental
124 magmas to the voluminous ferroan rhyolites (Christiansen and McCurry 2008; McCurry and
125 Rodgers 2009). Basaltic activity typically precedes and follows silicic volcanism at individual
126 volcanic centers along the YSRP, with intervening periods of no basaltic volcanism while the
127 silicic magmatic systems act as an effective density trap (Christiansen 2001). Petrological
128 and isotopic studies collectively infer that the olivine tholeiites result from the hybridization
129 of young, asthenosphere-derived melts with partial melts of the lithospheric mantle, which
130 last equilibrated in an ancient, dry lithospheric mantle keel at depths of 70-100 km and at
131 temperatures consistent with an only slightly elevated geothermal gradient (Doe et al. 1982;
132 Hildreth et al. 1991; Hanan et al. 2008; Leeman et al. 2009). The basalts are also considered
133 to show signatures of minimal crustal contamination on their rise to the surface (Doe et al.
134 1982; Menzies et al. 1984; Hildreth et al. 1991; Leeman et al. 2009). The tholeiites are
135 dominantly olivine and plagioclase phyric with a notable lack of clinopyroxene, indicating
136 minimal significant mid-crustal crystallization, where clinopyroxene would be the primary
137 liquidus phase (Thompson 1975; Leeman et al. 2009). Olivine tholeiites erupted in the
138 Yellowstone area are similar to those elsewhere in the Snake River Plain but extend to more
139 evolved compositions (Christiansen and McCurry 2008). In addition to the olivine tholeiites,
140 some mafic to minor silicic lavas of contrasting affinity are also found in the eastern Snake

141 River Plain. The mafic lavas, predominantly found at the Craters of the Moon lava field, form
142 a distinctly alkalic trend characterized by high TiO_2 , P_2O_5 , Ba, Zr and rare-earth elements
143 (REE), and lower MgO and CaO relative to olivine tholeiites and are referred to as the
144 Craters of the Moon (COM) trend (Leeman et al. 1976; Christiansen and McCurry 2008;
145 McCurry et al. 2008; Putirka et al. 2009).

146 Whether the silicic magma erupted in the YSRP province is generated through
147 fractional crystallization (e.g. McCurry et al. 2008) or through partial melting of basaltic
148 and/or felsic crust (e.g. Hildreth et al. 1991; Bindeman and Simakin 2014), a large amount of
149 basaltic magma is required to provide heat, volatiles and fractionated material. The $>3,700$
150 km^3 of silicic magma erupted in the YPVF alone over the last ~ 2 Ma requires a voluminous
151 supply of basaltic melt from the mantle to drive the silicic volcanism (Christiansen 2001;
152 McCurry and Rodgers 2009; Stelten et al. 2017). This basaltic flux has also been inferred
153 from the modern large thermal and He and CO_2 fluxes within the volcanic field (Hurwitz and
154 Lowenstern 2014) and is linked to the imaging of a large ($46,000 \text{ km}^3$) lower-crustal, melt-
155 bearing body inferred to be basaltic in composition (Huang et al. 2015).

156 Although there is voluminous basaltic volcanism preceding and subsequent to the
157 focus of silicic volcanism at Yellowstone, and its presence has been invoked as a key driver
158 in the generation and triggering of rhyolitic bodies (e.g. Loewen and Bindeman 2015), there
159 is a paucity of basaltic material erupted penecontemporaneously with the voluminous silicic
160 eruptions (Christiansen 2001). Rarely in the volcanic field, and mostly in extra-caldera
161 rhyolites, is evidence for magma mingling observed (Wilcox 1944; Pritchard et al. 2013).
162 Therefore, previous inferences on the mantle inputs into Yellowstone have been solely
163 derived from basalts erupted peripherally to the field or long after caldera formation (Doe
164 et al. 1982; Hildreth et al. 1991; Hanan et al. 2008). To provide a contrasting perspective, we

165 here document the mafic components (including the 'scoria' previously described by
166 Christiansen 2001) that were discharged during the large caldera-forming rhyolitic eruption
167 of the Huckleberry Ridge Tuff. We compare their compositions to mafic (basaltic to andesitic)
168 compositions erupted before and afterwards in the geographic area within and around the
169 Huckleberry Ridge Tuff caldera to investigate the nature of the mafic lineages in this area
170 and consider their likely origins.

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172 **The Huckleberry Ridge Tuff**

173 The ~ 2.08 Ma, $\sim 2,500$ km³ Huckleberry Ridge Tuff (HRT) is the product of the
174 climactic eruption of the first of three volcanic cycles in the YPVF (Christiansen 2001; Rivera
175 et al. 2014; Singer et al. 2014; Wotzlaw et al. 2015). The HRT consists of initial fall deposits,
176 which are overlain by three ignimbrite packages (members A, B and C), with additional fall
177 deposits beneath member C (Christiansen 2001).

178 The HRT eruption was shortly preceded by two episodes of volcanism, generating
179 the rhyolitic Snake River Butte lava, and the lavas collectively mapped by Christiansen (2001)
180 as the Junction Butte Basalt. Other precursory eruptions may well have occurred in areas
181 now down-dropped and buried in the HRT and younger calderas, but any direct evidence for
182 these is now lost. The Junction Butte Basalt is composed of multiple independent flows
183 which crop out at the northern end of the Yellowstone Plateau and northeast of the
184 mapped HRT caldera (Fig. 1; Christiansen 2001). Although the Junction Butte Basalt flows
185 are amongst the least primitive mafic compositions erupted at Yellowstone (and Junction
186 Butte itself is intermediate in composition), they are here grouped in with the SRP olivine
187 tholeiites, albeit with these flows containing plagioclase phenocrysts and groundmass
188 olivine (Hildreth et al. 1991; Christiansen 2001).

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SAMPLED MATERIALS

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We present data here on eruptive materials from the Yellowstone area and for descriptive convenience group these into three suites (Table 1). These suites encompass juvenile mafic material erupted in the Huckleberry Ridge event, together with comparator lavas which are temporally close to the eruption and/or were erupted spatially adjacent to or within the HRT caldera. Sample and locality information are given in Supplementary Table 1.

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Suite 1: HRT mafic materials. Although the HRT is composed dominantly of high-silica rhyolite, scoriaceous material has been reported as commonly present in the upper part of member B (Hildreth et al. 1991; Christiansen 2001). Reported analyses of this scoriaceous material (samples 81YH-79: Hildreth et al. 1984; 74IP-149B: Hildreth et al. 1991; Christiansen 2001), however, return rhyodacite compositions (70.5 and 70.8 wt % SiO₂ respectively). Extensive fieldwork on the HRT by Wilson shows, however, that there is a comparable juvenile mafic component in both ignimbrite members A and B (Table 1). This component matches physically the scoria described by Christiansen (2001) where it is found in upper ignimbrite B. However, our analyses yield greatly contrasting compositional characteristics when compared to the previously reported data. In member A, especially at exposures southwest of the caldera, poorly to non-vesicular clasts of dark-grey to greenish-grey, rarely red-oxidized material (Fig. 2a) occur within the dense-welded rheomorphic tuff attributed by Christiansen (2001) to member B, but identified by us as member A. Briefly, this identification is made on the basis that the relevant tuff can be traced continuously downwards to the HRT basal contact, and upwards to where a horizon representing a short

212 hiatus (reworked ash materials, minor welding reversal) separates the lower ignimbrite (i.e.
213 A) from an upper ignimbrite unit that has scoria present in its upper parts (i.e. B). Non-
214 vesicular mafic material is also rarely found in lower ignimbrite B in the same area
215 southwest of the caldera. Moderately to highly vesicular black to purple- to red-oxidised
216 scoria is also widespread as a component in mingled pumices (Fig. 2b) and as discrete lapilli-
217 grade clasts in upper member B, as previously reported. We sampled individual clasts of the
218 poorly vesicular material from ignimbrite A mostly from localities southwest of the caldera,
219 and extracted the vesicular scoria from mingled pumices in upper ignimbrite B from a site
220 southwest of the caldera where glassy, non-welded material occurs (Supplementary Table 1).
221 Elsewhere, the discrete scoria clasts in upper ignimbrite B are either too small to sample
222 effectively, or are vapor-phase altered and recrystallized along with their host tuff. Also,
223 rare macroscopically mingled scoria clasts were found in member A and sampled, and are
224 labelled as such here.

225 **Suite 2: SRP olivine tholeiites.** We sampled a selection of the lavas generally
226 attributed to the SRP olivine tholeiite suite (Supplementary Table 1), including several
227 examples reported on by Hildreth et al. (1991). These lavas represent flows (i) pre-dating
228 the HRT and exposed around the caldera margin (including one example underlying the HRT
229 southwest of the caldera), and (ii) post-dating the HRT, forming part of the extensive flows
230 flooring the Island Park segment of the combined HRT and Mesa Falls Tuff-related calderas,
231 and elsewhere in the Yellowstone caldera.

232 **Suite 3: COM eruptives.** We sampled pyroclasts from several scoria cones linked to
233 young eruptions of the COM suite in the Spencer-High Point Field (Fig. 1; Supplementary
234 Table 1). These cones were sampled from an area on, or immediately west of, the western

235 rim of the mapped HRT caldera and were selected on the basis of information in Iwahashi
236 (2010).

237 **Published data.** We compare our data from all three suites to published data from
238 spatially and chemically comparable deposits. The Suite 2 literature field comprises data
239 from olivine tholeiites along the Yellowstone-Snake River Plain volcanic area (but does not
240 include data from the Columbia River Basalt group). This field also includes examples of
241 olivine tholeiites from the Craters of the Moon lava field (NEOT flows of Putirka et al. 2009),
242 from the Spencer-High Point Field (Type 1 flows of Iwahashi 2010) and compositions from
243 two databases: the North American Volcanic Rock Data Base (NAVDAT:
244 <http://www.navdat.org/>) and the Geochemistry of Rocks of the Oceans and Continents
245 (GEOROC: <http://georoc.mpch-mainz.gwdg.de/georoc/>). Published data for Suite 3
246 comparisons were derived from studies of the Craters of the Moon area (Leeman 1974;
247 Leeman et al. 1976; COME flows of Putirka et al. 2009) and from the Spencer-High Point
248 Field (Type 2 flows of Iwahashi 2010). All mafic data (i.e. <65 wt% SiO₂, the range of
249 compositions analyzed in this study) were selected based on location, as we did not want to
250 restrict comparisons to basalts only (cf. Hildreth et al. 1991). No screening for perceived
251 contamination or primitive compositions was undertaken so as to allow us to compare
252 compositional ranges and degrees of evolution in all suites related to the broad range of
253 petrogenetic processes occurring in volcanic rocks in this region.

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ANALYTICAL METHODS

256 Samples from the three suites were collected and any adhering matrix or visible
257 xenocrysts removed by hand picking to isolate the mafic component. In two clasts (YP185

258 and YP188, both from early member A) mingling was too complex to allow complete
259 separation. These clasts therefore represent a mixture of mafic and rhyolitic components
260 (denoted by 'M' in geochemical plots). Samples were subsequently crushed and milled in an
261 agate Tema mill to yield a homogenous powder. Powders were analyzed for major element
262 concentrations by X-ray fluorescence (XRF) at the Open University (OU), UK and at the
263 University of Auckland, New Zealand following the methods of Ramsey et al. (1995).
264 Replicate analyses of standards give approximate 2 standard deviation (2sd) external
265 precisions of <3% for all elements, with most <1% (Electronic Appendix 1). Accuracies are
266 within 5% for all elements compared to preferred values for replicate standard analyses.
267 Duplicates run between institutions show <5% offset for all elements with >0.1 wt%
268 abundance. Trace element concentrations were measured by solution inductively-coupled
269 plasma mass-spectrometry (ICP-MS) at the OU and Victoria University of Wellington, New
270 Zealand (VUW) using an Agilent 7500 and a Thermo Scientific Element2 sector-field ICP-MS,
271 respectively. Abundances of trace elements were calculated by external normalization
272 relative to a bracketing standard (BHVO-2). A secondary standard (BCR-2) was used to
273 estimate external precisions, which were <6-7% for most elements except Li, Nb, Cs, Lu, Ta,
274 Tl, Pb, Th, U, Ni, Cu, Zn (<20%) and Mo (>20%). Accuracies are <6-7% for all elements, apart
275 from Ta, Tl, Cu (<15%) and Mo (>15%).

276 Isotope analyses were conducted using a Finnigan Triton Thermal Ionization Mass-
277 Spectrometer (TIMS) in the laboratories at the OU and VUW, but on the same instrument.
278 Chromatographic element separation was based on the procedure of Pin et al. (2014; see
279 Supplementary Material for more information). Sr was analyzed on single Re filaments,
280 using a TaF₂ activator (Charlier et al. 2006), for 240 ratios with an integration time for each
281 ratio of 16.777 s. Measurements were internally normalized to $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$ and Rb

282 interference was corrected by measuring ^{85}Rb and applying a correction using $^{87}\text{Rb}/^{85}\text{Rb} =$
283 0.385707 (Rosman and Taylor 1998). Repeated analyses of NBS987 yield an average of
284 0.710254 ± 0.000015 (2sd, n=16). Isotopic ratios from different runs were normalized to a
285 value of 0.71025 for NBS987, the long-term mean reported by Thirlwall (1991). Procedural
286 blank was 110 pg, insignificant when compared to the ~ 1000 ng loaded onto each filament
287 and thus necessitated no blank correction.

288 Nd was analyzed on double Re filaments with a H_3PO_4 loading solution over 270
289 ratios with an integration time of 8.389 s for each measurement. Ratios were internally
290 normalized using a value of $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$ and any presence of Ce and Sm corrected
291 using values of $^{140}\text{Ce}/^{142}\text{Ce} = 7.97297$ and $^{144}\text{Sm}/^{147}\text{Sm} = 0.20667$ (Rosman and Taylor 1998).
292 Repeated analyses of standards J&M (internal) and La Jolla yield values of $0.511818 \pm$
293 0.000004 (2sd, n=11) and 0.511845 ± 0.000002 (2sd, n=3) respectively, the former
294 consistent with the long-term laboratory average (0.511821 ± 0.000002 2sd) and the latter
295 similar to 0.511856 ± 0.000007 (2sd) measured by Thirlwall (1991). Procedural blank was 8
296 pg and thus warranted no blank correction.

297 Pb was analyzed using a double-spike method (see Todt et al. 1996). Half the sample
298 (approximately 150 ng), was run naturally on single Re filaments for 180 ratios using a silica
299 gel activator (Gerstenberger and Haase 1996). Subsequently, the remaining sample and a
300 $^{207}\text{Pb}/^{204}\text{Pb}$ double spike (Thirlwall et al. 2000) was thoroughly admixed mixed and analyzed
301 on single Re filaments for 120 ratios in the same way as the natural run. Integration times
302 for both runs were 8.389 s. The data were then deconvolved to determine the isotopic
303 ratios. Values for NBS981 were $^{206}\text{Pb}/^{204}\text{Pb} = 16.945 \pm 0.001$, $^{207}\text{Pb}/^{204}\text{Pb} = 15.503 \pm 0.001$
304 and $^{208}\text{Pb}/^{204}\text{Pb} = 36.737 \pm 0.004$ (2sd, n = 32). Isotopic measurements from different runs

305 were normalized to the values of Todt et al. (1996). The procedural blank for Pb was 10 pg
306 and warranted no correction.

307

308 RESULTS

309 Full major element, trace element and isotopic data for the samples analyzed in this
310 study are given in Electronic Appendix 1, and a representative selection of analyses is given
311 in Tables 2 and 3.

312

313 Major elements

314 HRT mafic dense and scoria clasts (hereafter HRT mafics, constituting Suite 1) form a
315 coherent trend from 49-63 wt% SiO₂, regardless of their stratigraphic position or degree of
316 vesiculation. They are characterized by high alkalis (Na₂O + K₂O = 4.5-7.3 wt%; Fig. 3a), P₂O₅
317 (0.52-1.8 wt%; Fig. 3b), TiO₂ (1.5-3.5 wt%) and FeO (9.6-16.1 wt%; Supplementary Fig. 1).
318 Suite 1 samples also have low MgO (0.6-3.0 wt%) and CaO (1.6-7.2 wt%; Supplementary Fig.
319 1) values.

320 Samples from flows with olivine tholeiitic affinity (Suite 2) erupted throughout the
321 volcanic history of Yellowstone form a coherent compositional group with SiO₂ ranging from
322 46-54 wt%. They are distinct from Suite 1 samples with higher MgO (4.2-11 wt%) and CaO
323 (7.7-11 wt%; Supp. Fig. 1), which are negatively correlated with SiO₂, and contain low total
324 alkalis (Na₂O+K₂O; 2.4-4.7 wt%; Fig. 3a) and P₂O₅ (0.19-0.62 wt%; Fig 3b). Mg# values
325 (defined as $Mg\# = 100[X_{MgO}/(X_{MgO}+X_{FeO})]$ where $FeO = 0.9[Fe_2O_3 (T)]$ on a wt% basis) of up to
326 64 show that the primitive end of the range in YSRP tholeiites is represented in our
327 Yellowstone Suite 2 samples (cf. Leeman et al. 2009).

328 There are broad major element similarities between our COM-type samples (Suite 3)
329 and HRT mafics (Suite 1). Suite 3 samples cover a similar SiO₂ range to Suite 1 samples (47-
330 57 wt%), with similarly high FeO (10-14 wt%), TiO₂ (up to 3.0 wt%; Supplementary Fig. 1)
331 and P₂O₅ (0.60-2.2 wt %; Fig. 3b), and depletions in MgO (1.6-3.7 wt%) and CaO (4.7-8.3 wt%)
332 when compared to the olivine tholeiites. Their compositions are mildly alkalic (total alkalis
333 5.0-7.6 wt%; Fig. 3a) and are similar to the published COM-trend for rocks with this silica
334 percentage (Fig. 3; Leeman et al. 1976; Christiansen and McCurry 2008; Putirka et al. 2009).
335 Note, however, that in general, the Suite 1 HRT mafic compositions form arrays in Harker
336 plots that are parallel to, but slightly offset from, our Suite 3 samples and the overall COM
337 fields (see Fig. 3), particularly with respect to lower total alkalis, P₂O₅, and TiO₂ in the Suite 1
338 samples.

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340 **Trace elements**

341 The clear separation between the Suite 1 HRT mafic samples and Suite 3 COM-type
342 samples when compared to the Suite 2 olivine tholeiites is also apparent in trace element
343 abundances. The Suite 1 HRT mafics and Suite 3 COM-type materials have notably high
344 concentrations of incompatible elements including large-ion lithophile elements (LILE), e.g.
345 Rb (25-99 ppm; Supplementary Fig. 2a) and Ba (1120-3800 ppm: Fig. 4a), high field strength
346 elements (HFSE), e.g. Zr (660-1970 ppm: Fig. 4b) and Nb (29-87 ppm), and actinides e.g. U
347 (1.2-3.5 ppm with outliers at 4.7 and 5.7 ppm). In contrast, olivine tholeiite samples have
348 notably higher V (210-280 ppm; Supplementary Fig. 2c), Ni (26-190 ppm) and Cr (20-540
349 ppm). Sr concentrations are similar among both the mafic suites 1 and 3 (140-550 ppm;
350 Supplementary Fig. 2b) and within the range defined by our Suite 2 olivine tholeiite samples.
351 Suite 1 HRT mafic and Suite 3 COM-type samples are elevated in all rare-earth elements

352 (REE) relative to the Suite 2 olivine tholeiites and have sub-parallel trends (Fig. 5) with
353 (La/Yb)_N ratios (5.0-10 with outlier YP242 at 18) overlapping with those of the olivine
354 tholeiites (2.6-11). Zr/Hf ratios are elevated in the Suite 1 (47-61) and Suite 2 samples (52-
355 55) relative to the Suite 3 samples (40-49) but other incompatible element ratios are similar
356 between the different suites (e.g. Ce/Pb: Supplementary Fig. 3).

357 The only major divergence between the trace element compositions of the Suite 1
358 HRT mafic samples and Suite 3 COM-samples is in Ba where there are two apparent trends
359 when plotted versus SiO₂ (Fig. 4a). The moderate Ba trend in our Suite 3 samples aligns with
360 the typical COM-trend reported elsewhere (Leeman et al. 1976; Christiansen and McCurry
361 2008) whereas the Suite 1 HRT mafic samples define a parallel trend with roughly double
362 the Ba concentrations at a given value of SiO₂ (Fig. 4a).

363

364 **Isotopic ratios**

365 ⁸⁷Sr/⁸⁶Sr values for all samples range from 0.70373-0.70808, with the highest and
366 lowest values measured from Suite 2 olivine tholeiite samples, similar to the range of
367 0.70377-0.70886 reported by Hildreth et al. (1991) from Yellowstone basalts. Suite 3
368 samples are tightly clustered (0.70574-0.70585) with one outlier (YR425: 0.70788 ± 0.00004
369 2se; Fig. 6). Suite 1 HRT dense mafics and scoria have broadly similar ⁸⁷Sr/⁸⁶Sr values
370 (0.70709-0.70771), which are more radiogenic than our Suite 3 COM-type samples (Fig. 6, 7).
371 However, both suites are notably less radiogenic than the COM samples reported on by
372 Putirka et al. (2009: ⁸⁷Sr/⁸⁶Sr = 0.70784-0.71130) and Leeman (1974: ⁸⁷Sr/⁸⁶Sr = 0.70810-
373 0.71240). ⁸⁷Sr/⁸⁶Sr variations in the Suite 1 HRT mafic samples correlate positively with
374 respect to whole-rock Rb/Sr ratios (R² = 0.65) whereas the olivine tholeiites show isotopic

375 variability at similar Rb/Sr ratios (Fig. 7a). There is no clear correlation between $^{87}\text{Sr}/^{86}\text{Sr}$ and
376 $1/\text{Sr}$ in any of our samples (Fig. 7b).

377 In similar fashion to Sr, the full range in $^{143}\text{Nd}/^{144}\text{Nd}$ values (0.51207-0.51251) for our
378 data is spanned by Suite 2 olivine tholeiite samples (Fig. 6). HRT dense mafic clasts from
379 ignimbrite A (0.51218-0.51232), HRT vesicular scoria from ignimbrite B (0.51231-0.51235)
380 and Suite 3 samples (0.51225-0.51240) all cover similar, largely overlapping ranges. All
381 samples give negative ϵ_{Nd} values (-2 to -10), where

$$\epsilon_{\text{Nd}} = \left[\frac{\left(^{143}\text{Nd}/^{144}\text{Nd} \right)_{\text{meas}}}{\left(^{143}\text{Nd}/^{144}\text{Nd} \right)_{\text{CHUR}}} - 1 \right] \times 10^4$$

382 from DePaolo and Wasserburg (1976). A value of CHUR (chondritic uniform reservoir) of
383 0.51263 was used, from Bouvier et al. (2008). Three of four Suite 3 samples have Nd isotopic
384 values very similar to the median of Yellowstone-Snake River Plain basalts (0.512405:
385 McCurry and Rodgers 2009), whereas three of four Suite 2 samples have ratios more
386 radiogenic than the median and the Suite 1 samples are less radiogenic. However, all except
387 three samples (one from each suite), have ϵ_{Nd} values >-7 , below which value basalts are
388 interpreted by McCurry and Rodgers (2009) to be contaminated. There is also no clear
389 correlation in any of the suites between $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ (Fig. 6).

390 Pb isotopic compositions also follow Sr and Nd in showing the greatest range in the
391 Suite 2 olivine tholeiite samples ($^{206}\text{Pb}/^{204}\text{Pb} = 15.86\text{-}17.64$; $^{207}\text{Pb}/^{204}\text{Pb} = 15.27\text{-}15.53$;
392 $^{208}\text{Pb}/^{204}\text{Pb} = 36.47\text{-}38.23$; Fig. 8). In contrast in Suite 1, HRT dense mafics ($^{206}\text{Pb}/^{204}\text{Pb} =$
393 $16.88\text{-}16.95$; $^{207}\text{Pb}/^{204}\text{Pb} = 15.45\text{-}15.46$; $^{208}\text{Pb}/^{204}\text{Pb} = 37.92\text{-}37.94$) and HRT vesicular scoria
394 ($^{206}\text{Pb}/^{204}\text{Pb} = 16.95\text{-}16.98$; $^{207}\text{Pb}/^{204}\text{Pb} = 15.48$; $^{208}\text{Pb}/^{204}\text{Pb} = 38.02\text{-}38.03$) show much more
395 restricted ranges with the scoria being more radiogenic. Our Suite 3 COM-type samples
396 ($^{206}\text{Pb}/^{204}\text{Pb} = 17.16\text{-}17.36$; $^{207}\text{Pb}/^{204}\text{Pb} = 15.48\text{-}15.51$; $^{208}\text{Pb}/^{204}\text{Pb} = 38.07\text{-}38.15$) also show a

397 modest range. Suite 1 samples collectively form a sub-parallel trend to the Suite 2 olivine
398 tholeiite samples, offset to higher $^{207}\text{Pb}/^{204}\text{Pb}$ for a given value of $^{206}\text{Pb}/^{204}\text{Pb}$ (Fig. 8). All
399 except one sample (the $^{207}\text{Pb}/^{204}\text{Pb}$ composition of Basalt of the Narrows [YR292], which
400 also has notably unradiogenic Sr isotopic characteristics) plot above the Northern
401 Hemisphere Reference Line (Hart 1984).

402

403

DISCUSSION

404 **The context of the HRT mafic magmas**

405 In considering the nature and range of compositions of the Suite 1 HRT mafic
406 compositions and the comparator materials from suites 2 and 3 there are two aspects that
407 need to be borne in mind. First, as seen elsewhere (e.g. Bacon and Metz 1984; Hildreth et al.
408 1991; Wilson et al. 2006; Pritchard et al. 2013), these mafic compositions related to silicic
409 systems show features in their trace element and isotopic characteristics that suggest they
410 do not generally represent pristine melts or magmas coming directly from a uniform mantle
411 source. Even the least evolved compositions sampled in each of the case studies cited above
412 show evidence for some variability in the mantle source, and/or fractionation or
413 assimilation, but when and where these processes occurred is open to debate, as is the role
414 of mantle versus crustal processes (cf. Rasoazanamparany et al. 2015). Second, the mafic
415 compositions sampled in single silicic eruptions are rarely uniform in composition (see
416 examples cited above), and the processes whereby the compositional variations within each
417 suite are generated also need to be considered.

418 In addition, there needs to be taken into consideration the regional setting of the
419 HRT mafics we document. Although lavas of olivine tholeiite affinity were erupted before

420 and after the HRT in the Yellowstone area, major and trace element characteristics of the
421 Suite 1 HRT mafics show closer links with COM-type compositions represented here by Suite
422 3 samples from the late Pleistocene Spencer-High Point volcanic field. The latter eruptives
423 have been in turn been linked with distinctly alkalic lava flows from the Craters of the Moon
424 lava field (Kuntz et al. 1992; Iwahashi 2010). The Craters of the Moon lava field consists of
425 ~30 km³ of late Quaternary (~15-2.1 ka) lava flows, erupted from monogenetic and
426 polygenetic vents along a volcanic rift zone, the 85 km long Great Rift in the eastern Snake
427 River Plain (Fig. 1: Kuntz et al. 1986). During the total lifespan of the Craters of the Moon
428 lava field, several other olivine tholeiite lava fields were erupted in the eastern Snake River
429 Plain, two of which (Kings Bowl and Wapi lava fields) were also sourced from the Great Rift
430 (Fig. 1) and were contemporaneous with the final eruptive phase of the Craters of the Moon
431 lava field (Kuntz et al. 1986, 1992).

432 The contemporaneity of young COM and olivine tholeiitic lavas along the Great Rift is
433 analogous to what we report here from the HRT, which occurred between episodes of
434 olivine tholeiite extrusion. This complexity within the YPVF has not been recognized before,
435 with the prevailing view being that olivine tholeiites dominate the YPVF mafic suite and are
436 intimately linked to the rhyolites (e.g. Hildreth et al. 1991; Christiansen and McCurry 2008).
437 It is thus apparent that the sharp contrasts in composition between the COM and olivine
438 tholeiite magma types reflect sources and processes that can act contemporaneously to
439 feed vents contained within quite limited geographic areas.

440 In the subsequent discussion, we first consider the various controls on the
441 compositions of the least-evolved magmas erupted within the three suites reported on here,
442 and in particular address whether the Suite 1 HRT mafics could be generated from the same
443 source region/primary magma as gave rise to the volumetrically dominant olivine tholeiites

444 at Yellowstone and elsewhere in the SRP. Evidence for and against each of the processes put
445 forward is summarized in Table 4. We then focus on the variability within each of the suites
446 in order to address whether these variations follow a common pattern, or whether there
447 are unique features to the compositional variations shown within each suite. Finally, we
448 compare and contrast our data and inferences with the published array of compositional
449 information on SRP volcanism, with particular reference to the main Craters of the Moon
450 area.

451

452 **Generation of the parental magma for the Suite 1 HRT mafic compositions**

453 The least evolved of the Suite 1 HRT mafics (Table 2) has a bulk SiO₂ composition
454 close to those of the olivine tholeiites in the Yellowstone area (Suite 2 data, and Hildreth et
455 al. 1991), but radically contrasting values for many major and trace elements (Table 2, Figs.
456 3, 4). We here consider the diverse and sometimes contradictory models (summarized in
457 Table 4) proposed for generation of the parental COM-type magmas (including our Suite 3
458 samples) and compare them against the data we present here for the Suite 1 HRT mafics.

459 **Fractional crystallization.** The most commonly invoked mechanism for generation of
460 magmas in the COM-trend in general has been through extreme fractional crystallization of
461 an olivine tholeiite parent (Christiansen and McCurry 2008; McCurry et al. 2008). In support
462 of this view, experimental studies by Whitaker et al. (2008) derived liquids with major
463 element compositions similar to primitive COM lava flows through ~80 % crystallization
464 (~40 % plagioclase, ~22 % olivine and ~18 % pyroxene) of an olivine tholeiite starting
465 material at 1,100 °C, 4.3 kbar and 0.4 wt% H₂O. However, there was also early recognition
466 that fractional crystallization alone cannot completely replicate the observed compositional
467 variations, particularly with regard to trace elements (Leeman et al. 1976). Our Suite 1 HRT

468 mafics data allow us to consider a fractional crystallization mechanism through simple
469 modeling of trace elements to try and replicate the distinctive signatures of the least
470 evolved members of the suite. Fractional crystallization modeling used the Rayleigh
471 fractionation equation:

$$472 \quad \frac{C_L}{C_O} = F(\bar{D}-1),$$

473 where C_L and C_O are the elemental concentrations in the derived and original liquids
474 respectively, F is the fraction of melt remaining and \bar{D} is the bulk distribution coefficient:

$$475 \quad \bar{D}_a = \sum W_B D_{aB},$$

476 where W_B is the weight % of the mineral B in the rock and D_{aB} is the distribution coefficient
477 of element a in mineral B .

478 Warm River basalt (sample YR291) was used as the starting composition due to its
479 similarity in composition to the starting material of Whitaker et al. (2008) and it having
480 characteristics of a relatively 'primitive' lava (e.g. Mg# of 64.1 [Leeman et al. 2009] with high
481 Ni and Cr: Table 2). Using $\bar{D} = 0$ (i.e. perfect incompatibility) for Ba, Zr, Rb, U, Hf, Th, and Y,
482 $\geq 84\%$ crystallization generates liquids similar in major element composition to the aphyric
483 samples at the least-evolved end of our HRT Suite 1 samples. This degree of crystallization is
484 similar to that proposed by Whitaker et al. (2008) from experimental studies. To test this
485 possible fractionation pathway, we model Sr values on the basis that the experimental and
486 petrographic crystal assemblages are dominated by plagioclase in which Sr is compatible. To
487 generate a composition similar to the HRT suite with $\sim 84\%$ crystallization, $\bar{D} = 0.7$ is
488 required. Using the partitioning relationship of Sr in plagioclase from Blundy and Wood
489 (1991), at a temperature of 1,100 °C (Whitaker et al. 2008; Putirka et al. 2009) and $X_{An} =$
490 0.63 (Whitaker et al. 2008), $D_{Sr} = 2.4$, (similar to $D_{Sr} = 2.31$ at 1194 °C, 5 kbar and $An = 0.59$

491 proposed by Sun et al. 2017). Based on the experimental crystallization of 50% plagioclase,
492 and ignoring the minimal effects of olivine and clinopyroxene on the Sr partition coefficient,
493 this approach would yield a value of $\overline{D}_{Sr} = 1.4$ and generate Sr-depleted melts (105 ppm with
494 84% crystallization) that have lower Sr concentrations than any Suite 1 samples analyzed in
495 this study (Electronic Appendix 1; Fig. 9). Using only the most calcic plagioclase composition
496 ($X_{An} = 0.85$) from Putirka et al. (2009), with the other parameters unchanged, would a
497 plagioclase $D_{Sr} = 1.4$ and therefore a required $\overline{D}_{Sr} = 0.7$ be generated. Although it is possible
498 to replicate the observed patterns with variable Sr partition coefficients, we additionally
499 note that there are no strong negative Eu anomalies in any Suite 1 samples (Fig. 5), which
500 would be expected with significant plagioclase fractionation. A similar degree of
501 fractionation (>80%) would be required to generate the Eu signature of the HRT Suite 1
502 samples with $D = 0$, a very unlikely situation in a plagioclase-dominated assemblage. Using
503 $D_{Eu} = 2.39$ at 1194 °C, 5 kbar and An = 0.59 (Sun et al. 2017), a resulting $\overline{D}_{Eu} = 1.2$ would
504 generate melts depleted in Eu (0.6 ppm with 84% crystallization), the opposite to what is
505 seen (Fig. 5). Therefore we consider extreme fractionation of an olivine tholeiite parent to
506 be not viable as the sole or even necessarily a major mechanism for the generation of the
507 parental Suite 1 HRT compositions.

508 **Crustal assimilation.** Another possibility to generate the least-evolved HRT Suite 1
509 melts is assimilation of crustal rocks by olivine tholeiites as they ascend. Any variations
510 within the HRT suite, e.g. Ba (Fig. 4a), would then be explained through varying degrees of
511 assimilation. In this context, Geist et al. (2002) proposed assimilation of a P-rich ferrogabbro,
512 whereas Putirka et al. (2009) proposed a two-stage assimilation-fractional crystallization
513 (AFC) model. The latter invoked early assimilation of 'mafic pods' in the lower crust followed
514 by mid-crustal assimilation of wall-rocks similar in composition to the rhyolite inclusions

515 reported from the COM suite (but not found in the centers sampled for this work). We use a
516 similar approach to Putirka et al. (2009) here to evaluate possible assimilants to explain the
517 least evolved Suite 1 HRT mafic compositions.

518 To generate the HRT primitive end-member through dominantly assimilation
519 processes requires that the assimilant has elemental concentrations at least as high as that
520 end-member. NAVDAT and GEOROC databases were thus used to search for plausible end
521 members (see Electronic Appendix 2 for search parameters). Although no results fitted the
522 search criteria in NAVDAT, 18 samples were returned from GEOROC, predominantly
523 lamproites. Although these samples have the low-moderate SiO₂ (37-59 wt%) required to be
524 a plausible assimilant, they are generally much higher in Sr, with all except one with Sr
525 concentrations least twice as high (>1110 ppm), and with K₂O values mostly greater than
526 three times the concentration of any Suite 1 sample at a given SiO₂ concentration.
527 Furthermore, the REE trend of the GEOROC sample group is very unlike the sub-parallel
528 trends of the olivine tholeiites and HRT suites, being highly LREE enriched and having
529 (La/Yb)_N ratios (≥52) at least three times those of any samples reported here. Any AFC trend
530 with a lamproitic or similar assimilant would then require a second AFC stage with a strongly
531 heavy-REE enriched assimilant of which a composition has yet to be found. Additionally,
532 although lamprophyres occur on the Colorado Plateau and in Wyoming, they have low Zr
533 and high Sr concentrations (Mirnejad and Bell 2006; Lake and Farmer 2015), contrasting
534 strongly with the Suite 1 mafics.

535 Considering other possible assimilants, rhyolite and granulite xenoliths found in lavas
536 along the SRP have low TiO₂, lower REE abundances and, in most cases, steeper REE slopes
537 than their host lavas (Leeman et al. 1985), antithetic to the elevated TiO₂, parallel REE
538 trends but enriched concentrations of the least-evolved Suite 1 melts compared to Suite 2

539 parental compositions. Another characteristic precluding a strong control of assimilation on
540 magma genesis is the high FeO of the Suite 1 parental compositions. The GEOROC
541 comparator samples, with the exception of one, and COM rhyolite inclusions have lower
542 FeO contents than olivine tholeiites (<7.4 and <3.5 wt% respectively) and thus cannot cause
543 the FeO increase required to generate the Suite 1 parent from an olivine tholeiite.
544 Furthermore, the lower K/Ba, and similar K/Rb with increasing SiO₂ in the Suite 1 samples
545 relative to Suite 2 samples (Supplementary Fig. 4), which ratios would be expected to
546 increase and decrease, respectively, with crustal assimilation, argue against significant
547 contamination (as concluded by Leeman et al. 1976). High K/Ba values occur in HRT rhyolitic
548 samples (Hildreth et al. 1991), assimilation of which would lead to increased K/Ba in the
549 basalts, as is observed particularly in Suite 2 samples. Nevertheless, these HRT analyses
550 have <650 ppm Zr, values that would be insufficient to generate the Suite 1 mafic parent
551 through assimilation alone. There is an apparent trend defined by scoria samples (Fig. 4)
552 with a visibly mingled sample (YP188) at the silicic end. However, this trend is antithetic to
553 that of the overall Suite 1 mafics, indicating that mixing with rhyolite is not a plausible
554 general mechanism for generation of the overall Suite 1 geochemical characteristics.
555 Furthermore, the HRT rhyolites have Zr/Nb ratios of 4-13 (Hildreth et al. 1991). Mixing with
556 rhyolite would require a mafic end-member Zr/Nb ratio of >50, which in turn would require
557 the presence of an implausible and unseen Nb-rich phase (Fig. 9).

558 All samples analyzed in this study have higher Sr and lower Nd isotopic signatures
559 than normal mantle-derived magmas, but which are consistent with values in basalts from
560 the Yellowstone-Snake River Plain area that have equilibrated with lithospheric mantle (Fig.
561 6; Hanan et al. 2008; McCurry and Rodgers 2009). The elevated ⁸⁷Sr/⁸⁶Sr ratios of the
562 published COM-type compositions relative to olivine tholeiites have been used to suggest

563 that crustal contamination is important in the generation of the COM-type parent (Leeman
564 and Manton 1971; Menzies et al. 1984; Putirka et al. 2009). In contrast, the Suite 1 HRT
565 mafic $^{87}\text{Sr}/^{86}\text{Sr}$ values are bracketed by our olivine tholeiite data, and all suite 1 and 3
566 samples fall within the broad field of SRP basalts (Fig. 6). It is possible that Suite 2 olivine
567 tholeiites may have assimilated small amounts of very radiogenic Archean upper crust,
568 whereas suites 1 and 3 magmas assimilated larger amounts of moderately radiogenic lower
569 crust. From the perceived off- and on-axis spatial relationships of the COM-type and olivine
570 tholeiites respectively, it has been argued that the ascent of the former is hindered by
571 passage through primary granitic crust compared to the relatively fast ascent of the olivine
572 tholeiites through crust replaced by basaltic sills (Christiansen and McCurry 2008; Putirka et
573 al. 2009). A similar explanation for the differences between the least evolved samples from
574 suites 1 and 2 presented here is difficult to uphold. This difficulty arises from the close
575 spatial and temporal proximity of the earliest olivine tholeiite eruptions, the Junction Butte
576 Basalt, traditionally viewed as contaminated, ($^{87}\text{Sr}/^{86}\text{Sr} = 0.70562 \pm 0.00004$ and $0.70756 \pm$
577 0.00004 2se, in our examples), to the HRT mafics ($^{87}\text{Sr}/^{86}\text{Sr} = 0.70709$ - 0.70771). All these
578 compositions were erupted at the beginning of known Yellowstone volcanism when the
579 least replacement of the pre-existing crust would be expected to have occurred.

580 **Depth/degree of partial melting.** Another possible mechanism for generating mafic
581 melts with differing chemical characteristics in the same geographic area is through source
582 partial melting variations. Leeman et al. (2009) proposed that the Snake River Plain olivine
583 tholeiites reflected melting of and equilibration with a spinel lherzolite source containing
584 minimal or no garnet at 70-100 km depth (≤ 2.8 GPa). They cited the flat chondrite-
585 normalized REE trends of primitive Snake River Plain olivine tholeiites, and low chondrite-
586 normalized Gd/Yb ratios ($[\text{Gd}/\text{Yb}]_{\text{N}} = 1.1$ - 1.7) which would increase strongly if there was

587 residual garnet in the source. The parallel REE slopes of the suites 1 and 3 samples when
588 compared to those of Suite 2 (Fig. 5) suggest, however, that there was no significant
589 difference in the depth of melting, consistent with the overlapping (Gd/Yb)_N values of
590 samples from Suite 2 (1.4-2.5) versus suites 1 and 3 (1.5-2.8).

591 Isotopic ratios of the different mafic suites can also provide clues on their source
592 zones. The slope of the Suite 2 olivine tholeiite ²⁰⁷Pb/²⁰⁴Pb and ²⁰⁶Pb/²⁰⁴Pb ratios (Fig. 8), if
593 interpreted as a pseudochron, gives an age of 2.77 ± 0.52 Ga (Supplementary Fig. 5a). This
594 apparent age is broadly comparable to the 'secondary-isochron' age of 2.5 Ga reported by
595 Doe et al. (1982), and a 2.8 Ga age from crustal xenoliths, the latter inferred to represent
596 establishment of the Wyoming craton (Leeman et al. 1985). Similar pseudochrons can be
597 regressed through data from our Suite 1 mafic samples (3.0 ± 0.22 Ga; excluding YP122;
598 Supplementary Fig. 5b) and Suite 3 samples (2.56 ± 1.1 Ga; Supplementary Fig. 5c). The
599 similar pseudochron ages between all three mafic suites indicate, based on our earlier
600 conclusion that assimilation has been minimal, that the melts of all three mafic suites
601 equilibrated with similar Archean-age regions. This inference, coupled with the
602 unradiogenic and overlapping ¹⁴³Nd/¹⁴⁴Nd isotope ratios between the different mafic suites
603 (Fig. 6) and, with the exception of one sample from our data set, plotting above the
604 Northern Hemisphere Reference Line (Fig. 8), collectively require sources for all three suites
605 within regions with ancient U and Nd enrichment. As the Suite 2 olivine tholeiite values are
606 taken to indicate that they last equilibrated with Archean subcontinental lithospheric
607 mantle (Doe et al. 1982; Hanan et al. 2008; Leeman et al. 2009), then the similarity in REE
608 patterns and Pb and Nd isotopic systematics requires that our suites 1 and 3 compositions
609 also last equilibrated with a similar source with a similar Archean history.

610 We now consider the possibility of varying degrees of partial melting of this common
611 source to generate the contrasts between the different suites, using Ba, Nb and Zr as
612 discriminants. Although all three elements are incompatible with respect to mantle mineral
613 assemblages, their degrees of incompatibility vary, with $Ba > Nb > Zr$ ($D_{Ba} = 0.01$, $D_{Nb} = 0.04$,
614 $D_{Zr} = 0.08$; Hofmann 1988). Therefore, smaller degrees of partial melting than that
615 associated with the olivine tholeiites should lead to elevated Ba/Nb, Ba/Zr and Nb/Zr ratios
616 in the resulting melts. Although the Suite 1 mafics show elevated Ba/Nb ratios, their Nb/Zr
617 ratios are lower and negatively correlated with Ba/Nb (this is also seen in Suite 3 samples).
618 These relationships are the opposite to what would be expected were the Suite 1 mafics
619 were derived from smaller-degree partial melts from a source common to the olivine
620 tholeiites (Supplementary Fig. 6). Our data are thus consistent with variations within the
621 olivine tholeiites being due to varying degrees of partial melting (Leeman et al. 2009) but
622 extrapolation of this hypothesis to generation of the parental melts for suites 1 and 3 is
623 incompatible with our data.

624 **Enrichment of the mantle source.** We next consider the possibility that magmas of
625 suites 1 and 3 were generated from mantle sources contrasting in some respect to the Suite
626 2 samples. Although the ferroan and isotopically more crustal nature of the least evolved
627 COM lavas have led to this possibility being dismissed by earlier workers (e.g. Leeman and
628 Manton 1971; Leeman et al. 1976; Reid 1995; Putirka et al. 2009), Suite 1 compositions
629 show contrasts with these 'type' COM flows (see subsequent section) that make a mantle
630 contrast worth investigating. Differing mafic compositions erupted within a single volcanic
631 field or province have elsewhere been attributed to source heterogeneity, with origins in
632 regions of variably enriched or metasomatized mantle (e.g. Feeley 2003; McGee et al. 2013;
633 Rasoazanamparany et al. 2015). Aqueous fluid has often been invoked as a mechanism for

634 locally enriching mantle sources, particularly in subduction zones through dehydration of
635 the subducting slab (e.g. McCulloch and Gamble 1991). Such aqueous-rich fluids are
636 typically enriched in LILE, and depleted in HFSE and REE that remain in the slab (McCulloch
637 and Gamble 1991; Green and Adam 2003, and references therein).

638 Eocene subduction and source enrichments associated with the Farallon slab have
639 previously been invoked in the timing and characteristics of the 55-45 Ma Absaroka Volcanic
640 Province, overlapping with the eastern border of Yellowstone (Fig. 1: Christiansen and Yeats
641 1992; Feeley 2003; Schmandt and Humphreys 2011). The calc-alkaline, andesitic Absaroka
642 eruptives have eastward-increasing K_2O contents and elevated LILE/HFSE ratios (Fig. 10:
643 Chadwick 1970; Feeley 2003). These features have been linked to partial melting of a
644 lithospheric mantle source that had been metasomatically enriched at <100 Ma by aqueous
645 fluids derived from the Farallon slab (Feeley 2003). As the slab foundered, asthenospheric
646 upwelling is proposed to have led to partial melting of the enriched region and generation
647 of Absaroka magmas. A comparable scenario has also been invoked for other volcanic fields
648 in the western U.S. (e.g. Mirnejad and Bell 2006; Lake and Farmer 2015; Brueseke et al.
649 2018). A modern low-velocity layer atop the mantle transition zone beneath the YSRP has
650 also been attributed to a zone of partial melt caused by ascending volatiles from a still-
651 dehydrating Farallon slab (Hier-Majumder and Tauzin 2017).

652 Although such a model may be applicable to the Quaternary Yellowstone eruptives,
653 a distinctive signature of samples from suites 1 and 3 is their enrichment not only in typically
654 fluid-mobile LILE (Ba, Rb) but also immobile HFSE (Zr, Ti, Nb) and REE relative to Suite 2
655 samples and the Absaroka volcanics (Fig. 10). In suites 1 and 3, the Ba enrichment coupled
656 with the lack of a significant K_2O enrichment (or depletion) precludes a significant role for
657 phlogopite while the lack of Sr enrichment precludes a significant role for carbonate in any

658 source enrichments, as mantle carbonates are commonly Sr enriched (Hoernle et al. 2002).
659 These features indicate that aqueous fluids alone cannot have generated any proposed
660 enrichment of the source zones for suites 1 and 3. However, at high pressures and
661 temperatures (>600 °C and >0.5 GPa), HFSE, such as Zr and Ti, can be soluble in high total
662 solute aqueous fluids or hydrous melts relative to dilute aqueous fluids (Manning et al. 2008;
663 Hayden and Manning 2011; Wilke et al. 2012; Louvel et al. 2013, 2014). Under such
664 circumstances HFSE may have been transported upwards from the Farallon slab to enrich
665 the overlying mantle in HFSE. Although normally this metasomatized region would descend
666 with the subducted plate to the deep mantle, thus causing the HFSE-depleted signature of
667 subduction-zone volcanism (McCulloch and Gamble 1991; Louvel et al. 2013, 2014), the
668 slowly sinking nature of the Farallon slab beneath the YSRP would mean that this HFSE-
669 enriched zone remained in the upper mantle and became available as a source region for
670 subsequent melting.

671 The geochemical signatures of suites 1 and 3 place further constraints on the nature
672 of any fluids from the Farallon slab. The similar REE patterns between the different suites
673 (Fig. 5) suggest that enrichment was uniform for all REE. Experimental data show that
674 minimal REE fractionation occurs in melts when compared to aqueous fluids, suggesting that
675 a hydrous melt was more likely the agent for HFSE enrichment (Tsay et al. 2014). Although
676 fluids generated within the garnet stability field would have an elevated LREE/HREE ratio
677 (Green and Adam 2003; Green et al. 2000), equilibration with spinel lherzolite lithospheric
678 mantle melts would likely mask any deeper signature. We thus cannot preclude ascent of
679 HFSE-enriched melts contributing to mantle enrichment from depths appropriate to the
680 garnet stability field, but the source of the parental melts to suites 1 and 3 has to have been
681 at levels above the garnet stability field.

682 The HFSE-enriched nature of suites 1 and 3 relative to the olivine tholeiites thus
683 suggests that melting of a melt-enriched mantle source is a plausible mechanism for the
684 generation of their parental melts. Following the 2.8 Ga magmatic/metamorphic
685 enrichment event (inferred here and in other works from Pb isotopic signatures: Doe et al.
686 1982; Leeman et al. 1985), a second, Cretaceous-Eocene enrichment event has been
687 proposed for the Absaroka Volcanic Province by Feeley (2003) and for volcanism farther east
688 by Mirnejad and Bell (2006). We consider the possible role of this second enrichment event
689 in our Suite 1 samples through the Sr isotopic systematics, where there is a positive
690 correlation between $^{87}\text{Sr}/^{86}\text{Sr}$ and Rb/Sr and no apparent relationship to 1/Sr (Fig. 7). We
691 argue (as contrasted previously for the olivine tholeiites) that this trend is not due to crustal
692 contamination but instead controlled by Rb, which is enriched in the Suite 1 samples (Fig. 9).
693 One interpretation for this trend, therefore, is as a pseudochron, potentially reflecting the
694 age of the secondary Rb-enrichment event. Using age-corrected (to 2.08 Ma) $^{87}\text{Sr}/^{86}\text{Sr}$ and
695 $^{87}\text{Rb}/^{86}\text{Sr}$ ratios, a pseudochron of 68 ± 45 Ma (MSWD=193) is generated using Isoplot
696 (Supplementary Fig. 7). Although imprecise, this age estimate is the same within error as
697 that proposed for the metasomatic event associated with volcanism in the Absaroka field
698 and elsewhere in Wyoming (Feeley 2003; Mirnejad and Bell 2006; Schmandt and
699 Humphreys 2011). Note also that this pseudochron correlation, if valid, would indicate an
700 original $^{87}\text{Sr}/^{86}\text{Sr}$ for the source region of the Suite 1 HRT mafics of 0.7069 ± 0.0004 , identical
701 within error to the inferred initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios of Yellowstone-Snake River Plain olivine
702 tholeiites (0.7067 ± 0.001 : McCurry and Rodgers 2009). If, in contrast, measured $^{87}\text{Sr}/^{86}\text{Sr}$
703 values are age-corrected to an Archean age (i.e. ≥ 2.5 Ga), then initial $^{87}\text{Sr}/^{86}\text{Sr}$ values would
704 have been < 0.7 and the slope of the pseudochron much steeper (Fig. 7a). This initial value is
705 implausibly low and indicates that Rb/Sr enrichment, if related to metasomatism, did not

706 occur in the Archean but must be a more recent event. We consider that the younger event
707 is recorded in Sr but not Pb isotopic systematics due to the elevated Rb/Sr in Suite 1 relative
708 to Suite 2 samples. In contrast the U/Pb values are similar between the two suites, thus,
709 allowing for a selective Sr isotopic overprint.

710 We therefore suggest that as the Farallon slab foundered in the Cretaceous-Eocene,
711 a variety of fluids (aqueous fluids and hydrous melts) with varying compositions were
712 released from the slab to migrate upwards into the lithospheric mantle resulting in a
713 heterogeneous source region for subsequent melting and volcanism (Fig. 11). Aqueous
714 fluids, enriched in LILE, likely migrated further (cf. Rubatto and Hermann 2003) and the LILE-
715 enriched regions preferentially melted as a consequence of asthenospheric upwelling to
716 generate the Absaroka volcanics with their high LILE/HFSE signature (Fig. 10). We infer that
717 during subsequent impingement of the modern Yellowstone thermal anomaly (plume) the
718 lithospheric mantle beneath the YPVF underwent partial melting. This 'modern' melting
719 event generated parental melts for the enriched Suite 1 mafics plus Suite 3 COM-type
720 magmas from HFSE-enriched regions, and olivine tholeiitic melts from adjacent unaltered
721 regions that were unaltered or from which the LILE were stripped during Absaroka
722 magmatism.

723

724 **Intra-suite trends**

725 Following generation of the parental melts, subsequent processes controlled the
726 distinct intra-suite compositional arrays, predominantly fractional crystallization and
727 assimilation, that we discuss in turn below. Both processes, particularly at low pressures,
728 have been invoked for the COM-trend along the Snake River Plain (Leeman et al. 1976;
729 Christiansen and McCurry 2008; McCurry et al. 2008). Although plagioclase and olivine are

730 commonly present in olivine tholeiite and COM eruptives (Putirka et al. 2009), major and
731 trace element trends may fingerprint any other 'cryptic' fractionation effects, reflecting
732 crystallization of phases not observed in the erupted material. Sub-parallel major element
733 trends (e.g. increasing $\text{Na}_2\text{O} + \text{K}_2\text{O}$ with SiO_2 , and decreasing MgO , FeO and CaO) in all three
734 suites suggest a control by similar olivine plus plagioclase fractionation assemblages (Tilley
735 and Thompson 1970), with the additional presence of apatite to explain a decrease in P_2O_5
736 in suites 1 and 3, as inferred for COM flows (Fig. 3b: Leeman et al. 1976; Reid 1995). Trace
737 element patterns, however, indicate a contrast in the relative role of fractionation of the
738 different phases in generating the intra-suite trends. The sharp decrease in Ni/Sc ratios with
739 decreasing MgO and $\text{CaO}/\text{Al}_2\text{O}_3$ in Suite 2 olivine tholeiite samples, compared to a relatively
740 flat trend defined by suites 1 and 3, suggest a much greater role for olivine in Suite 2
741 samples (Supplementary Fig. 8). Conversely, the positive relationship between Sr and other
742 LILE (e.g. Rb and Ba) in Suite 2 samples, coupled with an increase of Rb with SiO_2 , indicates a
743 bulk incompatibility of Sr and a reduced role for plagioclase (Fig. 9). This inference is
744 supported by an increase in Sr/Sc with increasing Ni/Cr in Suite 2 samples (Supplementary
745 Fig. 9). In contrast, samples from suites 1 and 3 show an inverse relationship between Sr and
746 Rb (Fig. 9) and, as Rb increases with SiO_2 in all suites, these relationships collectively indicate
747 a greater influence of plagioclase. Deviations from these trends (e.g. YR294, a Junction Butte
748 Basalt sample with 36 ppm Rb : Fig. 9) are likely to reflect variable degrees of contamination
749 by country rocks.

750 Quantitative fractional crystallization modelling was undertaken using the modeling
751 program of Ersoy and Helvacı (2010) with their built-in partition coefficients for "basic"
752 magmas. Starting compositions were the samples from each suite with the highest $\text{Mg}\#$
753 (YR291 and YR422 for suites 2 and 3 respectively) or a combination of high $\text{Mg}\#$ and low

754 SiO₂ (YP122: Suite 1). Results from this modeling support the previous inferences, with the
755 Suite 2 trend most consistent with a crystallizing assemblage of 60% clinopyroxene, 35%
756 olivine and 5% plagioclase (Fig. 9, Supplementary Fig. 9). In contrast, as shown by an
757 antithetic evolutionary trend (Supplementary Fig. 9), the Suite 3 sample variations can be
758 replicated by a fractional crystallization of an assemblage containing 80% plagioclase, 15%
759 olivine and 5% clinopyroxene. This assemblage is plagioclase richer and pyroxene poorer
760 than those observed in the experiments of Whitaker et al. (2008), conducted on a similar
761 range of compositions. However, the model mineral assemblage and fractionation trend are
762 at odds with the lack of a negative Eu anomaly in the Suite 3 samples (Fig. 5) as would be
763 expected with significant plagioclase fractionation. The compositional variation of this suite
764 is thus somewhat enigmatic and would benefit from further study. Both curves detailed
765 above show uniform Zr/Nb ratios with changing Sr (Fig. 9), consistent with the similar
766 incompatible nature of both elements in the fractionating assemblage.

767 In Suite 1 samples, although the uniform Sr/Sc ratio with decreasing Ni/Cr
768 (Supplementary Fig. 9) is consistent with a moderately plagioclase-dominant assemblage (45%
769 plagioclase, 30% olivine and 25% clinopyroxene), this assemblage does not fully replicate Sr,
770 Ba and Rb trends observed in the Suite 1 data. In addition, the modeled trend is antithetic to
771 the strong correlation between Zr/Nb and Sr observed in the sample data (Fig. 9). This
772 correlation requires either the fractionation of zircon or a Nb-rich phases. It is possible that
773 the decrease in Zr/Nb ratios, with decreasing Sr, is related to zircon fractionation
774 accompanying the bulk fractionation trend, as zircon is inferred to be stable at ~60 wt%
775 SiO₂ in similarly Zr-rich compositions (McCurry et al. 2008). There is no petrographic
776 evidence, however, for zircon or a Nb-rich phase being present, either in Suite 1 materials or
777 the COM lava flows (Leeman et al. 1976). Furthermore, zircon saturation temperatures

778 (Watson and Harrison 1983 calibration), yield temperatures of 860-960 °C for the HRT
779 mafics, similar to temperatures of 770-960 °C for Suite 3 samples that show constant Zr/Nb
780 values. These are too low to be plausible pre-eruptive temperatures for these melts,
781 particularly given that only one COM flow has yielded average plagioclase thermometry
782 estimates of <1,000 °C (Putirka et al. 2009). We also note that Zr concentrations broadly
783 increase in Suite 1 dense mafics across the compositional range (Fig. 4b), the opposite of
784 what would be expected with zircon fractionation. Although Zr/Nb correlates closely with Sr
785 contents in Suite 1 samples, Sr is scattered throughout the suite and does not show clear
786 trends with other proxies of evolution (e.g. Rb: Fig. 9).

787 The large degrees of scatter in data from all three suites and the poor fit of some
788 modelled trends (e.g. Supplementary Fig. 9), indicate that not all intra-suite trends can be
789 explained exclusively by fractional crystallization and some crustal assimilation/mixing is
790 also required. Although there is a rhyolite-mixing trend observed within the Suite 1 scoria
791 samples (Fig. 4), as discussed earlier (Crustal assimilation section), this process does not
792 explain the overall increases in Ba and Zr with SiO₂ observed in data from this suite. A
793 diminished role for assimilation is further supported by the decoupled behavior of P₂O₅ and
794 Zr within the Suite 1 mafics (cf. Figs. 3b, 4b), which behavior is typically attributed to
795 fractionation rather than mixing (e.g. Lee and Bachmann 2014). Furthermore, the lack of a
796 relationship between ⁸⁷Sr/⁸⁶Sr and 1/Sr (Fig. 7) in the samples from suites 1 and 3, is
797 inconsistent with a strong assimilation control on the compositional array.

798 Whilst the Suite 2 samples show a large range in ⁸⁷Sr/⁸⁶Sr and wide ranges in
799 incompatible elements (e.g. Rb: Fig. 9), commonly attributed to crustal assimilation (e.g.
800 Hildreth et al. 1991; Christiansen and McCurry 2008), there are complexities to this simple
801 model. For example, samples with Sr differing by a factor of 2 have very similar Sr isotopic

802 compositions and the highest-Sr lava (YR 292, Basalt of the Narrows; 546 ppm) has the
803 lowest $^{87}\text{Sr}/^{86}\text{Sr}$ (0.70373 ± 0.00005 2se). There is also no strong correlation between
804 $^{87}\text{Sr}/^{86}\text{Sr}$ and $1/\text{Sr}$, which would be expected with significant crustal assimilation of a
805 homogenous contaminant (Fig. 7).

806 Therefore, although assimilation is likely to occur in all suites, any simple explanation
807 involving incorporation of a common assimilant is insufficient to explain the compositional
808 arrays within each suite. The diversity within the Suite 1 samples, which is incompatible with
809 simple fractional crystallization and assimilation models discussed above, is surprising. It is
810 possible that variations in the suite are related to subtle differences in the
811 degree/composition of the initial enrichment, but why any such intra-suite source variation
812 should remain distinct during ascent through the crust and eruption is puzzling.

813

814 **Comparisons and contrasts with the Craters of the Moon lava field**

815 Our data allow us to compare our Suite 1 HRT mafics and Suite 3 local COM-type
816 samples with their counterparts from the Craters of the Moon lava field (Leeman 1976;
817 Putirka et al. 2009). In the first instance there are some contrasts. Suite 1 HRT mafics show
818 relatively elevated TiO_2 , Ba and Zr, and no correlation between Ba and Sr whereas Suite 3
819 samples and published COM data show a negative correlation (Fig. 9b). Furthermore, Suite 1
820 samples show a strong decrease in Zr/Nb with decreasing Sr, whereas Suite 3 samples show
821 uniformity of Zr/Nb (Fig. 9c). Different model crystallizing assemblages (see Fig. 9 caption)
822 yield broadly constant Zr/Nb with changing Sr, indicating that the Zr/Nb versus Sr
823 relationships in Suite 1 samples is unlikely to be controlled by fractional crystallization. The
824 above two features suggest that rocks of Suite 3 and the 'type' COM areas have signatures

825 that are more fractionation-controlled, whereas the Suite 1 HRT mafics have a further,
826 enriched signature, which we relate to mantle source variations (previous section).

827 As previously mentioned, the COM samples reported by Putirka et al. (2009) are
828 more radiogenic (i.e. higher $^{87}\text{Sr}/^{86}\text{Sr}$) than our samples from suites 1 and 3 which, in simple
829 terms, reflects variations in the amount of crustal assimilation. This explanation is
830 supported by the negative correlation between $^{143}\text{Nd}/^{144}\text{Nd}$ and $^{87}\text{Sr}/^{86}\text{Sr}$ in COM samples,
831 forming a trend towards the average crustal isotopic composition (Fig. 6: Leeman 1976;
832 Putirka et al. 2009). Furthermore, the initial increase in $^{87}\text{Sr}/^{86}\text{Sr}$ with Rb/Sr ratios in COM-
833 type flows from the Craters of the Moon (Fig. 7a), starting from values similar to the least
834 radiogenic Suite 1 HRT mafics, indicates enhanced incorporation of a more radiogenic
835 assimilant than that inferred in the HRT mafics. The trend observed is that expected in
836 material of Archean age (Fig. 7a), consistent with the assimilation of ancient crust in the
837 type Craters of the Moon area.

838

839

IMPLICATIONS

840 Our results have a number of implications for the onset of large-scale volcanism in
841 the Yellowstone area. The early eruption of two contrasting mafic suites (1 and 2) reflects a
842 complex mafic root zone beneath the early Yellowstone magmatic system. Our data
843 represent the first documentation of COM-type material associated with rhyolites within the
844 YPVF, as all mafic rocks reported so far have been of olivine tholeiitic affinity, whether
845 erupted close to, or outside the focus of silicic volcanism (Christiansen 2001; Christiansen
846 and McCurry 2008; Pritchard et al. 2013). The presence of the Suite 1 HRT mafic materials
847 indicate that genesis of such 'COM-like' magmas is more widespread than previously

848 thought, and that such magmas can be present at the onset of, rather than always post-
849 dating, any focus of silicic volcanism. It also shows that a COM-like mafic lineage can also be
850 intimately associated with large silicic eruptions, in contrast to its currently viewed
851 association with small-scale basaltic and fractionated intermediate to silicic eruptives in the
852 Snake River Plain (Kuntz et al. 1986; McCurry et al. 2008; Putirka et al. 2009).

853 The Suite 1 HRT compositions have implications for modelling of the silicic
854 magmatism at Yellowstone. Although the HRT mafics are distinct in their elemental
855 compositions, their isotopic compositions fall within the range of the olivine tholeiites. Any
856 isotopic leverage on the HRT silicic system from the HRT mafic compositions is thus
857 comparable to those from olivine tholeiites. However, the HRT mafic compositions are
858 critical when conducting elemental modelling of the Yellowstone silicic system. The least
859 evolved published HRT analysis has 70.8 wt% SiO₂ and 2670 ppm Ba (Hildreth et al. 1991).
860 Our discovery of basaltic compositions (~50 wt% SiO₂) with ~2,500 ppm Ba, as opposed to
861 ~500 ppm in the tholeiites at a similar silica content, has significant impacts when
862 discussing petrogenesis of the least-evolved rhyolites at Yellowstone and the nature of the
863 components involved (e.g. Christiansen and McCurry 2008).

864 Our study has shown a general spatial variation in the distribution of mafic
865 components in the HRT, with dense clasts erupted with ignimbrite member A concentrated
866 to the SW and scoria associated with upper member B concentrated to the north. Although
867 only mafic eruptives belonging to the olivine tholeiite suite have so far been documented in
868 the YPVF, we consider it possible that evidence for COM-type mafic contributions may be
869 found in younger silicic deposits. However, such contributions may have been diluted out by
870 continuing ascent of more voluminous olivine tholeiite melts. The close proximity of young
871 eruptions of olivine tholeiite and COM-type magmas in and just west of the Island Park

872 segment of the HRT caldera (e.g. Christiansen 2001; Iwahashi 2010) demonstrates that
873 these contrasting magma types do not reflect mutually exclusive magma-generating
874 systems.

875 Although our Suite 2 Yellowstone olivine tholeiites represent a limited data set, it is
876 clear that there is significant complexity within it, as well as within the broader SRP olivine
877 tholeiite literature data. Large variations within major and trace elemental compositions
878 (e.g. SiO₂ and Sr), and isotopic compositions (e.g. Pb and Sr isotopes) have previously been
879 ascribed to varying degrees of contamination from crustal rocks or rhyolite melts (Hildreth
880 et al. 1991; Christiansen and McCurry 2008). However, the lack of observed trends between
881 elemental and isotopic compositions (e.g. Fig. 7) suggests there are further complexities
882 than these. Furthermore, there is considerable variation within stratigraphically-grouped
883 basalts. The multiple flow pre-HRT Junction Butte Basalt (Christiansen 2001) is considered
884 evolved and contaminated (Hildreth et al. 1991). Although possibly the case for some flows
885 (e.g. YR294: SiO₂ = 54.3 wt%, Rb = 36 ppm, ⁸⁷Sr/⁸⁶Sr = 0.70756), others show a more
886 primitive signature (e.g. YR297: SiO₂ = 49.1 wt%, Rb = 5 ppm, ⁸⁷Sr/⁸⁶Sr = 0.70561). This
887 diversity suggests that contamination processes are localized and variable, and likely involve
888 a variety of assimilants. Consequently, we feel that the Yellowstone olivine tholeiites
889 deserve renewed scrutiny using current compositional and geochronological capabilities to
890 fully identify variations within the suite and the compositional role they play in the silicic
891 magmatic system after the HRT eruption.

892 Although the complex compositional characteristics of the Suite 1 HRT mafics (and
893 other COM compositions) mean there are issues with any proposed petrogenetic process
894 (Table 4), we would argue that our proposed model of HFSE-enriched melt metasomatism in
895 the mantle source for generation of the least-evolved melts is the most compatible with our

896 data. Although the generation of high P-T, HFSE-rich fluids is likely to be a common process
897 in subduction zones (Louvel et al. 2013, 2014), causing enrichment of the overlying mantle
898 wedge, primary enrichment is likely to be in a zone immediately overlying the subducted
899 slab. The lower solubility HFSE elements would be likely to be precipitated first, leaving
900 dominantly more-soluble elements (e.g. LILE) to ascend farther into the sub-continental
901 lithospheric mantle into higher temperature regions and the focus of melting, while the
902 HFSE-enriched zones founder with the subducted slab (Louvel et al. 2013, 2014). The
903 unusual slab geometry beneath the YPVF, of a stationary or slowly descending slab in the
904 asthenosphere, means that in this case the HFSE-enriched mantle, generated during Eocene
905 and onwards foundering of the slab, has remained in the zone of potential melt generation.
906 Therefore, with the arrival of a focused, albeit weak, thermal anomaly (the Yellowstone
907 plume, *sensu lato*), the HFSE-enriched mantle material was located in the melting window
908 when the lithospheric mantle was raised above its solidus at the onset of YPVF volcanism
909 and birth of the HRT silicic system. The higher alkali contents of these metasomatized
910 regions would also promote melting through reducing the solidus temperature (Hirschmann
911 2000).

912 It seems clear that the formation of the HRT mafics and COM-type compositions is a
913 localized feature requiring the alignment of a variety of processes. There are additional
914 complexities related to the local setting and conditions, particularly within the Sr isotopic
915 signatures, between the Suite 1 HRT mafics, the Suite 3 COM-type flows analyzed here, and
916 the more radiogenic nature of the flows from the Craters of the Moon type area (Leeman
917 1974; Leeman et al. 1976; Putirka et al. 2009). However, with a full major, trace and isotopic
918 dataset we are able to offer a more consistent mechanism for generation of the parental
919 magmas for the HRT mafic suite that may also be applicable to the broader COM suite. We

920 suggest that re-evaluation of the overall characteristics of the Craters of the Moon series of
921 rocks may also be required.

922

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933

934

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1229 **Figure Captions**

1230 **Figure 1:** Map of the Yellowstone- Snake River Plain area (adapted from Kuntz et al. 1982
1231 and Christiansen et al. 2007). Within the Yellowstone Plateau volcanic field, the red
1232 line marks the mapped rim of the caldera for the Huckleberry Ridge Tuff (HRT)
1233 eruption, and the location of the pre-HRT Junction Butte Basalt (JBB) is also shown
1234 (from Christiansen 2001). Quaternary basaltic lava fields along the Snake River Plain
1235 (SRP) are also outlined, including those of olivine tholeiitic affinity (purple; SH-
1236 Shoshone, W-Wapi, KB-King's Bowl, NR-North Robbers, SR-South Robbers, CG-Cerro
1237 Grande, HHA-Hells Half Acre) and those containing Craters of the Moon-type
1238 magmas (orange; COM-Craters of the Moon, SHP-Spencer-High Point). The 85 km
1239 Great Rift (GR) and the boundary of Yellowstone National Park (YNP) are also shown
1240 for reference.

1241 **Figure 2:** Photographs of representative HRT dense mafic (top) and scoria (bottom) clasts
1242 found dominantly in HRT members A and B, respectively. The crenulated margin of
1243 the HRT dense mafic clast indicates its juvenile nature.

1244 **Figure 3:** SiO₂ versus (a) total alkalis and b) P₂O₅ for the three suites of mafic samples

1245 analyzed in this study and relevant compositional fields from published data (see

1246 text for details). Although there is overlap between the alkali compositions of the

1247 mafic suites, P₂O₅ compositions are distinctly higher in the Suite 1 HRT samples. M

1248 denotes mingled clasts (YP185 and YP188 discussed in the text). Classification fields

1249 in panel (a) are from Le Bas and Streckeisen (1991). 2sd uncertainties are smaller

1250 than the symbol sizes.

1251 **Figure 4:** Compositional diagrams for the samples analyzed in this study and relevant

1252 compositional fields from published data for SiO₂ versus (a) Ba and (b) Zr, showing

1253 the enrichment in both LILE and HFSE in samples from suites 1 and 3. HRT dense

1254 mafics and scoria (Suite 1) are further enriched in Ba relative to COM-type flows,

1255 including our Suite 3 samples. See text for sources of published data fields. A

1256 possible mixing trend with rhyolite is defined by the scoria samples, trending

1257 towards the mingled clast (M). 2sd uncertainties are smaller than the symbol size.

1258 **Figure 5:** Chondrite-normalized (McDonough and Sun 1995) REE plot showing enriched but

1259 sub-parallel trends in the samples from suites 1 and 3 relative to Suite 2. There is no

1260 significant Eu anomaly in any of the samples analyzed in this study.

1261 **Figure 6:** ¹⁴³Nd/¹⁴⁴Nd versus ⁸⁷Sr/⁸⁶Sr (age corrected) for samples analyzed in this study and

1262 isotopic fields for relevant comparators from the YSRP area (see text for published

1263 data sources). All of the samples analyzed in this study fall within the field covered

1264 by Snake River Plain basalts. The radiogenic nature of the Sr isotopic systematics,

1265 and unradiogenic Nd isotopic compositions, is taken to indicate interaction of the

1266 mafic suites with Archean subcontinental lithospheric mantle (Hanan et al. 2008).

1267 Arrow indicates trend of evolved lava flows from Craters of the Moon (Putirka et al.

1268 2009) towards average continental crust ($^{87}\text{Sr}/^{86}\text{Sr} = 0.72$; $^{143}\text{Nd}/^{144}\text{Nd} = 0.5118$:
1269 Hofmann 1997). Black asterisk represents the focus of collated Yellowstone-Snake
1270 River Plain basalts ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7067$, $^{143}\text{Nd}/^{144}\text{Nd} = 0.510245$: McCurry and Rodgers
1271 2009). Archean crust typically has $\epsilon_{\text{Nd}} < -15$ (McCurry and Rodgers 2009, and
1272 references therein). Approximate values for enriched mantle 1 (EM1) and 2 (EM2)
1273 from Zindler and Hart (1986). See text for explanation of ϵ_{Nd} . 2se errors are smaller
1274 than the symbol size.

1275 **Figure 7:** Plots showing the relationship between elemental and isotopic Sr compositions.
1276 Age-corrected $^{87}\text{Sr}/^{86}\text{Sr}$ values versus (a) Rb/Sr and (b) 1/Sr show a positive
1277 relationship between $^{87}\text{Sr}/^{86}\text{Sr}$ and Rb/Sr for the Suite 1 HRT mafics ($R^2 = 0.65$) but no
1278 clear relationship with 1/Sr. Suite 2 olivine tholeiite samples analyzed for this study
1279 show a large range in isotopic compositions with a minimal range in Rb/Sr. See text
1280 for published data sources. Vectors show modelled gradients for hypothetical Rb/Sr
1281 enrichment events at 2.77 Ga and 68 Ma from a common initial $^{87}\text{Sr}/^{86}\text{Sr}$ and
1282 multiple initial Rb/Sr ratios. 2se error bars ($^{87}\text{Sr}/^{86}\text{Sr}$) are smaller than the symbols.

1283 **Figure 8:** Pb-isotopic compositions for the samples analyzed for this study and fields for
1284 published data (see text for published data sources). All samples, except for the
1285 Basalt of the Narrows (YR292) in panel (a), plot above the Northern Hemisphere
1286 reference line (NHRL: Hart 1984). Suite 1 HRT mafics show elevated $^{207}\text{Pb}/^{204}\text{Pb}$ and
1287 $^{208}\text{Pb}/^{204}\text{Pb}$ ratios for a given value of $^{206}\text{Pb}/^{204}\text{Pb}$ relative to the Suite 2 olivine
1288 tholeiites. Enriched mantle 1 (EM1) and 2 (EM2) and depleted mantle (DMM) values
1289 from Zindler and Hart (1986). Individual 2se errors are smaller than the symbol size
1290 unless indicated otherwise.

1291 **Figure 9:** Sr vs (a) Rb, (b) Ba and (c) Zr/Nb for samples analyzed in this work. Suite 1 HRT
1292 mafics are differentiated from Suite 3 COM samples by their elevated Ba and no
1293 correlation with Sr, (panel b) and strongly decreasing Zr/Nb (panel c), the latter
1294 which requires crystallization of zircon or a Nb-rich phase, both of which are unlikely
1295 to have been present. Note the positive correlation between Sr and other LILE in
1296 Suite 2 olivine tholeiite samples, indicating the incompatibility of Sr in these samples
1297 despite the presence of plagioclase. Red, purple and orange dashed lines show
1298 modelled fractional crystallization trends for Suite 1 (YP122 as starting composition,
1299 crystallizing assemblage of 45% plagioclase, 30% olivine, 25% clinopyroxene), Suite 2
1300 (YR291 as starting composition, crystallizing assemblage of 60% clinopyroxene, 35%
1301 olivine, 5% plagioclase) and Suite 3 (YR422 as starting composition, crystallizing
1302 assemblage of 80% plagioclase, 15% olivine, 5% clinopyroxene), respectively.
1303 Modelling was done using the modeler from Ersoy and Helvacı (2010). Black crosses
1304 represent 9% increments of crystallization. Suite 1 samples have Zr/Nb ratios not
1305 consistent with a fractionation control. Suite 2 and 3 samples are consistent with an
1306 clinopyroxene- and plagioclase-dominated fractionation signature respectively,
1307 suggested by antithetic behavior of Sr with increasing Ba (panel b) and uniformity in
1308 Zr/Nb (panel c). See text for sources of published data. M denotes mingled scoria
1309 clast YP188.

1310 **Figure 10:** Ocean Island Basalt-normalized (Sun and McDonough 1989) multi-element
1311 diagram showing the enriched nature in LILE and HFSE of the Suite 1 HRT mafics and
1312 Suite 3 COM-type materials relative to Suite 2 olivine tholeiites. Absaroka Volcanic
1313 Province data from Feeley (2003) show a high LILE/HFSE ratio typical of hydrous
1314 fluid-enriched subduction zone volcanism.

1315 **Figure 11:** Summary cartoon showing our proposed late Cretaceous-Eocene enrichment of
1316 the lithospheric mantle beneath Yellowstone by fluids derived from the subducted
1317 and foundering Farallon slab. Aqueous fluids with high LILE/HFSE ratios ascended
1318 and enriched the source region of the Eocene Absaroka Volcanic Province.
1319 Contemporaneously, solute-rich, LILE plus HFSE-enriched hydrous melts ascended
1320 into the base of the lithospheric mantle. Aqueous fluid enriched regions
1321 preferentially melted during asthenospheric upwelling to generate the Absaroka
1322 Volcanic Province. The renewal of volcanism in the YPVF in the Quaternary, with the
1323 arrival of a thermal anomaly (the Yellowstone plume), melted the heterogeneous
1324 lithospheric mantle. Unaltered regions yielded the parental melts to the olivine
1325 tholeiites (Suite 2, e.g. the Junction Butte Basalt) and enriched zones yielded the
1326 parental melts to the Suite 1 HRT mafics. The latter ascended to be intercepted by
1327 the growing HRT silicic magma system, syn-eruptively in the case of the samples
1328 analyzed here. Younger COM-type lavas erupted immediately west of the HRT
1329 caldera (Suite 3) also show evidence for derivation from a HFSE-enriched source.

1330 Table 1. Summary of the sample suites analyzed for this study.

Sample group	Host unit	Characteristics
Suite 1: dense mafics	HRT member A and rarely in member B	Small (<10 cm), dense, rounded, grey-green (rarely oxidised red) clasts often with a crenulated margin indicating a juvenile nature (Fig. 2a). Commonly aphyric (devitrified) with rare sparsely porphyritic samples containing euhedral feldspars.
Suite 1: scoria	Top of HRT member B	Black, or purple-red (where oxidised), aphyric, moderately vesicular material which occurs as discrete clasts or streaks within rhyolitic pumice. Where mingled in pumice, the scoria often includes up to 1 cm feldspar xenocrysts derived from the host pumice (Fig. 2b).
Suite 2: Snake River Plain olivine tholeiites	Pre and post HRT olivine tholeiites lava flows (Christiansen 2001)	Aphyric to sparsely porphyritic lava flows with olivine and plagioclase dominant mineral assemblages.
Suite 3: COM eruptives	Spencer-High Point (SHP) volcanic field (Iwahashi 2010)	Black-red scoriaceous bombs and lava flows (only pyroclastic material sampled). Ranges in crystallinity from 0-15%. Porphyritic crystal-rich materials contain euhedral feldspars up to 2 cm in length.

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	Sample	Host unit	SiO ₂	TiO ₂	FeO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅	Mg#	Sc	V	Cr	Ni	Rb	Sr	Zr	Nb	Ba	La	Yb	Hf	U
Suite 1	YP122	HRT A Dense mafic	50.79	3.00	14.22	2.42	7.24	3.32	1.80	1.52	23	36	132	5.1	5.8	46	340	1575	29	2863	95	10.1	26	1.7
	YP244	HRT A Dense mafic	59.50	2.09	10.46	0.60	4.12	3.62	2.97	1.09	9	21	69	5.9	6.0	63	247	1386	43	3508	76	5.8	26	2.6
	YP246	HRT A Dense mafic	58.10	2.51	9.58	0.83	4.94	3.84	2.92	1.39	13	25	107	5.3	5.2	61	269	1099	58	2641	85	6.2	22	2.4
	YP449	HRT A Dense mafic	52.28	3.47	13.10	0.74	5.66	3.26	2.23	1.69	9	43	121	21.2	4.7	46	342	1828	40	3430	142	9.9	33	2.1
	YP071 BLACK	HRT B Scoria	55.08	2.20	12.46	2.52	6.03	3.39	2.40	1.12	27	29	88	5.2	5.4	54	277	1545	37	3802	59	6.6	28	2.2
	YP266 SCORIA	HRT B Scoria	57.98	1.94	11.10	2.19	5.21	2.90	2.95	0.97	26	24	76	2.8	5.0	67	247	1266	39	3358	67	6.3	23	2.5
	YP334B	HRT B Scoria	56.79	2.03	11.50	2.36	5.40	3.41	2.90	1.00	27	26	80	5.4	5.2	68	265	1353	39	3530	72	6.6	26	2.6
Suite 2	YR291	Warm River Basalt	47.18	1.20	10.71	10.77	10.70	2.19	0.21	0.19	64	22	241	538	190	3.1	152	77	17	120	6.3	1.3	1.9	0.1
	YR292	Basalt of the Narrows	49.39	1.91	10.35	6.68	8.48	3.30	1.15	0.36	53	20	223	48	44	22	546	172	33	423	26	1.5	3.9	0.6
	YR294	Junction Butte Basalt	54.28	1.94	10.24	4.24	7.68	3.06	1.62	0.41	42	20	213	20	26	36	366	236	19	641	30	2.1	5.4	0.9
	YR297	Junction Butte Basalt	49.09	1.92	11.45	6.90	9.97	2.65	0.40	0.23	52	17	258	65	83	5.2	320	146	12	168	9.0	1.2	3.6	0.2
	YR302	Gerrit Basalt	46.39	1.96	12.63	7.67	10.02	2.74	0.27	0.30	52	21	278	101	104	2.1	250	137	10	174	8.9	1.3	3.2	0.2
Suite 3	YR305	High Point scoria cone	56.53	1.31	10.72	1.55	4.74	4.28	3.35	0.60	20	19	2.8	37	1.0	58	185	1973	87	2185	63	8.1	36	3.5
	YR418	Un-named scoria cone	50.19	2.47	13.71	3.09	6.58	3.69	2.30	1.61	29	24	68	11	2.2	65	234	1089	83	1716	107	9.2	20	2.2
	YR420	Un-named scoria cone	50.53	2.45	13.62	3.07	6.49	3.58	2.33	1.58	29	25	73	3.2	2.4	70	245	1182	87	1814	114	9.2	22	2.3
	YR425	Blacks Knoll	47.26	3.01	14.13	3.72	8.01	3.79	1.57	2.20	32	23	104	2.9	3.3	34	347	753	61	1262	92	7.7	14	1.3

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1333 Table 2: Major and trace element analyses of selected samples from each of the suites in this study. Oxides are in wt% and elemental concentrations in ppm.
 1334 Full data set can be found in Electronic Appendix 1.

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	Sample	Host unit	⁸⁷ Sr/ ⁸⁶ Sr	2se	¹⁴³ Nd/ ¹⁴⁴ Nd	2se	²⁰⁶ Pb/ ²⁰⁴ Pb	2se	²⁰⁷ Pb/ ²⁰⁴ Pb	2se	²⁰⁸ Pb/ ²⁰⁴ Pb	2se
Suite 1	YP122	HRT A Dense mafic	0.70721	3.69E-06	0.51231	2.79E-06	16.952	2.75E-03	15.445	3.65E-03	37.919	1.18E-02
	YP244	HRT A Dense mafic	0.70754	3.43E-06	0.51229	6.18E-06	16.892	1.12E-03	15.463	1.37E-03	37.943	4.28E-03
	YP246	HRT A Dense mafic	0.70728	3.57E-06	0.51232	4.73E-06	16.878	9.26E-04	15.458	9.98E-04	37.925	2.92E-03
	YP449	HRT A Dense mafic	0.70731	3.94E-06	0.51229	3.84E-06	16.882	9.26E-04	15.461	1.05E-03	37.937	3.18E-03
	YP071BLACK	HRT B Scoria	0.70751	3.46E-06	0.51235	5.15E-06	16.968	1.98E-03	15.481	2.55E-03	38.028	8.17E-03
	YP266SCORIA	HRT B Scoria	0.70771	3.63E-06	0.51231	2.42E-06	16.947	7.74E-04	15.476	8.22E-04	38.016	2.36E-03
	YP334B	HRT B Scoria	0.70769	3.76E-06	0.51232	4.10E-06	16.978	1.07E-03	15.482	1.24E-03	38.023	3.79E-03
Suite 2	YR291	Warm River Basalt	0.70604	3.13E-06	0.51244	3.82E-06	17.309	2.41E-03	15.511	2.35E-03	38.233	6.31E-03
	YR292	Basalt of the Narrows	0.70373	5.03E-06	0.51251	4.81E-06	16.665	3.35E-03	15.297	4.55E-03	36.466	1.44E-02
	YR294	Junction Butte Basalt	0.70756	3.87E-06	0.51207	3.71E-06	15.859	1.68E-03	15.272	2.26E-03	36.782	7.05E-03
	YR297	Junction Butte Basalt	0.70561	3.89E-06	0.51251	4.03E-06	16.943	1.27E-03	15.415	1.32E-03	37.505	3.68E-03
	YR302	Gerrit Basalt	0.70808	3.81E-06	0.51244	4.25E-06	17.031	2.04E-03	15.478	2.62E-03	37.811	8.32E-03
Suite 3	YR305	High Point scoria cone	0.70577	3.66E-06	0.51239	5.56E-06	17.218	1.06E-03	15.479	1.28E-03	38.142	3.97E-03
	YR418	Un-named scoria cone	0.70581	3.70E-06	0.51239	3.25E-06	17.208	1.36E-03	15.476	1.34E-03	38.131	3.71E-03
	YR420	Un-named scoria cone	0.70588	3.56E-06	0.51240	2.65E-06	17.158	1.10E-03	15.479	1.05E-03	38.150	2.82E-03
	YR425	Blacks Knoll	0.70789	4.43E-06	0.51225	3.32E-06	17.361	5.73E-03	15.507	5.28E-03	38.069	1.34E-02

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1342 Table 3: Isotopic ratios from selected samples from each of the suites in this study.

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Process	Evidence for	Evidence against
Extreme fractional crystallization of an olivine tholeiitic parent magma	Using $\bar{D} = 0$ for selected trace elements (e.g. Ba, Zr, U), >80% crystallization required to generate Suite 1 compositions. Similar amount to experimentally-determined degree required to generate liquids with similar major element characteristics to a COM from a tholeiitic parent (Whitaker et al. 2008)	Similar Sr concentrations between the olivine tholeiite and HRT/COM suites and absence of an Eu anomaly in suites 1 and 3 which would be expected from plagioclase dominated fractionation
Crustal assimilation	Elevated $^{87}\text{Sr}/^{86}\text{Sr}$ isotopic ratios in suites 1 and 3, and particularly in published data from the Craters of the Moon lava field (Leeman 1976; Putirka et al. 2009)	Absence of a viable assimilant in databases with high Ba, Zr and FeO, moderate K_2O and Sr, and with a flat REE pattern. Comparable $^{87}\text{Sr}/^{86}\text{Sr}$ between the different mafic suites and lack of a strong relationship between $^{87}\text{Sr}/^{86}\text{Sr}$ and $1/\text{Sr}$, which would be expected with assimilation
Depth of melting	Elevated HREE concentrations in the suites 1 and 3 apparently rules out residual garnet in the source	Similar $(\text{Gd}/\text{Yb})_N$ between the different suites indicating similar depths of equilibration for suite 1 and 3 melts to the olivine tholeiites that equilibrated with spinel lherzolite lithospheric mantle (Leeman et al. 2009)
Degree of melting	Incompatible element enrichment favours small degree of partial melting	Negative correlation between Nb/Zr and Ba/Nb in Suite 1 samples. Different degrees of partial melting would generate a positive trend due to relative degrees of incompatibility ($\text{Ba} > \text{Nb} > \text{Zr}$).
Aqueous fluid enrichment	Enrichment of LILE and mechanism invoked for Eocene regional volcanism due to dehydration of underlying Farallon slab (Feeley 2003)	Enrichment in aqueous fluid immobile HFSE (e.g. Zr, Ti) in suites 1 and 3, resulting in similar LILE/HFSE ratios in the mafic suites, and lack of significant K_2O enrichment in suites 1 and 3
Hydrous melt enrichment	Enrichment in LILE and HFSE in suites 1 and 3, and parallel REE patterns between the mafic suites, with enrichment in all REE aided by hydrous melts (Tsay et al. 2014)	Localised nature of enrichment and significantly different characteristics to contemporaneously derived aqueous fluids

1348 Table 4: Summary of the possible end-member processes for generation of the parental melt to the Suite 1 HRT mafics, together with respective evidence
 1349 for or against the relevant process. See text for further discussion.

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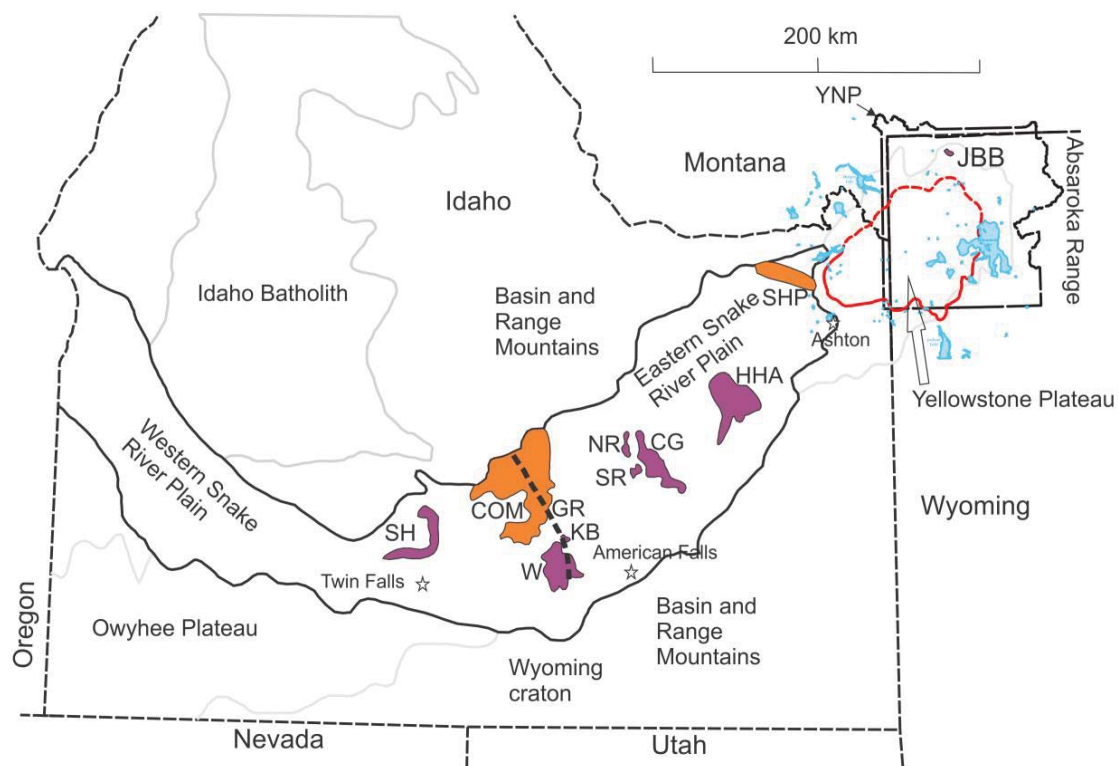


Figure 1



Figure 2

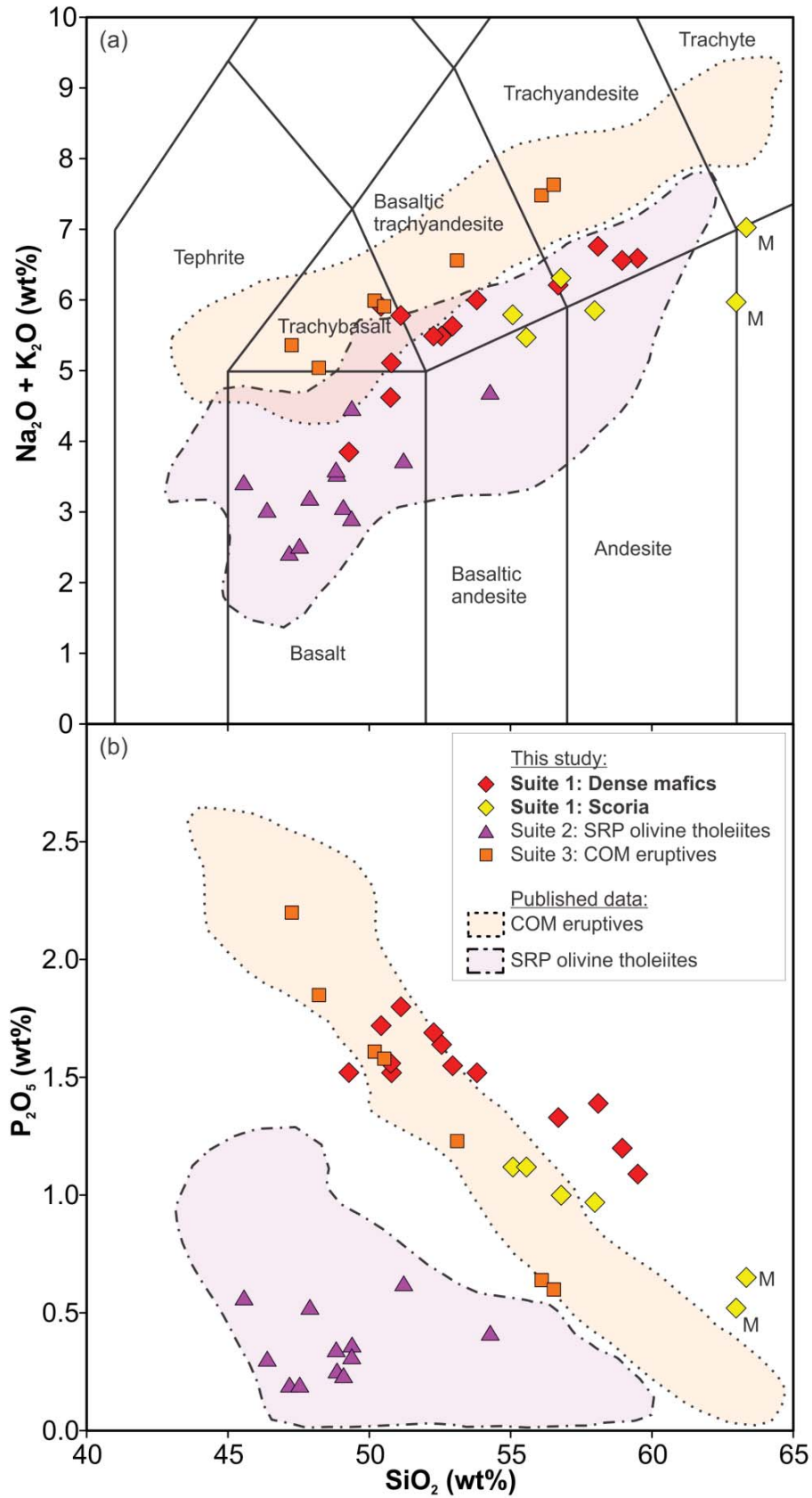


Figure 3

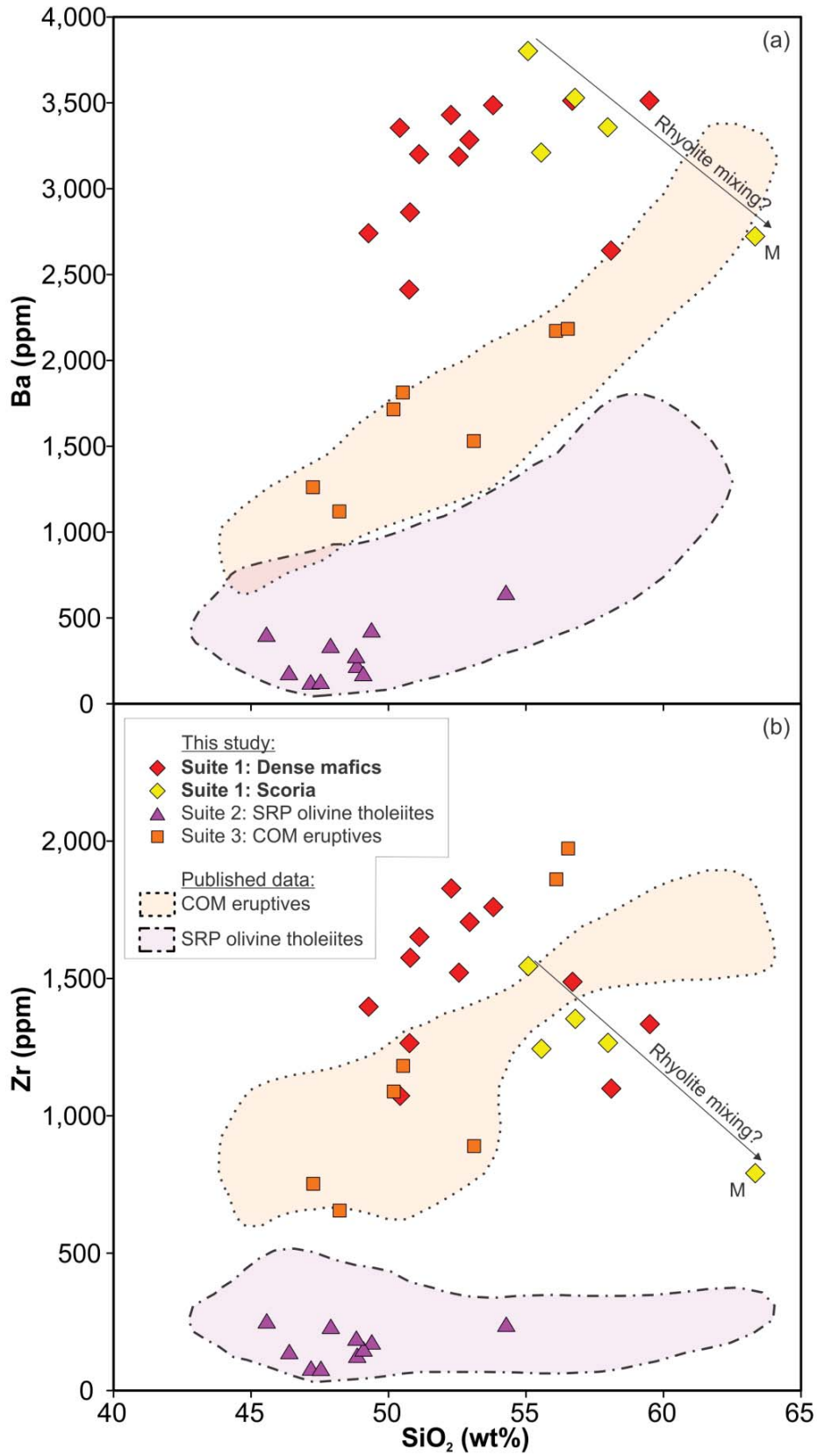


Figure 4

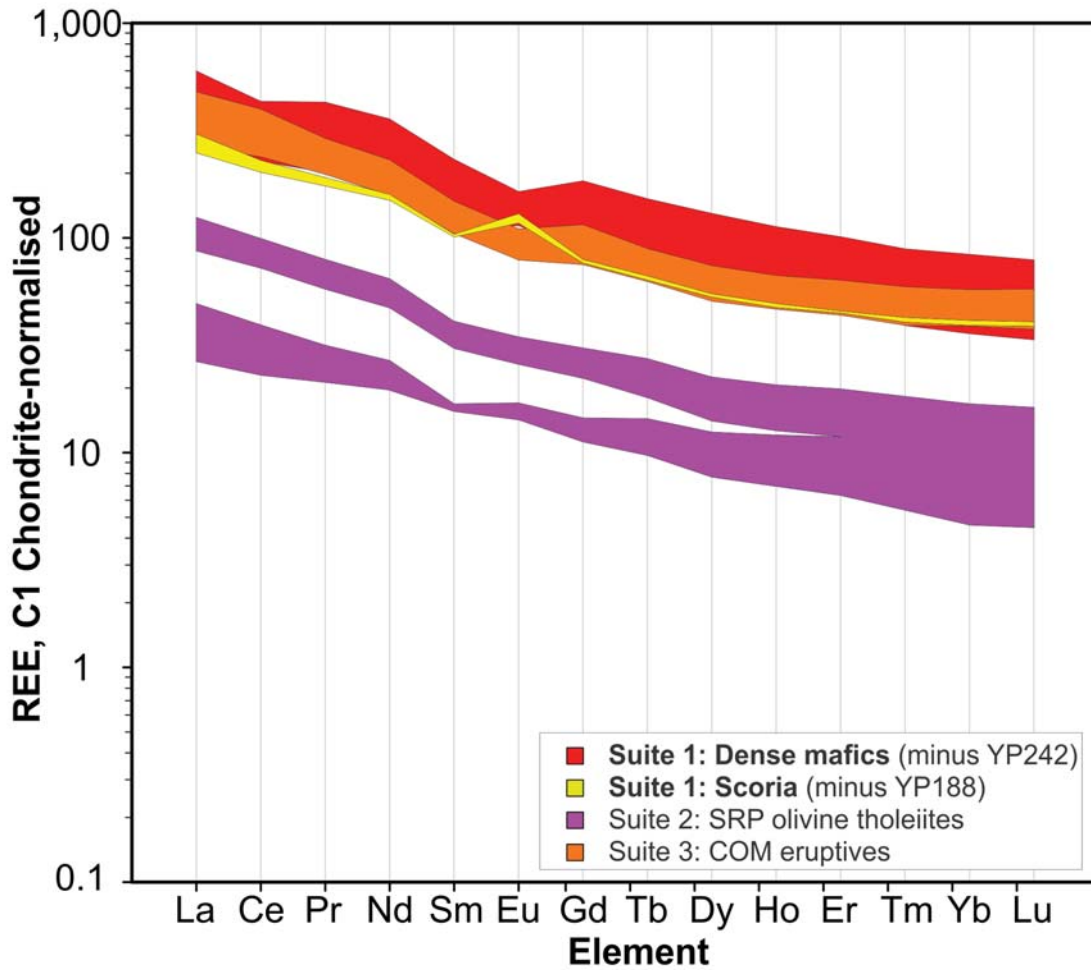


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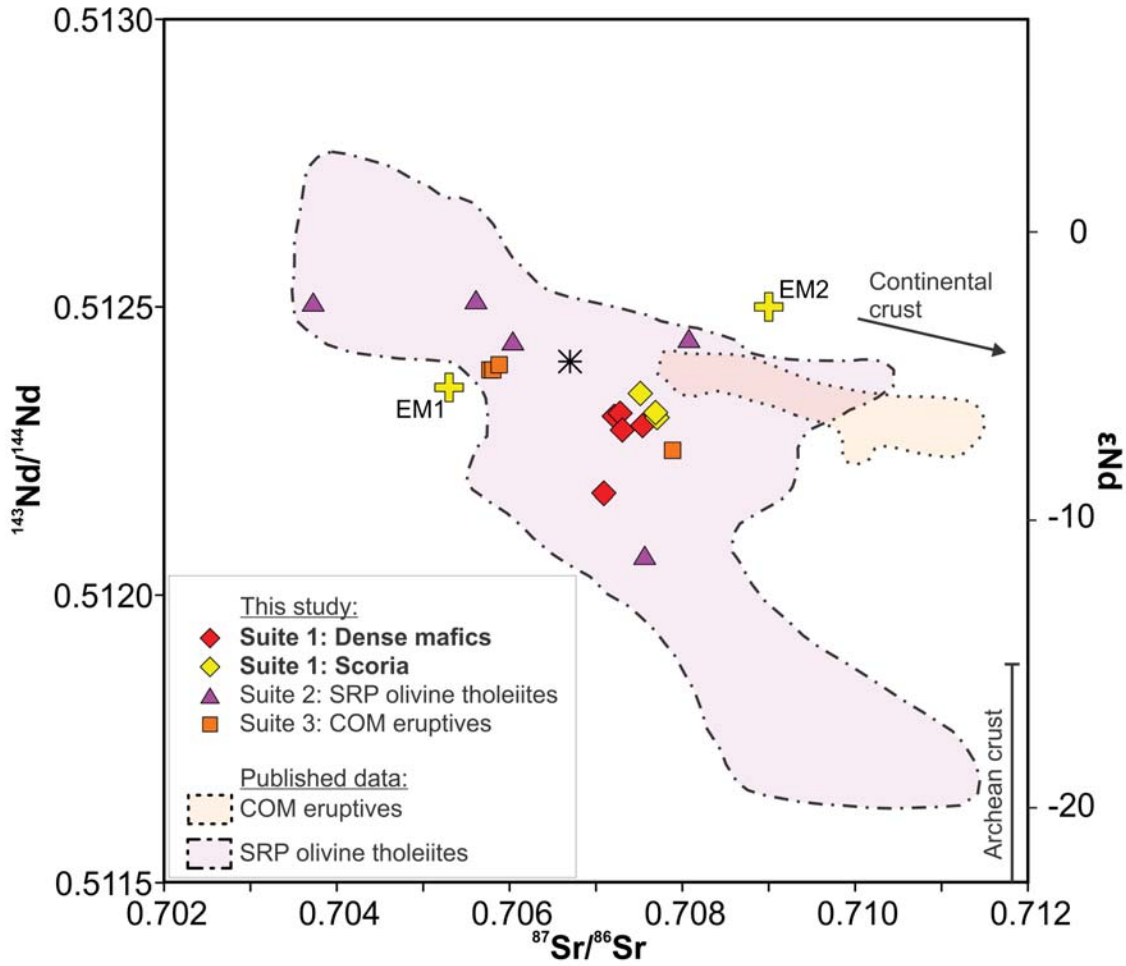


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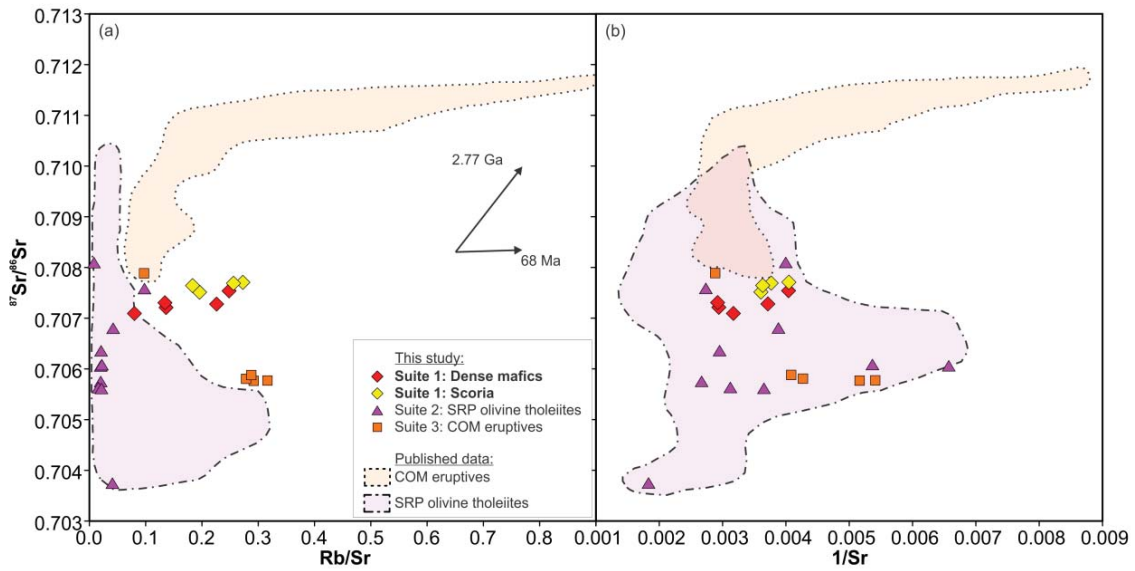


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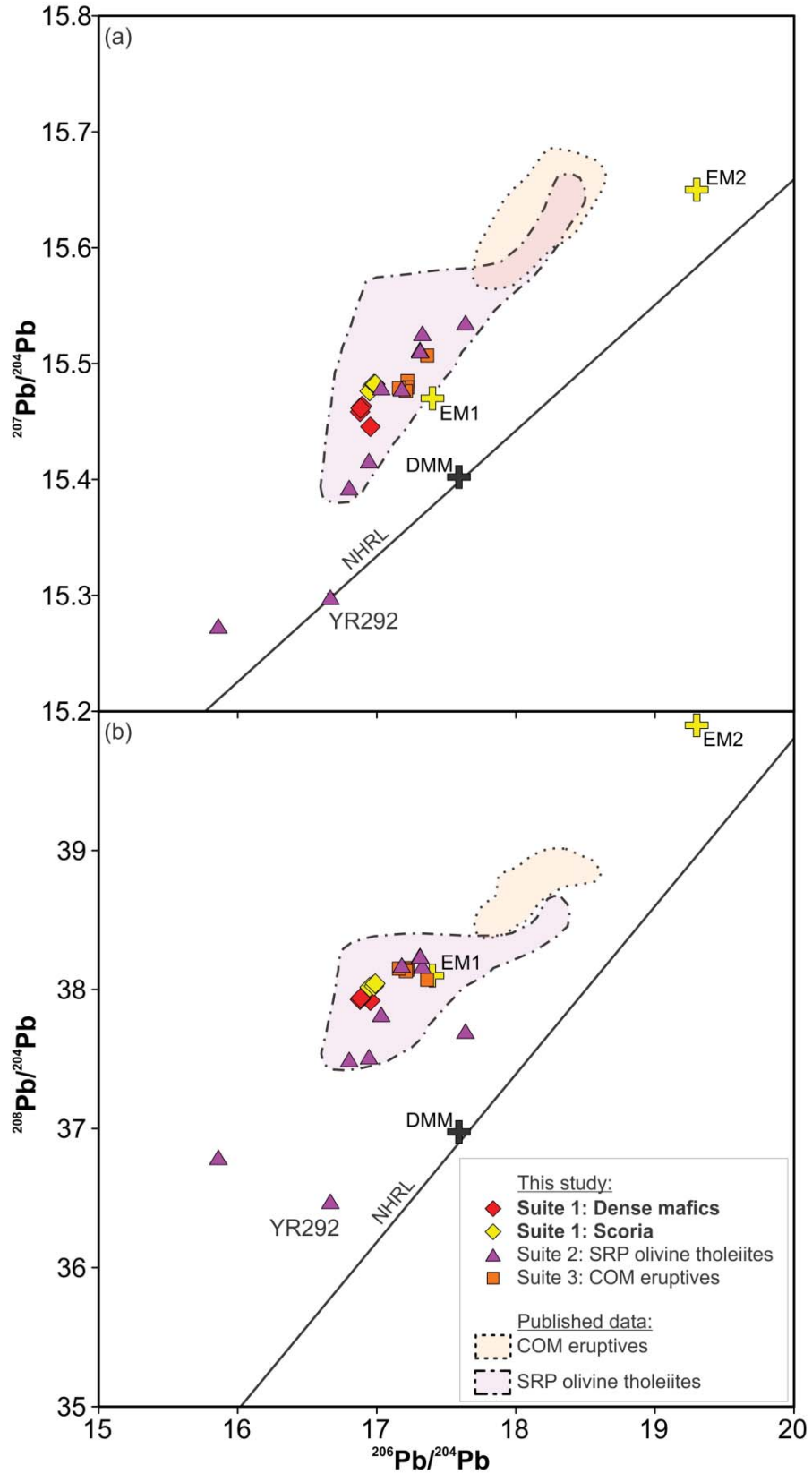


Figure 8

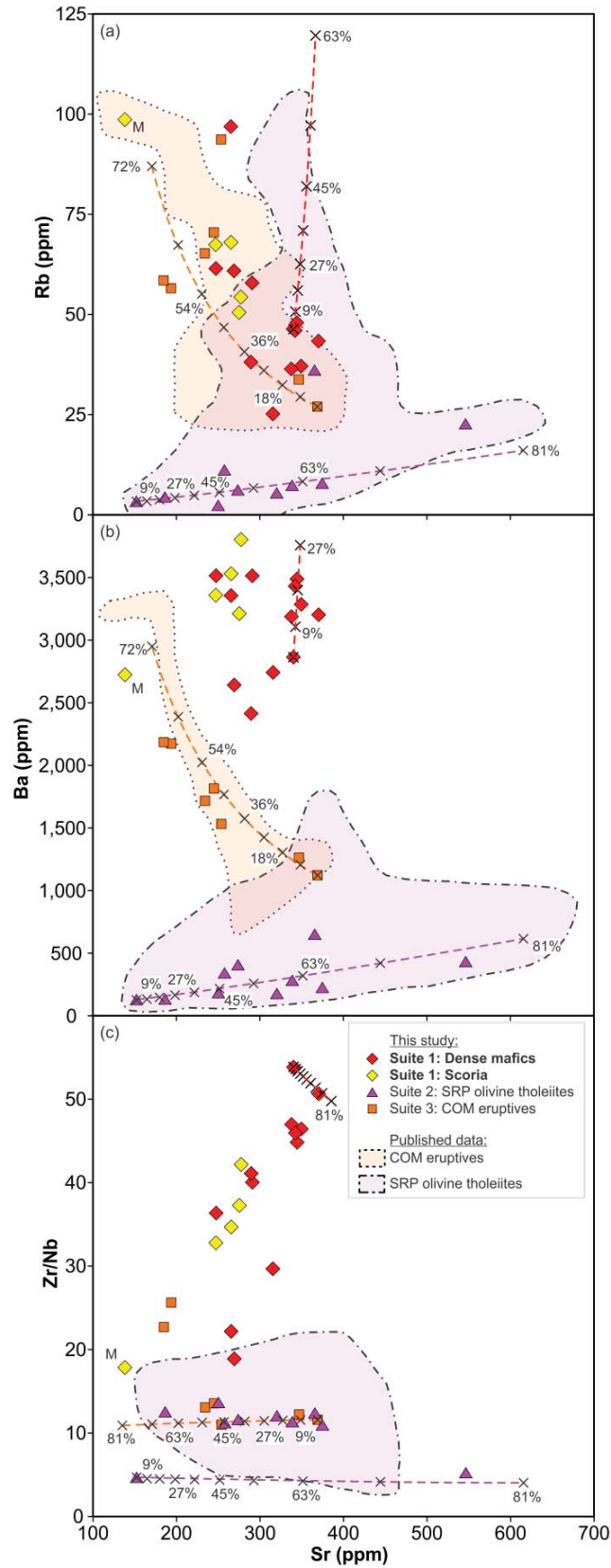


Figure 9

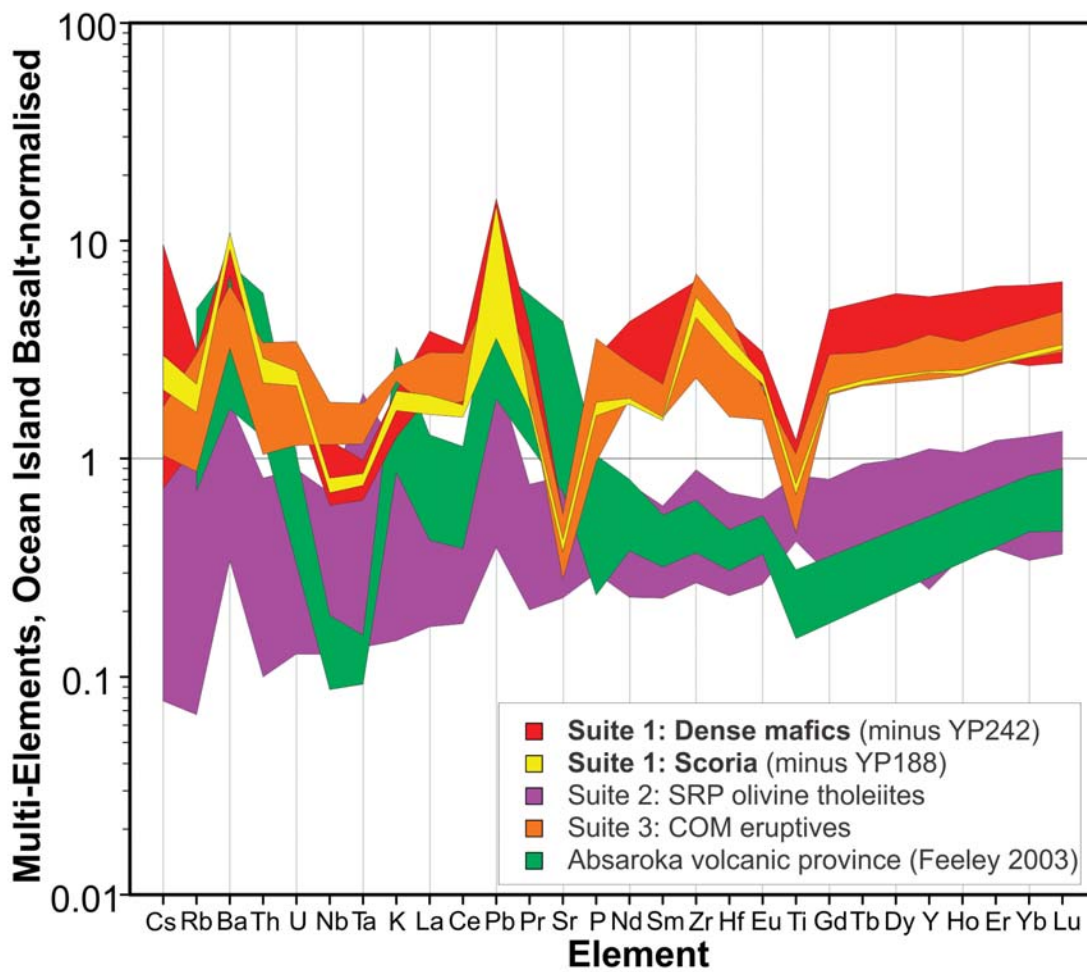


Figure 10

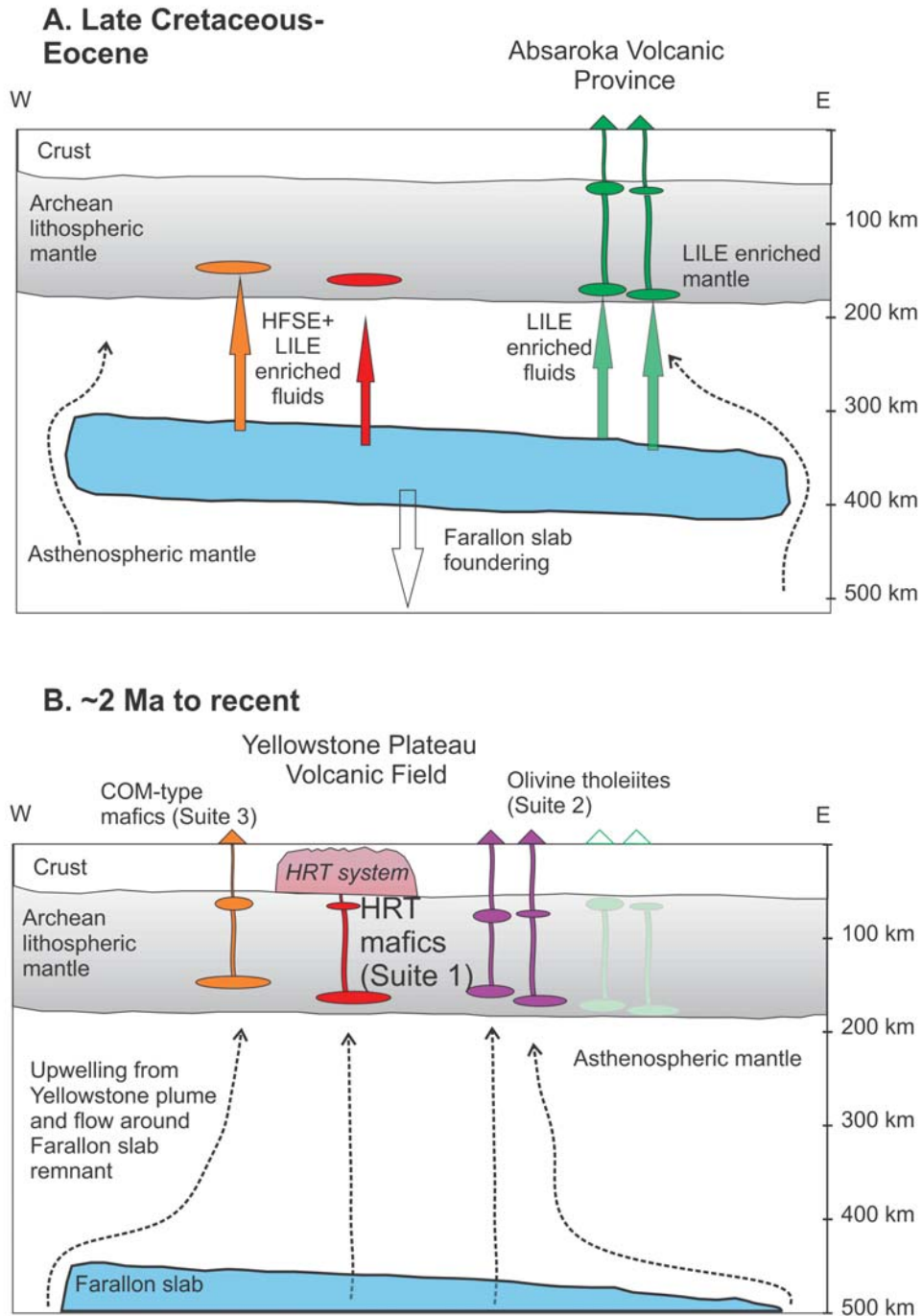


Figure 11