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Testing the Recycled Gabbro Hypothesis for the Origin of "Ghost Plagioclase" Melt Signatures Using ⁸⁷Sr/⁸⁶Sr of Individual Olivine-Hosted Melt Inclusions from Hawai'i

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- 13

14 Key Points:

- Hawaiian melt inclusions are variable in ⁸⁷Sr/⁸⁶Sr, but are within the ⁸⁷Sr/⁸⁶Sr range of the lavas from their host volcano.
- Mauna Loa melt inclusions with high (Sr/Ce)_N (ghost plagioclase) signatures have the same ⁸⁷Sr/⁸⁶Sr as inclusions with normal (Sr/Ce)_N.
- Diffusive interaction between rising melt and Hawaiian gabbros can produce the ghost plagioclase signature in Mauna Loa melt inclusions.
- 21

22 Abstract

23 Melt inclusions with large, positive Sr anomalies have been described in multiple tectonic

24 settings, and the origins of this unusual geochemical feature are debated. Three origins have been

- 25 proposed involving plagioclase as the source of the elevated Sr: (i) direct assimilation of
- 26 plagioclase-rich lithologies, (ii) recycled lower oceanic gabbro in the mantle source, and (iii)
- 27 shallow-level diffusive interaction between present day lower oceanic crust (i.e., plagioclase-
- bearing lithologies) and the percolating melt. A "ghost plagioclase" signature (i.e., a large,
 positive Sr anomaly without associated high Al₂O₃) is present in melt inclusions from Mauna
- 30 Loa. We present new ⁸⁷Sr/⁸⁶Sr measurements of individual olivine-hosted melt inclusions from
- 31 three Hawaiian volcanoes, Mauna Loa, Loihi, and Koolau. The dataset includes a Mauna Loa
- melt inclusion with the highest reported Sr anomaly (or highest (Sr/Ce)_N, which is 7.2) for
- 33 Hawai'i. All melt inclusions have 87 Sr/ 86 Sr values within the range reported previously for the
- 34 lavas from each volcano. Critically, the 87 Sr/ 86 Sr of the high (Sr/Ce)_N melt inclusion lies within
- 35 the narrow range of Mauna Loa melts that lack high $(Sr/Ce)_N$ signatures. To explain the high
- 36 (Sr/Ce)_N ratio of the ghost plagioclase signature using an ancient recycled gabbro would
- 37 therefore require the gabbro-infused mantle source to evolve, by chance, to have the same
- ⁸⁷Sr/⁸⁶Sr as the source of the Mauna Loa melts that lack a recycled gabbro (ghost plagioclase)
- 39 signature. Alternatively, shallow-level diffusive interactions between Mauna Loa plagioclase-
- 40 rich cumulates and a percolating mantle-derived melt provides a simpler explanation for the
- $41 \qquad \text{presence of the high } (Sr/Ce)_N \text{ Mauna Loa melts.}$

42 **1. Introduction**

43 Melt inclusions, encapsulated within growing magmatic phenocrysts at depth, provide 44 "snapshots" of melt evolution that may not be preserved in whole rock compositions. One 45 geochemical signature found in melt inclusions is high Sr concentrations relative to trace 46 elements of similar incompatibility (such as Ce, Nd, and Sm). The magnitude of the Sr anomaly 47 can be quantified with the ratio $(Sr/Ce)_N$ (where N signifies normalization to primitive mantle; 48 McDonough & Sun, 1995). Melt inclusions with high (Sr/Ce)_N ratios are significant because the 49 Sr anomaly found within these inclusions suggest plagioclase was involved in melt petrogenesis. 50 However, plagioclase accumulation, or assimilation, is not always obvious in the major element 51 chemistry of these melt inclusions (Danyushevsky et al., 2003, 2004; Gurenko & Chaussidon, 52 1995; Michael et al., 2002; Peterson et al., 2014; Saal et al., 2007; Slater et al., 2001; Sobolev et 53 al., 2000).

54 Melt inclusions with high (Sr/Ce)_N have been described from samples taken from mid-55 ocean ridges (MORs; e.g., Danyushevsky et al., 2003, 2004; Michael et al., 2002) and hotspot 56 lavas erupted at ocean islands such as Galapagos (Peterson et al., 2014), Hawai'i (Sobolev et al., 57 2000, 2011), and Iceland (e.g., Gurenko & Chaussidon, 1995; Maclennan, 2008; Slater et al., 58 2001). A common geologic material with high $(Sr/Ce)_N$ ratios is cumulate gabbro, which can be 59 a plagioclase-rich lithology. Its abundance in the deep oceanic crust represents the intrusive part 60 of the MOR sequence, and makes gabbro an obvious candidate for imparting large positive Sr 61 anomalies on percolating melts at MOR and ocean island settings (Peterson et al., 2014; Saal et 62 al., 2007; Sobolev et al., 2000; Yaxley & Sobolev, 2007). Plagioclase likely plays a central role 63 in generating large positive Sr anomalies because Sr is compatible in plagioclase compared to 64 elements of similar behavior (Sm, Nd) during peridotite melting and subsequent differentiation in mafic magmatic systems. 65

Melts with large, positive Sr anomalies signal interaction with plagioclase-rich lithologies like gabbro. Paradoxically, a subset of these high (Sr/Ce)_N melt inclusions lack coexisting enrichments in major elements associated with plagioclase (e.g., Al), giving rise to the concept of "ghost plagioclase" signatures in rare melt inclusions trapped in magmatic phenocrysts (Sobolev et al., 2000). Three hypotheses invoked to explain the rare occurrences of large, positive Sr anomalies in melt inclusions include: (i) direct assimilation of gabbro by upwelling melts (Danyushevsky et al., 2003), (ii) melts of recycled lower oceanic (gabbroic) crust in the mantle

- source that are hybridized with peridotite and trapped as melt inclusions in growing magmatic
- 74 phenocrysts (Chauvel & Hémond, 2000; Hofmann & Jochum, 1996; Sobolev et al., 2000;
- 75 Yaxley & Sobolev, 2007), and (iii) diffusive interaction between percolating melt and a

76 plagioclase-rich crystal mush (Peterson et al., 2014; Saal et al., 2007).

Direct assimilation has been ruled out to explain Sr anomalies in many oceanic hotspot
melt inclusions from Hawai'i (Sobolev et al., 2000) and Galapagos (Peterson et al., 2014)
because the ghost plagioclase melt inclusions have large, positive Sr anomalies that do not have
corresponding elevated Al₂O₃, as would be expected for direct assimilation of Al-rich
plagioclase (a.g. Peterson et al., 2014; Sobolev et al., 2000)

81 plagioclase (e.g., Peterson et al., 2014; Sobolev et al., 2000).

82 Regarding the second hypothesis, melts of recycled gabbroic lower oceanic crust may 83 generate trace element chemistry similar to many high (Sr/Ce)_N inclusions from ocean islands. 84 The recycled gabbro model suggests eclogite, metamorphosed gabbroic material, would partially 85 melt earlier (deeper) than surrounding peridotite in an upwelling mantle plume (Sobolev et al., 86 2000; Yaxley & Sobolev, 2007). The melt of the eclogite is silicic (e.g., dacitic) and thus would 87 react with surrounding peridotite, producing garnet-bearing pyroxenites (Yaxley & Green, 1998). 88 The large, positive Sr anomaly is transferred from the silicic melt to the reacted pyroxenite and 89 resides mainly within clinopyroxene. When the reacted pyroxenite partially melts during 90 continued plume upwelling, high-MgO liquids with elevated Sr are generated and mix with 91 normal melts formed by partial melting of peridotite. In the recycled gabbro melting model, the 92 Sr budget in a high (Sr/Ce)_N melt will be dominated by Sr from the gabbro, because the 93 peridotite in the melt source of this model is not assumed to have a large, positive Sr anomaly. As a result, this model predicts that Mauna Loa melt inclusions with large, positive Sr anomalies 94

95 will have essentially the same 87 Sr/ 86 Sr as the recycled gabbro.

96 The third hypothesis predicts that diffusive interaction between percolating melt and 97 plagioclase-rich mush will produce trace element patterns reflecting a gabbro contribution. Due 98 to the slow diffusive nature of major elements (e.g., Al₂O₃), the diffusion model will not generate 99 significant changes in the major element concentration of a melt. In addition, diffusive 100 interaction will cause the high (Sr/Ce)_N melt inclusions to have the same Pb (Peterson et al., 101 2014) and Sr (this study) isotopic composition as the gabbro through which they percolated 102 (because Sr and Pb have high diffusivities in plagioclase). If the melt percolated through and 103 interacted with the gabbro of the ~100 Ma (Müller et al., 2008) MOR crust beneath Hawai'i, melt 104 inclusions will adopt ⁸⁷Sr/⁸⁶Sr of the relatively modern MOR crust. Conversely, if the melt 105 inclusions with high (Sr/Ce)_N result from interaction with gabbroic mush in a Mauna Loa magma chamber, these inclusions will have ⁸⁷Sr/⁸⁶Sr that is indistinguishable from lavas erupted at 106 Mauna Loa. In both cases (i.e., MOR gabbro or Mauna Loa gabbro), the resulting ⁸⁷Sr/⁸⁶Sr ratio 107 108 of high (Sr/Ce)_N melt inclusions is predicted to be indistinguishable from the ⁸⁷Sr/⁸⁶Sr of the 109 gabbro with which the percolating melt interacted.



Figure 1. Map of the Hawaiian Islands showing sample locations (red dots) for the six samples in this study: two from Loihi, three from Mauna Loa, and one from Koolau. The three Mauna Loa samples (red dot on Mauna Loa), and the two Loihi samples (red dot on Loihi) are located very close geographically and are represented by a single symbol. Black dots mark the volcanoes that are used for comparison in this study. Map was created using GeoMapApp (geomapapp.org).

110 This study presents strontium isotopes measured on melt inclusions with high $(Sr/Ce)_N$ to distinguish between the two hypotheses-diffusive interaction between percolating melt and 111 112 modern plagioclase-rich mush versus melts of ancient recycled gabbro-for generating "ghost plagioclase" signatures in melt inclusions. In addition, major, volatile (Cl and S), and trace 113 114 element concentrations are included for individual melt inclusions isolated from six different 115 lavas from three Hawaiian volcanoes: Mauna Loa, Loihi, and Koolau. One of the melt inclusions 116 from Mauna Loa reported here has the highest $(Sr/Ce)_N$ (7.2) ratio ever reported in a Hawaiian 117 lava or melt inclusion.

118 **2. Methods**

Olivine-hosted melt inclusions and glasses from six Hawaiian picrites from Mauna Loa (samples Puu2, Puu7, and ML-2012-27), Loihi (samples 6K490-1 and 6K490-2), and Koolau (sample K89-6; Figure 1) were analyzed. Melt inclusions in this study were not homogenized, as most melt inclusions analyzed were naturally glassy, and characterized for ⁸⁷Sr/⁸⁶Sr and elemental concentrations. However, five inclusions (Puu2-1-2, Puu7-2-15, Puu7-2-92b, K89-6_52, 6K490-1_51) were partially crystalline and were characterized only for ⁸⁷Sr/⁸⁶Sr, and not for elemental concentrations.

The Supplement describes the samples selected, melt inclusion preparation methods,including grinding the olivines into tiny (several hundred microns on a side) cubes to remove all



Figure 2. Method for isolating individual olivine-hosted melt inclusions. (a) Olivines often have multiple melt inclusions and all except for the target melt inclusion must be removed prior to sample leaching and dissolution. First, the target inclusion is exposed, the host olivine is mounted in epoxy, and the melt inclusion is analyzed for major, trace, and volatile elements using *in situ* techniques. (b) After *in situ* analysis, accessory melt inclusions are removed by mounting the olivine grain onto a glass slide using CrystalbondTM. (c) The olivine is then ground on silicon carbide grinding mats until the accessory melt inclusions on one side of the olivine grain are removed. (d) The olivine grain is reoriented to optimally remove remaining accessory melt inclusions, and this process is repeated until only the target melt inclusion is left within a case of its olivine host (e). The "cubed" melt inclusion is then leached and cleaned in ultra-pure acid before dissolution and column chemistry.

- 128 accessory inclusions within an olivine grain (Figure 2), thereby isolating the melt inclusion of
- 129 interest. The Supplement also includes detailed major (electron microprobe), volatile (electron
- 130 microbrobe and ion probe), and trace (laser ablation ICP-MS) element analytical techniques of
- 131 melt inclusions, host olivines, and host pillow glass/tephra. Finally, the Supplement includes
- details of Sr separation chemistry for melt inclusions, and ⁸⁷Sr/⁸⁶Sr analyses by thermal
- 133 ionization mass spectrometry (TIMS) at the University of California, Santa Barbara Isotope
- 134 Geochemistry Facility.

135 **3. Results**

136 The melt inclusions are hosted in olivine crystals ranging from Fo_{80.4} to Fo_{90.1} (Table S1). 137 Melt inclusions span a wide range of sizes, most having maximum major axis lengths between 138 120 and 630 μ m (Figure S3). A high, positive Sr anomaly can be quantified using (Sr/Ce)_N. 139 Below we group the melt inclusions from Mauna Loa (including this study) into two sets: High 140 $(Sr/Ce)_N$ inclusions (with $(Sr/Ce)_N > 2)$, and normal-type melt inclusions (with $(Sr/Ce)_N$ between 141 0.8 and 1.3, and excluding the ultra-calcic melt inclusion). Three Pu'u Wahi melt inclusions 142 (samples Puu7-2-15, Puu7-2-92b, and Puu2-1-2), and one melt inclusion each from Koolau (sample K89-6 52) and Loihi (sample 6K490-1 51), are partially crystalline. Partially crystalline 143 inclusions are not included in major and trace element plots as crystallization can modify major 144 and trace element concentrations, but they are included within the ⁸⁷Sr/⁸⁶Sr dataset. 145 146 In a plot of Al₂O₃ versus MgO (Figure 3), the highest (Sr/Ce)_N melt inclusion from 147 Mauna Loa is shown with new data on normal-type melt inclusions and previously published 148 Mauna Loa melt inclusions (Reinhard et al., 2018; Sobolev et al., 2000, 2011) and Hawaiian 149 whole rock and glass data (Frey et al., 2016; Garcia et al., 1993, 1995, 1998). Frey et al. (2016) compiled a dataset of Hawaiian shield-stage lavas and provides a high-quality dataset of major 150 and trace elements and ⁸⁷Sr/⁸⁶Sr compositions for comparison to the lavas and melt inclusions in 151 152 this study. The melt inclusions presented here, including the highest (Sr/Ce)_N melt inclusion, 153 have Al₂O₃ concentrations similar to the host glasses and plot on, or close to, the array formed by 154 Hawaiian lavas (the exception is a single ultra-calcic melt inclusion which has ~2 wt.% lower 155 concentration of Al₂O₃ than the Hawaiian array at the same MgO content). Gabbroic xenoliths

- 156 from Mauna Loa (sampling the deep recesses of Mauna Loa volcano; Gaffney, 2002), and
- 157 gabbros from the Gabal Gerf ophiolite (Zimmer et al., 1995), plot above the array formed by
- 158 Hawaiian melt inclusions and whole rocks. Critically, the highest $(Sr/Ce)_N$ melt inclusion (ML-
- 159 2012-27_19b) is not shifted off of the Hawaiian array toward the gabbroic compositions.
- 160 Melt inclusions have major element characteristics consistent with Hawaiian whole rock
- 161 and glass data reported in the literature (Figure S4). In general, melt inclusions have lower FeOt 162 (total iron as FeO) compared to their host glass, which is similar to olivine-hosted melt
- 162 (total non as reo) compared to then nost glass, which is similar to onvine-nosted metr 163 inclusions from other localities that have experienced post-entrapment Fe-Mg diffusive exchange
- 164 (Danyushevsky et al., 2000; Gaetani & Watson, 2000; Kent, 2008; Sobolev & Danyushevsky,
- 165 1994). Some of the Mauna Loa inclusions have high CaO relative to the Hawaiian lava array,
- 166 including a single ultra-calcic melt inclusion reported here (sample Puu2-1-39, seen most clearly
- 167 in the panel showing CaO/Al₂O₃ versus MgO in Figure S4f), consistent with previous findings of
- 168 high CaO melts at Hawai'i (Huang & Humayun, 2016; Kogiso & Hirschmann, 2001). While
- some melt inclusions in this study have slightly different major element compositions that trend
- 170 outside of the field defined by Hawaiian lavas (Figure S4), below we show that none of the melt
- 171 inclusions have unusual ⁸⁷Sr/⁸⁶Sr that varies outside of the range found in whole rocks and
- 172 glasses at each respective volcano.
- 173 The Mauna Loa melt inclusions from Pu'u Wahi reported here have a range of (Sr/Ce)_N
- 174 of 0.8–1.5, and the ML-2012-27 Mauna Loa melt inclusions reported here have a $(Sr/Ce)_N$ range
- 175 of 1.0–7.2 (Figures 3 and 4), where $(Sr/Ce)_N$ of 7.2 is the highest value reported for any Mauna
- 176 Loa melt inclusion. Sobolev et al. (2000, 2011) reported (Sr/Ce)_N values up to 5.4, with 11
- 177 inclusions that have $(Sr/Ce)_N > 2$. In a plot of $(Sr/Ce)_N$ versus Al₂O₃ (Figure 3), the highest



Figure 3. High (Sr/Ce)_N Hawaiian melt inclusions and host pillow/tephra glasses compared to Hawaiian lavas, and to Mauna Loa and Gabal Gerf gabbros. (a) In (Sr/Ce)_N versus Al₂O₃ (wt.%) space, the melt inclusion data do not show a positive correlation. A mixing model (line with filled diamonds) is shown between the average Pu'u Wahi melt inclusion composition from this study (Table S10) and the Mauna Loa gabbro with the highest $(Sr/Ce)_N$ (sample 98-65, with $(Sr/Ce)_N = 13.5$) from Gaffney (2002), with 10% increments marked; a second mixing model (line with filled triangles) is shown between the average Pu'u Wahi melt inclusion composition (Table S10) and the Gabal Gerf gabbro with the highest $(Sr/Ce)_N$ (sample GG36, with $(Sr/Ce)_N = 148$) from Zimmer et al. (1995). (b) The same plot as in (a) except expanded to highlight melt inclusions with $(Sr/Ce)_N < 2$. (c) In Al_2O_3 (wt.%) versus MgO (wt.%) space; the mixing model uses the same mixing endmembers as panels (a) and (b). We use the comprehensive Hawaiian dataset (which includes both whole rock and glass analyses) from Frey et al. (2016) for comparison because it provides a compilation of the highest-quality major and trace element data from Hawai'i. The Frey et al. (2016) dataset has been filtered as described in Section S1.5 of the Supplementary Methods. Loa-trend volcanoes (Loihi, Mauna Loa, Koolau, Lanai, and Kahoolawe) and Keatrend volcanoes (Mauna Kea and Kilauea) from the Frey et al. dataset are shown here. The tephra/pillow glass for each sample hosting melt inclusions is also shown for reference. The Pu'u Wahi tephra glass major and trace element concentrations are from Sobolev et al. (2011). Low (Sr/Ce)_N Mauna Loa melt inclusions from Sobolev et al. (2000, 2011) include inclusions with $(Sr/Ce)_N < 2$. Concentrations shown in the figure have been corrected for addition or subtraction of equilibrium olivine so that the melt inclusions are in equilibrium with the host olivine. Only data for glassy melt inclusions are shown. The two ultra-depleted melt (UDM) inclusions from Sobolev et al. (2011) are identified. An outlier with low Al_2O_3 content, which is an ultra-calcic melt inclusion (UCMI) from Pu'u Wahi, is noticeable in all three panels.

178 $(Sr/Ce)_N$ inclusion in this study does not show an anomalous Al₂O₃ concentration relative to the



Figure 4. $(Sr/Ce)_N$ versus $(La/Sm)_N$ plot shows the diffusive interaction model results (black crosses) for Mauna Loa based on the Peterson et al. (2014) model, and the numbers next to the data points represent the number of years of diffusion. Mixing lines are shown between the ML-2012-27 host tephra composition (Table S8) and compositions generated at 1, 20, and 400 years of diffusive interaction. Average 2σ error bars are shown in the top right corner of plot.

179 melt inclusions with lower $(Sr/Ce)_N$.

180 In a plot of (Sr/Ce)_N versus (La/Sm)_N (Figure 4), the high (Sr/Ce)_N inclusions, all of 181 which are from Mauna Loa, tend to have lower (La/Sm)_N ratios than normal-type melt inclusions and Hawaiian lavas, which is consistent with prior observations at Hawai'i (Sobolev et al., 2000) 182 183 and Galapagos (Peterson et al., 2014). Excluding the ultra-depleted melt (UDM) inclusions from 184 Sobolev et al. (2011), the melt inclusion with the highest $(Sr/Ce)_N$ has the lowest $(La/Sm)_N$ 185 observed in Hawaiian melts. Like other high $(Sr/Ce)_N$ inclusions from Hawai'i, the high $(Sr/Ce)_N$ 186 inclusion is depleted in light rare earth elements (REEs) relative to middle REEs, an observation that has been made before at Hawai'i (e.g., Sobolev et al., 2000) and Galapagos (Peterson et al., 187 2014). Notably, all Hawaiian melt inclusions with high (Sr/Ce)_N in this study are hosted in 188 189 Mauna Loa lavas; none of the Loihi or Koolau samples have large positive (Sr/Ce)_N, but we note 190 that fewer Loihi and Koolau melt inclusions have been characterized.

191 Mauna Loa melt inclusions reported here have some variability in trace element ratios. 192 Primitive mantle (McDonough & Sun, 1995) normalized trace element patterns (spidergrams) of 193 all glassy melt inclusions reported in this study are shown together with spidergrams of the host 194 glass or tephra (Figure 5a–d). The highest $(Sr/Ce)_N$ inclusion exhibits the most significant 195 deviation in pattern shape (with overall depletion in incompatible trace element abundances 196 compared to the other Mauna Loa inclusions). For comparison, the Mauna Loa gabbro with the 197 highest (Sr/Ce)_N (sample 98-65; Gaffney, 2002) is plotted against Mauna Loa sample ML-2012-198 27 (Figure 5). The highest (Sr/Ce)_N inclusion has a similar spidergram to high (Sr/Ce)_N 199 inclusions from Sobolev et al. (2000, 2011), but different compared to the average normal Pu'u Wahi melt inclusion (Figure S5). Pu'u Wahi melt inclusions from Mauna Loa reported in 200 201 Reinhard et al. (2018) are plotted alongside this study's Pu'u Wahi inclusions, and they exhibit 202 less variability in spidergram patterns than the Mauna Loa ML-2012-27 inclusions. Similarly, the spidergrams for the Loihi and Koolau inclusions show relatively little variability and are similarto the host glass.

Figure 5 also shows that the Mauna Loa gabbros from Gaffney (2002) contain positive Pb anomalies whereas the Mauna Loa melt inclusions have negative Pb anomalies. Sulfides are observed in Hawaiian lavas (Stone & Fleet, 1991). If sulfides are responsible for the positive Pb anomalies in Mauna Loa gabbros (which are cumulates of Mauna Loa melts), this can be taken as evidence for sulfide fractionation from Hawaiian magmas at depth as being the mechanism generating the negative Pb anomalies in high (Sr/Ce)_N inclusions.

211 The ⁸⁷Sr/⁸⁶Sr of melt inclusions from the five lavas examined here (Table 1) are similar

to, or slightly larger, than the range observed for their respective host volcano. The ⁸⁷Sr/⁸⁶Sr

213 range for all of the Mauna Loa inclusions reported here is relatively small, ranging from

0.703700 to 0.703986 (⁸⁷Sr/⁸⁶Sr range of 406 ppm, Figure 5). The inclusions fall within the range

215 reported for Mauna Loa lavas (0.703599–0.703967; Hauri et al., 1996; Kurz et al., 1995). The

216 highest (Sr/Ce)_N melt inclusion (ML-2012-27_19b) has an 87 Sr/ 86 Sr of 0.703828 ± 0.000045, and

217 this inclusion, which also has the lowest $(La/Sm)_N$ in this study, has indistinguishable ${}^{87}Sr/{}^{86}Sr$

218 from the other Mauna Loa melt inclusions from the same lava (including Mauna Loa melt $(1 + 1)^{-1}$

219 inclusions with low $(Sr/Ce)_N$ (Figure 6)). The Loihi melt inclusions (from two different lavas)

have 87 Sr/ 86 Sr ratios ranging from 0.70328 to 0.70367 (87 Sr/ 86 Sr range of 555 ppm), which

extends the range of 87 Sr/ 86 Sr identified for Loihi lavas (0.70344 to 0.70378; Bennett et al., 1996;

Garcia et al., 1995) to slightly lower 87 Sr/ 86 Sr. The Koolau melt inclusions (all from a single lava

sample) exhibit the largest range of ⁸⁷Sr/⁸⁶Sr among melt inclusions in this

	- ()	- / .	(- (-)	is and the Blass o				ppm change in ⁸⁷ Sr/ ⁸⁶ Sr
Sample	Sr (ppm)	Ce (ppm)	(Sr/Ce) _N	Total Sr (ng)	[Sr] _{sample} /[Sr] _{blank}	°′Sr/°°Sr*	2σ (std. err.)	ratio from blank correction ^b
Mauna Loa - Pu'u	Wahi (Puu2 &	& Puu7)						
Puu2-1-2	-	-	-	2.5	222	0.703772	0.000032	46
Puu2-1-9	270	20.6	1.11	15.5	292	0.703800	0.000021	35
Puu2-1-18a	337	26.7	1.06	2.8	53	0.703885	0.000049	191
Puu2-1-19	338	24.1	1.18	1.1	97	0.703899	0.000024	104
Puu2-1-39	215	15.8	1.14	7.3	1038	0.703935	0.000010	10
Puu7-1-20	438	24.9	1.48	1.2	177	0.703867	0.000034	57
Puu7-2-15	-	-	-	3.7	527	0.703845	0.000018	19
Puu7-2-92b	-	-	-	0.9	78	0.703917	0.000065	129
Puu7-2-80	324	20.3	1.34	9.2	817	0.703953	0.000016	12
Puu7-2-19	325	35.2	0.78	18.9	2690	0.703838	0.000008	4
Puu7-1-29	330	25.2	1.10	1.2	168	0.703804	0.000038	61
Puu7-3-19a	314	21.9	1.21	17.5	329	0.703916	0.000009	31
Puu7-3-31	265	19.1	1.16	28.8	542	0.703700	0.000023	19
Puu7-1-14	279	28.0	0.84	3.6	67	0.703947	0.000021	149
Puu7-1-30	325	31.6	0.87	2.5	46	0.703896	0.000013	218
Puu7-1-36	338	25.9	1.10	23.5	441	0.703770	0.000014	23
Puu7-2-62	334	24.3	1.16	3.1	444	0.703887	0.000030	23
Puu7-1-41	330	22.8	1.21	7.7	682	0.703977	0.000023	15
Puu7-2-6	327	25.8	1.07	9.8	1400	0.703811	0.000010	7
Puu7-2-40a	280	19.2	1.22	9.6	855	0.703770	0.000021	12
Puu7-2-42	336	22.9	1.24	5.3	469	0.703986	0.000029	21
Puu7-2-7a	311	22.5	1.16	3.1	445	0.703942	0.000040	23
Puu7-1-10	355	26.3	1.14	13.5	1923	0.703949	0.00008	5
Puu7-2-52	345	23.0	1.26	5.0	447	0.703882	0.000035	23
host glass ^c	319	25.9	1.04			0.703912	0.000008	
Mauna Loa ML-20	12-27							
ML-2012-27_2	283	15.3	1.56	10.0	1027	0.703881	0.000013	10
ML-2012-27_5a	457	38.1	1.01	10.2	1042	0.704010	0.000016	10
ML-2012-27_7	441	34.1	1.09	2.9	300	0.703887	0.000016	34
ML-2012-27_18a	426	35.8	1.00	0.7	69	0.704039	0.000030	144
ML-2012-27_21a	437	34.4	1.07	1.8	181	0.704009	0.000045	55
ML-2012-27_25	411	30.2	1.15	2.1	215	0.703985	0.000021	46
ML-2012-27_19b	786	9.1	7.24	1.4	74	0.703828	0.000045	137
host glass	344	27.6	1.05			0.703909	0.000006	
Koolau - K89-6								
K89-6_52	-	-	-	3.5	363	0.704401	0.000031	26
K89-6_53	260	16.2	1.35	0.5	25	0.703654	0.000079	416
K89-6_55	244	14.1	1.45	10.5	1495	0.703960	0.000027	/
K89-6_65	327	20.0	1.38	1.4	86	0.703864	0.000026	118
K89-6_76a	267	17.6	1.27	0.6	65	0.703906	0.000041	155
	500	23.5	1.52			0.703905	0.000000	
CK400 1 51				0.0	50	0 702284	0.000003	711
6K49U-1_51	-	-	-	0.9	5Z 21	0.703284	0.000093	211
host glass	439 460	42.1	0.92	0.5	51	0.703665 0.703586	0.000090 0.000006	222
- Loihi - 6K490-2								
6K490-2 79-	507	44 5	0.96	1 1	63	0 703363	0 000033	173
host glass	412	35.0	0.99	1.1	00	0.703574	0.000006	1,5

Table 1. ⁸⁷Sr/⁸⁶Sr measurements made on melt inclusions and the glass of the host lava in this study.

Note: The Sr and Ce fractionation-corrected concentrations are reported here, but not for partially crystalline inclusions.

^a ⁸⁷Sr/⁸⁶Sr values have been blank corrected and corrected for the offset between the preferred (0.710240) and the measured ⁸⁷Sr/⁸⁶Sr of NBS987.

^b The change in ⁸⁷Sr/⁸⁶Sr ratio due to the blank correction (ppm) is calculated as (⁸⁷Sr/⁸⁶Sr_{meas-norm987}-⁸⁷Sr/⁸⁶Sr_{blank-corrected})/⁸⁷Sr/⁸⁶Sr_{meas-norm987}*10⁶.

^c Sr concentration and ⁸⁷Sr/⁸⁶Sr of Pu'u Wahi glass is from Reinhard et al. (2018).

224



Figure 5. Primitive mantle normalized trace element patterns, or spidergrams, (left panels) and corresponding 87 Sr/ 86 Sr values (right panels) of all Hawaiian melt inclusions and host pillow/tephra glass in this study (note that 5 melt inclusions—Puu2-1-2, Puu7-2-15, Puu7-2-92b, K89-6 52, 6K490-1 51—are crystalline, so their spidergrams are not shown, but ⁸⁷Sr/⁸⁶Sr are). Concentrations shown in the figure have been corrected for addition or subtraction of equilibrium olivine so that the melt inclusions are in equilibrium with the host olivine. (a-b) Spidergrams for melt inclusions and host tephra from the three Mauna Loa samples-ML-2012-27 and the Pu'u Wahi samples (Puu2 and Puu7)—are shown together with a spidergram from a Mauna Loa gabbro with the highest $(Sr/Ce)_N$ (sample 98-65 from Gaffney (2002)); spidergrams for melt inclusions from Pu'u Wahi from Reinhard et al. (2018) are also shown. The Pu'u Wahi tephra trace element concentrations are from Sobolev et al. (2011). (c) Spidergrams of Koolau melt inclusions from sample K89-6 with the host glass. (d) Spidergrams of Loihi melt inclusions from samples 6K490-1 and 6K490-2 are shown with the two respective host glasses. (e-f) The ⁸⁷Sr/⁸⁶Sr of the host pillow/tephra glass is shown as a straight line and dashed lines represent the 2σ (2SE) error bars. Panel (f) compares the ⁸⁷Sr/86Sr of melt inclusions from Mauna Loa samples "Puu2" and "Puu7" analyzed here (orange symbols) and by Reinhard et al. (2018) (grey symbols). The ⁸⁷Sr/⁸⁶Sr results (generated in two different labs, UCSB and Vrije University, respectively) are remarkably consistent (the ⁸⁷Sr/⁸⁶Sr of the host tephra glass is shown as a straight line and was analyzed by Reinhard et al. (2018)). Partially crystalline inclusions are indicated in panels (f-h), and spidergrams are not available for these inclusions. Note that the host pillow glass ⁸⁷Sr/⁸⁶Sr ratios for the two Loihi lavas—6K490-1 and 6K490-2—are indistinguishable in panel (h).



Figure 6. Important trace element ratios versus ⁸⁷**Sr**/⁸⁶**Sr. (a–b)** (Sr/Ce)_N versus ⁸⁷Sr/⁸⁶Sr. (c) (La/Sm)_N versus ⁸⁷Sr/⁸⁶Sr. (d) Cl/Nb versus ⁸⁷Sr/⁸⁶Sr. Note that melt inclusions in Sobolev et al. (2011) with ⁸⁷Sr/⁸⁶Sr are not shown as described in Section 3, and melt inclusions from Sobolev et al. (2000), including high (Sr/Ce)_N inclusions, do not have ⁸⁷Sr/⁸⁶Sr. Fields for individual Hawaiian volcanoes are shown in (b) and (c) and the data are from Frey et al. (2016), which were filtered to exclude altered and differentiated lavas, as described in the Supplementary Methods Section S1.5, and thus the full range of ⁸⁷Sr/⁸⁶Sr for the volcanoes is not necessarily represented in this figure. Each plot on the left-hand side shows the diffusive interaction model results (black crosses) for Mauna Loa based on the Peterson et al. (2014) model and the numbers next to the crosses represent the number of years of diffusion. Mixing lines (grey lines with small grey diamonds representing increments of 10%) are shown between the ML-2012-27 host tephra composition and diffusion model results of 1 year and 20 years. For panel (d), note that samples ML-2012-27 and K89-6 do not have published Cl concentrations and Cl was not analyzed in the glasses in this study. Pu'u Wahi glass Cl concentration is from Sobolev et al. (2011). Cl concentrations for Loihi samples 6K490-1 and 6K490-2 are from Pietruszka et al. (2011). All panels show average 2 σ error bars in the bottom right corner of plots.

227 study $(0.703654 \pm 0.000079 \text{ to } 0.704401 \pm 0.000031, 1062 \text{ ppm total variability})$, which is encompassed by the range of ⁸⁷Sr/⁸⁶Sr in Koolau lavas (0.703487 to 0.70451; Roden et al., 1994; 228 Tanaka et al., 2002). While there is ⁸⁷Sr/⁸⁶Sr heterogeneity in the Loihi and Koolau inclusions 229 230 examined here, the heterogeneity is not surprising given heterogeneity observed in olivine-hosted 231 melt inclusions at other OIB localities (Jackson & Hart, 2006; Harlou et al., 2009; Reinhard et al., 2016, 2018). What is surprising is the remarkable ⁸⁷Sr/⁸⁶Sr homogeneity across the different 232 233 Mauna Loa melt inclusions examined here, in spite of extreme trace element variability (e.g., 234 $(Sr/Ce)_N$). Below we focus the discussion on this unusual ${}^{87}Sr/{}^{86}Sr$ homogeneity. 235 Loihi melt inclusions have higher Cl contents (319 to 606 ppm) than the melt inclusions 236 from Mauna Loa (measured concentration of 10 to 183 ppm; see Table S1) or Koolau (20 to 45 237 ppm) (Figure 6e). Elevated Cl in Loihi melts was observed previously (Kent et al., 1999a, 1999b,

238 2002). Similarly, the three Loihi inclusions have the largest range in Cl/Nb (14 to 30), consistent
239 with the high Cl concentrations observed in Loihi glasses (Kent et al., 1999a, 1999b, 2002). In

240 comparison, Cl/Nb ratios tend to lower values in Koolau melt inclusions (5 to 7) and Mauna Loa

inclusions (4 to 14, excluding a single melt inclusion, sample Puu7-2-7a, with Cl/Nb = 21).

Critically, ⁸⁷Sr/⁸⁶Sr does not covary with Cl/Nb in the melt inclusion suite examined here,
 suggesting seawater-derived materials are not responsible for generating the ⁸⁷Sr/⁸⁶Sr
 heterogeneity (see Sections 4.1 and 4.3 for discussion on the interpretation).

245 Melt inclusions from Pu'u Wahi were previously analyzed for ⁸⁷Sr/⁸⁶Sr by Sobolev et al. 246 (2011) and Reinhard et al. (2018). Sobolev et al. (2011) analyzed a suite of 137 inclusions from 247 Pu'u Wahi using a laser ablation (LA) single-collector ICP-MS technique and reported an 248 extraordinary range in ⁸⁷Sr/⁸⁶Sr, from 0.7021 to 0.7081 (which is a range of ~8500 ppm, or 249 approximately ± 3000 ppm 2σ relative standard deviation, or 2RSD; Figure 7). They showed that 250 the two UDMs identified in Pu'u Wahi inclusions yield the most extreme high ⁸⁷Sr/⁸⁶Sr values 251 $(0.7081 \pm 0.0004;$ two melt inclusions analyzed at least ten times each), and if these UDM inclusions are excluded, the 87 Sr/ 86 Sr in their dataset has an average of 0.7039 ± 0.0019 (2SD) 252 253 (approximately ± 2700 ppm 2RSD). In contrast, the 33 Pu'u Wahi melt inclusions analyzed for 254 87 Sr/ 86 Sr by TIMS in this study and the study of Reinhard et al. (2018) show very little variability 255 (i.e., 0.703700 to 0.703986, or approximately ±200 ppm 2RSD across both TIMS studies). Like Reinhard et al. (2018), we did not identify UDM inclusions, which may partially explain the lack 256 257 of extreme ⁸⁷Sr/⁸⁶Sr in our study (but see below). However, even if the two UDM inclusions are excluded from the Sobolev et al. (2011) dataset, the 2RSD in ⁸⁷Sr/⁸⁶Sr of the remaining 135 258 259 inclusions (i.e., approximately ± 2700 ppm) is still ~13 times greater than the combined 2RSD in 260 87 Sr/ 86 Sr identified in this study and the study of Reinhard et al. (2018) (i.e., approximately ± 200 261 ppm).

In summary, the TIMS 87 Sr/ 86 Sr results for Pu'u Wahi inclusions from two different labs—Vrije University (N = 9 inclusions; Reinhard et al., 2018) and UCSB (N = 24 inclusions; this study)—are in close agreement, but disagree substantially with the reported 87 Sr/ 86 Sr variability in Pu'u Wahi inclusions from Sobolev et al. (2011) (Figure 7). One explanation is that

there is an inherent difference in precision between the two techniques. Rubidium (as well as



Figure 7. ⁸⁷Sr/⁸⁶Sr of Mauna Loa Pu'u Wahi melt inclusions by two different methods. (a) The ⁸⁷Sr/⁸⁶Sr of Pu'u Wahi melt inclusions measured by LA-ICP-MS (Sobolev et al., 2011) and TIMS (Reinhard et al., 2018; this study) are compared to the ⁸⁷Sr/⁸⁶Sr range of Mauna Loa lavas (orange bar = 0.703599–0.703967; Hauri et al., 1996; Kurz et al., 1995) and of the Pu'u Wahi glass (horizontal black line; Reinhard et al., 2018). The ultra-depleted melt (UDM) inclusions from Sobolev et al. (2011) are marked separately. Even when the UDM inclusions are excluded, the 2RSD in ⁸⁷Sr/⁸⁶Sr of the Pu'u Wahi melt inclusions determined by LA-ICP-MS greatly exceeds (by a factor of 13) the 2RSD determined by TIMS. Panel (b) shows only the ⁸⁷Sr/⁸⁶Sr of Pu'u Wahi melt inclusions analyzed by TIMS compared to the Pu'u Wahi matrix glass (horizontal black line with 2SE represented by the dashed lines) analyzed by Reinhard et al. (2018).

- 267 other isobaric interferences, e.g., ⁸⁴Kr, ⁸⁶Kr) limits the precision of ⁸⁷Sr/⁸⁶Sr analyses performed
- by LA-ICP-MS, whereas chemical separation of Sr provides a means to minimize Rb prior to
- analysis by TIMS, and TIMS does not suffer from the other isobaric interferences from the
- 270 carrier gas. We suggest that the difference in Pu'u Wahi melt inclusion ⁸⁷Sr/⁸⁶Sr variability
- determined by LA single-collector ICP-MS (Sobolev et al., 2011) and TIMS (this study;
- 272 Reinhard et al., 2018) is the result of the former technique generating lower-precision analyses of
- a relatively-homogeneous melt inclusion population (i.e., a population with ⁸⁷Sr/⁸⁶Sr variability
- similar to that identified by TIMS). To test this hypothesis, we calculate the average ⁸⁷Sr/⁸⁶Sr and
- 275 2SD of the Pu'u Wahi melt inclusions analyzed by the LA-ICP-MS technique, and compare this



Figure 8. The red histogram displays the resulting model variability in ⁸⁷Sr/⁸⁶Sr if a relatively homogeneous population (i.e., ⁸⁷Sr/⁸⁶Sr variability is approximately ±200 ppm, 2RSD) of melt inclusions, like that found by the TIMS studies of Pu'u Wahi melt inclusions (Reinhard et al., 2018; this study), is modelled using a Monte Carlo method that assigns the melt inclusions larger uncertainties from the distribution of measurement uncertainties associated with the Sobolev et al. (2011) ICP-MS analyses. The red histogram represents the ⁸⁷Sr/⁸⁶Sr of model melt inclusions resulting from 10⁶ iterations of the Monte Carlo simulation. The ⁸⁷Sr/⁸⁶Sr values of the Pu'u Wahi melt inclusions reported by Sobolev et al. (2011) are shown as a blue histogram). In summary, larger measurement uncertainties associated with a relatively homogeneous population of melt inclusions can generate apparent heterogeneity that mimics the extreme ⁸⁷Sr/⁸⁶Sr heterogeneity reported in the Puu Wahi inclusions by the Sobolev et al. (2011).

- to the average ⁸⁷Sr/⁸⁶Sr and 2SD of the Pu'u Wahi melt inclusions analyzed by TIMS. The 276
- average of Sobolev et al.'s (2011) 87 Sr/ 86 Sr measurements is 0.7040 ±0.0021 (2SD, N = 137) 277
- (with the two UDM inclusions), or 0.7039 ± 0.0019 (2SD, N = 135) (without the two UDM 278
- 279 inclusions). These average ⁸⁷Sr/⁸⁶Sr values are indistinguishable from the average ⁸⁷Sr/⁸⁶Sr
- obtained on Pu'u Wahi melt inclusions at the Vrije University TIMS lab (0.703867 ±0.00013, 280
- 2SD, N = 9) and the average 87 Sr/ 86 Sr of melt inclusions from the UCSB TIMS lab reported here 281
- 282 $(0.703873 \pm 0.000152, 2SD, N = 24)$, which are also indistinguishable from the Pu'u Wahi tephra
- glass ⁸⁷Sr/⁸⁶Sr (0.703912±0.000008, 2SE) and the range in ⁸⁷Sr/⁸⁶Sr reported in Mauna Loa 283
- whole rocks (0.70360 to 0.703967; Hauri et al., 1996; Kurz et al., 1995). This result is consistent 284

with the hypothesis that the disagreement between the TIMS ⁸⁷Sr/⁸⁶Sr analyses and the LA-ICP MS ⁸⁷Sr/⁸⁶Sr analyses are a result of the significantly larger errors associated with the Sobolev et
 al. (2011) ⁸⁷Sr/⁸⁶Sr measurements.

288 In order to further test the influence of measurement reproducibility on the total observed ⁸⁷Sr/⁸⁶Sr variability in the Pu'u Wahi melt inclusion suite, we performed a Monte Carlo 289 290 simulation. This Monte Carlo simulation (Figure 8) evaluates the ⁸⁷Sr/⁸⁶Sr variability in Pu'u 291 Wahi melt inclusions that would result if a relatively homogeneous population of Pu'u Wahi 292 melt inclusions (like that found by the TIMS studies) is assigned the larger measurement 293 uncertainties associated with the Sobolev et al. (2011) single-collector ICP-MS method. In the 294 simulation, we first calculate an uncertainty equal to the average 2SD of the 135 Pu'u Wahi melt 295 inclusion measurements (i.e., including only the melt inclusions with at least one replicate 296 analysis) reported by Sobolev et al. (2011), which is ±2919 ppm. This is used to generate a 297 Gaussian distribution, which has a 2RSD of ±2919 ppm, and from this distribution values are 298 drawn randomly in each iteration of the model and then assigned as errors to each of the 33 Pu'u 299 Wahi melt inclusions analyzed by TIMS in this study and Reinhard et al. (2018). The range of 300 ⁸⁷Sr/⁸⁶Sr in the 33 melt inclusions in the model is then calculated, and this process was then repeated 10⁶ times. This simulation yields a range in ⁸⁷Sr/⁸⁶Sr for each of a million different 301 302 model melt inclusion populations, giving a 95% confidence interval of ~8200 ppm for the ranges 303 in ⁸⁷Sr/⁸⁶Sr across all simulations. The 95% confidence interval in the range in ⁸⁷Sr/⁸⁶Sr for the 304 simulation approaches the ~8500 ppm range in 87 Sr/ 86 Sr reported in the suite of 135 melt 305 inclusions analyzed by Sobolev et al. (2011), and the comparison is shown in Figure 8. In 306 summary, if the larger measurement errors of Sobolev et al. (2011) data are applied to a relatively homogeneous population of melt inclusions analyzed by TIMS, it is possible to 307 generate apparent ⁸⁷Sr/⁸⁶Sr variability that is comparable to the reported ⁸⁷Sr/⁸⁶Sr variability in 308 Sobolev et al. (2011). Thus, the large reported ⁸⁷Sr/⁸⁶Sr variability previously found in Pu'u Wahi 309 melt inclusions appears to be the result of larger uncertainties associated with the ⁸⁷Sr/⁸⁶Sr 310 311 analyses by ICP-MS.

312 We did not encounter any melt inclusions with the UDM trace element patterns identified 313 by Sobolev et al. (2011). Therefore, we acknowledge that these UDM melt inclusions could have 314 different ⁸⁷Sr/⁸⁶Sr compositions than the Pu'u Wahi inclusions that we examine here. However, we note that the high ⁸⁷Sr/⁸⁶Sr values (~0.708) reported in the UDM inclusions are generated in 315 316 our Monte Carlo simulation, which can be seen in Figure 8 by the overlap of the model melt 317 inclusions in red with the Sobolev et al. (2011) melt inclusions in blue at ~ 0.708 . Therefore, the 318 ⁸⁷Sr/⁸⁶Sr data for Pu'u Wahi melt inclusions from Sobolev et al. (2011) are not included in Figure 319 6 or in the interpretation of Mauna Loa⁸⁷Sr/⁸⁶Sr data, but we do show the Sobolev et al. (2011) melt inclusions ⁸⁷Sr/⁸⁶Sr data in comparison to the TIMS data in Figure 7. Below we address 320 321 only the origin of the non-UDM inclusions with high (Sr/Ce)_N identified by Sobolev et al. (2000, 322 2011), which were argued by Sobolev et al. (2000) to be sourced by ancient recycled gabbro.

323 4. Discussion

4.1 Processes that can influence melt inclusion compositions prior to, and following,entrapment.

When working with melt inclusions, it is necessary to consider how the melts may have been modified before and after entrapment. For example, following entrapment, melt inclusion compositions can be affected by diffusive exchange, through the host crystal, with the surrounding melt. Relative to Hawaiian lavas, and host glasses, the melt inclusions in this study tend to have lower FeOt (Figure S4), which is indicative of diffusive Fe-Mg exchange between the melt inclusion and the host melt. Fe-Mg diffusive exchange in melt inclusions is relatively common, and has been attributed to slower cooling, as a longer residence time is required for Fe to diffuse out of the host mineral (e.g., Danyushevsky et al., 2000; Gaetani & Watson, 2000; Sobolev & Danyushevsky, 1994). However, Fe-Mg exchange does not influence trace element

ratios or the ⁸⁷Sr/⁸⁶Sr of the melt inclusion, therefore "Fe loss" does not affect the conclusions of this study and we do not attempt to correct for it.

337 Additionally, melt inclusions experience post-entrapment crystallization during cooling, 338 and we correct for post-entrapment loss of olivine to the melt inclusion wall. All Hawaiian melt 339 inclusion major and trace element data shown in the figures have been fractionation corrected to 340 be in equilibrium with the forsterite (Fo) content of the host olivine by adding or subtracting 341 equilibrium olivine at 0.1% increments. For the calculation, we assume (i) an olivine-melt Fe-Mg K_d of 0.30 (Ford et al., 1983; $K_d = (Fe^{2+}/Mg)_{olivine} / (Fe^{2+}/Mg)_{melt}$) and (ii) 10% of the total iron in 342 the melt is Fe³⁺ (following Hauri, 1996). Major, volatile, and trace element concentrations 343 344 corrected for olivine addition or subtraction are shown in Table S1, together with measured 345 concentrations.

346 Prior to entrapment, melts can assimilate altered oceanic crust. Chlorine is an indicator 347 for assimilation of seawater-derived materials, including altered oceanic crust. Thus, melts that 348 have assimilated seawater-derived materials can have elevated Cl, Cl/K₂O, and Cl/Nb ratios 349 (Jambon et al., 1995; Kendrick et al., 2013a, 2015; Kent et al., 1999a, 1999b, 2002; Koleszar et 350 al., 2009; Lassiter et al., 2002; Michael & Cornell, 1998). Chlorine is fluid mobile (e.g., Debret 351 et al., 2014; John et al., 2011; Kendrick et al., 2013b; Rose-Koga et al., 2012; Straub & Layne, 352 2003), therefore recycled oceanic crust that has gone through a subduction filter is not predicted 353 to retain high Cl/K₂O and Cl/Nb ratios (e.g., Kendrick et al., 2013a, 2015, 2017; Rowe & 354 Lassiter, 2009). Mantle-derived lavas have low Cl/Nb ratios relative to lavas influenced by 355 assimilation of altered oceanic crust (e.g., 5-17; Michael & Cornell, 1998; Rowe & Lassiter, 356 2009; Rowe et al., 2015; Saal et al., 2002; Stroncik & Haase, 2004). The Hawaiian melt 357 inclusions considered here generally have mantle-like Cl/Nb (ratios of 4 to 14, but inclusion 358 Puu7-2-7a has a higher ratio, 21), except for Loihi glasses (ratios of 34 to 65) and their melt 359 inclusions (ratios of 14 to 30), which confirms observations of elevated Cl and Cl/Nb in Loihi 360 glasses and melt inclusions (Kent et al., 1999a, 1999b). Critically for this study, the highest 361 (Sr/Ce)_N melt inclusion has mantle-like Cl/Nb, so assimilation of shallow-level altered oceanic 362 crust is unlikely to be involved in its origin. However, the low Cl/Nb ratio of the high (Sr/Ce)_N 363 inclusion does not exclude a contribution from the Cretaceous lower (gabbroic) oceanic crust 364 beneath Hawai'i (which may have mantle-like Cl/Nb, see Section 4.2), which is discussed below.

365 None of the high Cl/Nb inclusions (i.e., Loihi) have elevated (Sr/Ce)_N.

4.2 Source of high (Sr/Ce)_N "ghost plagioclase" in Mauna Loa melt inclusions: bulk assimilation of gabbro?

This study presents the highest magmatic $(Sr/Ce)_N$ ratio in Hawaiian melts measured to date, suggesting a role for a plagioclase-bearing lithology, such as cumulate gabbro, in the petrogenesis of this, and similar, high $(Sr/Ce)_N$ inclusions from Hawai'i. However, the mechanism for the incorporation of the high $(Sr/Ce)_N$ "ghost plagioclase" signature (i.e., elevated

372 (Sr/Ce)_N without concomitant enrichment in Al₂O₃) remains controversial (Peterson et al., 2014;

373 Saal et al., 2007; Sobolev et al., 2000; Yaxley & Sobolev, 2007).

374 Gabbros from the deeper regions of Mauna Loa volcano may have mantle-like Cl/Nb, so 375 assimilation of such gabbros may not be detected with Cl/Nb. In order to test for direct gabbro 376 assimilation, we present a mixing model between the average "normal" Pu'u Wahi melt inclusion 377 composition (an average composition of Pu'u Wahi melt inclusions lacking anomalous major and 378 trace elements signatures, defined in Table S10) and the Mauna Loa gabbroic xenolith with the 379 highest (Sr/Ce)_N-to minimize the quantity of gabbro needed to generate the elevated (Sr/Ce)_N in 380 the melt inclusion—from Gaffney (2002; Figure 3). The highest (Sr/Ce)_N melt inclusion (sample 381 ML-2012-27 19b) is offset from the normal melt inclusion-Mauna Loa gabbro mixing line to 382 lower Al₂O₃ at a given (Sr/Ce)_N, and is therefore not well described by assimilation of this or any 383 other available Mauna Loa gabbro composition. Additionally, the mixing line in Figure 3a 384 informs the minimum amount of Mauna Loa gabbro assimilation (90%) required to generate the 385 high (Sr/Ce)_N in this melt inclusion, given available gabbro compositions at Mauna Loa. This 386 high proportion of Mauna Loa gabbro is also inconsistent with the same mixing model in Al_2O_3 versus MgO space (Figure 3c): This is because 90% Mauna Loa gabbro assimilation by the 387 388 inclusion would cause it to plot off (toward higher Al₂O₃) of the array formed by Hawaiian lavas 389 in Al₂O₃ versus MgO space, but the high (Sr/Ce)_N inclusion is not displaced from this array 390 (Figure 3c).

391 Assimilation of gabbro from the Cretaceous oceanic crust that underlies Hawai'i was also 392 tested using Gabal Gerf ophiolite gabbros (Zimmer et al., 1995), which serve as a proxy for 393 Cretaceous oceanic crust gabbro. However, there is no mixing model between normal-type Pu'u 394 Wahi melts and Gabal Gerf gabbros that can account for the ghost plagioclase inclusion (one 395 mixing line is shown, which trends to the highest (Sr/Ce)_N Gabal Gerf composition, in Figure 3). 396 The mixing lines generate Al_2O_3 that is too high for the high $(Sr/Ce)_N$ inclusion in $(Sr/Ce)_N$ 397 versus Al₂O₃ and Al₂O₃ versus MgO space. Thus, we can exclude bulk assimilation of gabbro in 398 the genesis of the high (Sr/Ce)_N melt inclusion. This observation is central to the paradox of the 399 "ghost plagioclase" signature, where trace element signatures (i.e, high $(Sr/Ce)_N$) indicate 400 gabbroic influence, whereas major elements (i.e., Al₂O₃) are inconsistent with direct assimilation 401 of gabbro. While we can exclude direct assimilation of gabbro, these mixing models do not 402 address the two remaining hypotheses for the origin of high (Sr/Ce)_N signatures at hotspots— 403 ancient recycled gabbro or diffusive interaction between Hawaiian melts and Mauna Loa 404 gabbros-which we test with Sr isotopes below.

405

406 4.3 Source of high (Sr/Ce)_N "ghost plagioclase" in Mauna Loa melt inclusions: diffusive 407 exchange between upwelling melts and shallow gabbro?

408

409 Examining lavas and melt inclusions from the Galapagos that exhibit the trace element 410 signature commonly associated with plagioclase, Saal et al. (2007) and Peterson et al. (2014) 411 argue against the presence of ancient recycled oceanic gabbro in the mantle source. Instead, they 412 proposed that diffusive exchange between mantle-derived melts and gabbros in the deep volcanic 413 sections of Galapagos volcanoes generate the measured, plagioclase-related (i.e., high (Sr/Ce)_N) 414 trace element signatures. In addition to melts with plagioclase-influenced compositions, 415 Galapagos olivine-hosted melt inclusions from Fernandina exhibit the ghost plagioclase 416 signature. Peterson et al. (2014) examined the range of trace element and Pb isotopic 417 compositions that could be generated from both melting an ancient recycled oceanic gabbro in 418 the deep mantle and from diffusive interaction in the shallow crust. Peterson et al. (2014) 419 demonstrated that the recycled oceanic gabbro model cannot reproduce the trace element

420 signature and the Pb isotope compositions of the ghost plagioclase inclusions simultaneously.

421 However, the diffusive exchange model generates both the trace element and Pb isotope

422 compositions of the high $(Sr/Ce)_N$ inclusions with the ghost plagioclase signature.

423 Therefore, instead of reflecting a source signature related to recycled gabbro 424 heterogeneities embedded in the plume, as advocated by Sobolev et al. (2000) for Hawai'i, the 425 model for the generation of ghost plagioclase signatures proposed by Saal et al. (2007) and 426 Peterson et al. (2014) suggests that it is a shallow process, where diffusive interaction between 427 plagioclase in lithospheric gabbros and upwelling hotspot melts results in the hotspot melts 428 inheriting high (Sr/Ce)_N signatures. In this diffusive exchange model, Sr diffuses through the 429 crustal plagioclase toward the upwelling plume melt because it has a higher diffusivity in 430 plagioclase relative to other trace elements, such as La and Nd. In this way, a large positive Sr 431 anomaly can be inherited by the upwelling plume melt due to diffusive interaction with the 432 plagioclase, thereby generating the ghost plagioclase composition.

433 We apply the Peterson et al. (2014) "diffusive exchange" approach used in the Galapagos 434 to evaluate whether melt-gabbro diffusive interaction can generate the elevated (Sr/Ce)_N 435 signatures in Mauna Loa inclusions. The model parameters follow Peterson et al. (2014), except 436 that a Mauna Loa lava (sample J2-15-13, with a relatively high Sr content of 689 ppm; Wanless 437 et al., 2006) is used instead of the high-Sr concentration Galapagos melt used in Peterson et al. 438 (2014), and we track the influence of the diffusive exchange model on ⁸⁷Sr/⁸⁶Sr instead of Pb 439 isotopes. (An incompatible element rich melt, like J2-15-13 used here, may have been more 440 common during the pre-shield alkali volcanism at Mauna Loa.) A plagioclase trace element 441 composition is calculated that is in equilibrium with Mauna Loa lava J2-15-13 using a 442 plagioclase An₈₀ composition at 1200 °C and plagioclase-melt partition coefficients from Bédard 443 (2006). Hawaiian melt inclusions with ghost plagioclase signatures (similar to Galapagos melt 444 inclusions with ghost plagioclase signatures) are depleted in incompatible trace elements, except 445 those associated with plagioclase, which is reflected in the low (La/Sm)_N of the ghost plagioclase 446 inclusions. Because of this depleted nature, we use a 12% aggregated fractional melt of 447 Workman and Hart's (2005) depleted MORB mantle (DMM) as the percolating melt that 448 diffusively exchanges with the plagioclase-rich lithology. The average of East Pacific Rise ridge 449 segments from Gale et al. (2013) is used for the initial ⁸⁷Sr/⁸⁶Sr composition of the percolating 450 melt. This percolating melt undergoes diffusive exchange with a troctolite (for simplicity) 451 hosting 50% modal plagioclase with the modeled trace element composition, where diffusive 452 exchange uses published trace element diffusivities (Cherniak, 1995, 2002, 2003; Cherniak & 453 Watson, 1994; Giletti & Casserly, 1994; Giletti & Shanahan, 1997) at 1200°C (see Table 2 for 454 diffusivities used). Hawaiian lava temperatures approach 1200°C (Gansecki et al., 2019), making 455 this an appropriate temperature for model calculation. The duration of diffusive interaction varies 456 from 1 year to 400 years in our model, as plagioclase trace element signatures are inherited by 457 the percolating melt as a function of time (Table 2). Because Sr has a higher diffusivity than Ce 458 and Al in plagioclase, the percolating DMM melt can inherit high $(Sr/Ce)_N$, but not elevated 459 Al₂O₃, thereby providing an explanation for the ghost plagioclase signature without invoking 460 direct gabbro assimilation (Danyushevsky et al., 2003) or ancient recycled gabbro (Sobolev et al. 461 2000). Depletion in light REEs relative to middle REEs is an outcome of the diffusive exchange 462 model, where lower $(La/Sm)_N$ are generated as higher $(Sr/Ce)_N$ are produced (Figure 4). Finally, 463 we mix the diffusively modified melts with the host glass associated with the highest (Sr/Ce)_N 464 inclusion, sample ML-2012-27 (though mixing with average Pu'u Wahi normal-type melt 465 inclusions would not change the result), and then compare the mixtures to measured Hawaiian

melt inclusion compositions. Following 1 to 5 years of diffusive exchange (i.e., timescales that
are well within crystal residence times in Hawaiian magmas; e.g., Cooper et al., 2001; Vinet &
Higgins, 2010; Wieser et al., 2019), the (La/Sm)_N, (Sr/Ce)_N, and ⁸⁷Sr/⁸⁶Sr composition of the

469 highest Mauna Loa $(Sr/Ce)_N$ inclusion is matched in the diffusive exchange model.

470 This model requires the presence of Mauna Loa gabbros deep in the volcanic pile of the

471 volcano. Gaffney (2002) examined a suite of Mauna Loa gabbros (crystallized from Mauna Loa

472 magmas), which provide evidence of cumulus plagioclase in the deep stratigraphy of the

473 volcano. Although gabbroic xenoliths from Mauna Loa are uncommon, the lava pile is many

474 kilometers thick, and only a fraction of the lava pile has been sampled directly. Therefore,

gabbro could be more abundant in the deep shield than is suggested by the low abundance of

476 gabbroic xenoliths that have been transported to the surface.

Table 2. Model parameters and output data for the diffusive interaction model based on the model of Peterson et al. (2014).

	K _d (plag-melt) ^a	D _{plag} (1200°C,	Plagioclase	Initial	Output data	(concentratio	ons in ppm)					
		m² s ⁻¹) ^b	melt ^c	$melt^d$								
			(ppm)	(ppm)	1 year	5 years	10 years	20 years	50 years	100 years	200 years	400 years
Ba	0.230	8.09E -19	198	4.69	4.86	5.52	6.35	7.99	12.82	20.57	35.03	60.18
Th	0.050	3.10E -21	1.37	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.07
U	0.024	3.10E -21	0.47	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03
Nb	0.030	3.10E -21	20.4	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24
К	0.171	4.38E -16	7222	408	2364	5618	6368	6491	6494	6494	6494	6494
La	0.106	3.87E -19	21	1.60	1.60	1.62	1.64	1.67	1.78	1.96	2.32	3.01
Ce	0.081	3.87E -19	53.5	4.57	4.58	4.61	4.64	4.71	4.92	5.28	5.97	7.32
Pb	0.511	1.41E -17	1.69	0.15	0.23	0.52	0.80	1.17	1.55	1.63	1.63	1.63
Nd	0.084	2.92E -19	36	4.76	4.77	4.78	4.80	4.83	4.94	5.11	5.46	6.14
Sr	1.813	3.37E -17	689	63.5	217	534	647	680	682	682	682	682
Zr	0.004	3.10E -21	264	41.5	41.5	41.5	41.5	41.5	41.5	41.5	41.5	41.5
Hf	0.015	3.10E -21	6.33	1.27	1.27	1.27	1.27	1.27	1.27	1.27	1.27	1.27
Sm	0.056	2.92E -19	9.3	1.88	1.88	1.88	1.88	1.89	1.90	1.93	1.99	2.10
Eu	0.534	2.92E -19	3.06	0.74	0.83	1.13	1.39	1.67	1.88	1.94	2.01	2.15
Ti	0.076	3.10E -21	21049	5310	5310	5311	5311	5311	5311	5312	5314	5317
Dy	0.019	3.24E -19	6.77	3.37	3.58	3.58	3.58	3.58	3.58	3.58	3.58	3.58
Yb	0.015	5.06E -19	2.09	2.04	2.04	2.04	2.04	2.04	2.04	2.04	2.04	2.04
⁸⁷ Sr/ ⁸⁶ Sr			0.703872	0.702569	0.703585	0.703833	0.703863	0.703870	0.703871	0.703871	0.703871	0.703871

For the diffusion model we assume that the plagioclase grain size is 0.001 m, there is 1% melt, and 50% of the solid is plagioclase. ^aBédard (2006) for plagioclase of An_{an} at 1200°C

^bDiffusion coefficients for plagioclase at 1200°C from Cherniak and Watson (1994), Giletti and Casserly (1994), Cherniak (1995, 2002, 2003) and Giletti and Shanahan (1997). However, some trace elements do not have experimentally determined diffusion coefficients. For trace elements without this data, Peterson et al. (2014) estimated based on cation size and charge (Behrens et al., 1990; Saal and Van Orman, 2004). Peterson et al. (2014) assumed that the diffusion coefficients for Nb, Zr, and Ti are similar to those of Th and U. Like Peterson et al. (2014), we assumed that Eu is 50% Eu²⁺, and that the diffusion coefficient for Eu²⁺ is similar to that of Sr. ^c While Peterson et al. (2014) used a Galapagos lava to calculate an equilibrium plagioclase composition, we use a Mauna Loa lava (Mauna Loa sample J2-15-13; Wanless et al., 2006). The plagioclase composition in equilibrium with this melt is calculated using the partition coefficients in the table.

^dLike Peterson et al. (2014), the initial melt is a 12% aggregated fractional melt of DMM composition from Workman and Hart (2005), and the isotopic value is the average of the East Pacific Rise Ridge segment averages from Gale et al. (2013).

477 The diffusive exchange model for the high (Sr/Ce)_N melt inclusions relies on the fact that the percolating melt is depleted, and this may seem unexpected for a hotspot volcano far from a 478 479 MOR. Nonetheless, depleted components have been suggested to exist in the Hawaiian plume 480 (e.g., Garcia et al., 2010; Harrison et al., 2020). Garcia et al. (2010) hypothesize that an ancient 481 depleted upper mantle component exists in the Hawaiian plume, that could potentially supply the 482 depleted percolating melt that interacts with the Mauna Loa gabbro in our model. Additionally, 483 Mukhopadhyay et al. (2003) state that they "favor an asthenospheric origin for the DM 484 component" in Hawai'i, and that "the DM component is most apparent in lavas from the Kauai 485 shield and is present in varying proportion in other Hawaiian shield volcanoes," including Mauna 486 Loa (which has ~20% of the DM component in Figure 14 of Mukhopadhyay et al., 2003). As

487 such, evidence exists that supports the existence of a depleted component in the Hawaiian plume
488 that sources shield stage lavas, consistent with our model. While the precise origin of the
489 depleted component is unknown, we use the composition of the Workman and Hart (2005)
490 depleted mantle component to approximate its trace element composition.

491 The results of the melt-plagioclase diffusive exchange model are compared with the 492 Hawaiian melt inclusion data in Figures 4 and 6, and the model encompasses both the ⁸⁷Sr/⁸⁶Sr 493 composition and the unusual trace element composition—high (Sr/Ce)_N and low (La/Sm)_N—of 494 the Mauna Loa ghost plagioclase inclusion examined here. While diffusive exchange with a 495 Mauna Loa cumulate gabbro is consistent with the high (Sr/Ce)_N inclusion data presented here, 496 the Mauna Loa-like ⁸⁷Sr/⁸⁶Sr in the high (Sr/Ce)_N inclusion is inconsistent with diffusive 497 exchange between upwelling melts and MOR-basement gabbro. Diffusive exchange between 498 melts and MOR-basement gabbro in the Pacific lithosphere would predict low, MORB-like 499 87 Sr/ 86 Sr in the melt inclusion. This is because the model predicts that the percolating melt will rapidly (>5 years; Figure 6) become dominated by the ⁸⁷Sr/⁸⁶Sr of the gabbro, and the average 500 xenoliths from Hualalai sampling MOR-basement gabbros indicate that the Cretaceous oceanic 501 502 crustal gabbros beneath Hawai'i have 87 Sr/ 86 Sr (0.702733 ± 0.000392 (2SD); Gao et al., 2016) that is indistinguishable from East Pacific Rise MORB (0.70257 ± 0.00024 ; Gale et al., 2013) 503 504 due to relatively little modification by seawater. Furthermore, the low Cl/K (<0.01) in the highest 505 (Sr/Ce)_N inclusion rules out interaction with seawater-altered MOR gabbro with higher 506 (secondary) ⁸⁷Sr/⁸⁶Sr, because such a model would predict a high Cl/K ratio in the melt 507 inclusion. Incidentally, Peterson et al. (2014) exclude diffusive melt exchange with MORB-508 related gabbros in the Galapagos because this would generate a *negative* Sr anomaly, due to 509 lower Sr concentrations in oceanic gabbros than melts from oceanic hotspot volcanoes, a result 510 that is directly applicable to the Hawaiian case examined here. Thus, we can rule out Cretaceous 511 oceanic crustal gabbro as the origin of the ghost plagioclase signature.

512 An important remaining question is to explore whether the ⁸⁷Sr/⁸⁶Sr we measured in the 513 Mauna Loa inclusion with the ghost plagioclase signature is consistent with the recycled gabbro 514 model proposed by Sobolev et al. (2000). We explore this question below.

4.4 Using Sr isotopes to test models for the origin of the ghost plagioclase signature in Mauna Loa inclusions.

517 The primary result of this study is that the Mauna Loa melt inclusion with high $(Sr/Ce)_N$ 518 is indistinguishable in ⁸⁷Sr/⁸⁶Sr from other melt inclusions from the same lava (sample ML-2012-519 27; Figure 6). This melt inclusion also has ⁸⁷Sr/⁸⁶Sr that falls within the range of ⁸⁷Sr/⁸⁶Sr 520 identified in Mauna Loa lavas. We explore the implications of this observation below.

521 While a recycled gabbro signature may be present in the mantle for most Loa-trend 522 volcanoes, including Mauna Loa (Frey et al., 2016), recycled gabbro is unlikely to have 523 contributed the high (Sr/Ce)_N signature to the high (Sr/Ce)_N Mauna Loa inclusion examined here. 524 If high (Sr/Ce)_N is a measure of magnitude of the contribution of the recycled gabbro component, 525 as proposed by Sobolev et al. (2000), this raises the question as to why the highest (Sr/Ce)_N melt inclusion has the same ⁸⁷Sr/⁸⁶Sr as the Mauna Loa melt inclusions and lavas with normal 526 527 (Sr/Ce)_N. In their model, the recycled gabbro contributes approximately 73 to 79% of the total Sr 528 budget in ghost plagioclase melts, which is much larger than gabbro contributions suggested for 529 Hawaiian lavas. Therefore, if an excess contribution from recycled gabbro were responsible for 530 the high (Sr/Ce)_N signature in the unusual, high (Sr/Ce)_N Mauna Loa melt inclusion examined

531 here, then the high $(Sr/Ce)_N$ melt inclusions should have ${}^{87}Sr/{}^{86}Sr$ that is shifted closer to the

⁸⁷Sr/⁸⁶Sr of the recycled gabbro compositions than the Mauna Loa melt inclusions with normal 532 533 (Sr/Ce)_N. This is not observed: The high (Sr/Ce)_N melt inclusion and inclusions with normal (Sr/Ce)_N have indistinguishable ⁸⁷Sr/⁸⁶Sr. If one appeals to coincidence, and assumes that the 534 535 recycled gabbro has evolved to have exactly the same ⁸⁷Sr/⁸⁶Sr composition as the other component(s) sampled by the Mauna Loa inclusions with normal (Sr/Ce)_N, then one can explain 536 537 why high $(Sr/Ce)_N$ and normal $(Sr/Ce)_N$ inclusions from Mauna Loa have the same ${}^{87}Sr/{}^{86}Sr$. 538 However, we do not accept such a coincidence as plausible, for two reasons. First, possible 539 compositions of the ancient recycled gabbro protolith are highly variable-including, but not 540 limited to, gabbro from recycled seamounts, large igneous provinces, or oceanic lithosphere 541 (depleted-MORB, normal-MORB, or enriched-MORB)-and thus will run the gamut of Sr-542 isotopic compositions, making it unlikely that ancient recycled gabbro would coincidently evolve to have ⁸⁷Sr/⁸⁶Sr like Mauna Loa lavas that lack the ghost plagioclase inclusions. Second, 543 variable dehydration and melting processes operating on heterogeneous protoliths at different 544 545 times in distinct subduction zones, followed by varying periods of storage in the mantle and 546 admixture with diverse reservoirs, are unlikely to generate different components-a high 547 (Sr/Ce)_N recycled gabbro component and low (Sr/Ce)_N components—in the plume with 548 indistinguishable ⁸⁷Sr/⁸⁶Sr. In short, an ancient recycled gabbro component is unlikely to be 549 responsible for the high (Sr/Ce)_N "ghost plagioclase" melt inclusions.

550 A more straightforward explanation for the identical 87 Sr/ 86 Sr in the high (Sr/Ce)_N melt 551 inclusions and "normal" (Sr/Ce)_N inclusions in Mauna Loa is that the former inherited the high 552 (Sr/Ce)_N signature by interacting with Mauna Loa gabbros. Gabbros related to construction of 553 the hotspot volcano will have ⁸⁷Sr/⁸⁶Sr ratios that are similar to Mauna Loa lavas, normal-type 554 inclusions, and the ghost plagioclase inclusion in this study. The diffusive exchange model 555 generates the high (Sr/Ce)_N in ghost plagioclase melt inclusions—indicative of plagioclase-melt interaction—and is also consistent with the relationships between (Sr/Ce)_N and ⁸⁷Sr/⁸⁶Sr (Figures 556 557 4 and 6). This result, which employs Sr isotopic constraints on the origin of ghost plagioclase 558 melt inclusions, mirrors the findings of Peterson et al. (2014), who also showed that the Pb 559 isotopic compositions in Galapagos ghost plagioclase melt inclusions are inconsistent with an 560 ancient recycled oceanic gabbro origin.

561 5. Conclusion

562 This study uses Sr isotopic measurements in individual melt inclusions to evaluate the 563 origin of the ghost plagioclase signature—elevated (Sr/Ce)_N, but similar Al₂O₃, as melt 564 inclusions with (Sr/Ce)_N<2—in Mauna Loa melt inclusions. The ⁸⁷Sr/⁸⁶Sr of the highest (Sr/Ce)_N melt inclusion ((Sr/Ce)_N =7.2) matches the 87 Sr/ 86 Sr in lower (Sr/Ce)_N melt inclusions ((Sr/Ce)_N 565 < 2) from the same lava, and is also indistinguishable from ⁸⁷Sr/⁸⁶Sr in Mauna Loa lavas. 566 567 Because the highest (Sr/Ce)_N melt inclusion is isotopically indistinguishable from Mauna Loa 568 melts with small Sr anomalies, the source of the high (Sr/Ce)_N melt inclusions should have the 569 same ⁸⁷Sr/⁸⁶Sr as the component that generates other Mauna Loa lavas with smaller (Sr/Ce)_N 570 ratios. According to Sobolev et al. (2000), the ⁸⁷Sr/⁸⁶Sr of the recycled gabbro will dominate the ⁸⁷Sr/⁸⁶Sr signature in ghost plagioclase melt inclusions because the recycled gabbro contributes 571 572 to the majority of the total Sr budget in ghost plagioclase melts. While we cannot rule out 573 recycled gabbro as the source of the ghost plagioclase signature, it would be a great coincidence 574 that any recycled gabbro component in the Hawaiian mantle plume melting beneath Mauna Loa would have the same ⁸⁷Sr/⁸⁶Sr as the source of other Mauna Loa lavas that lack a recycled 575 576 gabbro signature. Therefore, the observation that the ⁸⁷Sr/⁸⁶Sr of the highest (Sr/Ce)_N inclusion

- 577 matches Mauna Loa lavas supports a petrogenetic origin for the ghost plagioclase melt inclusions
- 578 that relates to the Mauna Loa magma plumbing system. We find that the origin of the ghost
- 579 plagioclase signature within melt inclusions can be explained by diffusive interaction between
- rising melt and plagioclase-rich lithologies in the deep magma cumulates of Hawaiian volcanoes.
- 581 Thus, the ghost plagioclase signature may not be a signature directly derived from the mantle
- 582 source, but instead is likely a result of recent crustal processes operating in the deep regions of
- 583 Hawaiian volcanoes.

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▲ Gabal Gerf gabbros (Zimmer et al., 1995)



	Sample	Melt inclusions	Glass	Volcano						
	ML-2012-27	7 😑	0	Mauna Loa						
	Puu2 & Puu	17 🔴	0							
	6K490-1		◇	Loihi						
	6K490-2	\diamond	\diamond	Lonn						
	К89-6			Koolau						
Prev	iously publish	ied data								
• H	igh (Sr/Ce) _N m	nelt inclusions (Sobc	olev et al., 2	2000, 2011)						
• U	Ultra-depleted melt (UDM) inclusions (Sobolev et al., 2011)									
O Lo	• Low (Sr/Ce), Mauna Loa melt inclusions (Sobolev et al., 2000, 2011)									
Pu'u Wahi melt inclusions (Reinhard et al., 2018)										
• M	launa Loa gab	bros (Gaffney, 2002	2)							
G	abal Gerf gab	bros (Zimmer et al.,	1995)							











Table 1. '	°′Sr/°	°Sr measurements mac	le on melt i	nclusions and	the gla	ass of the	host lav	a in this stu	udy.
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Sample	Sr (ppm)	Ce (ppm)	(Sr/Ce),	Total Sr (ng)	[Sr]comple/[Sr]blank	⁸⁷ Sr/ ⁸⁶ Sr ^a	2σ (std. err.)	ppm change in ⁸⁷ Sr/ ⁸⁶ Sr
		(PP)	()					ratio from blank correction
Mauna Loa - Pu'u	Wahi (Puu2 8	k Puu7)						
Puu2-1-2	-	-	-	2.5	222	0.703772	0.000032	46
Puu2-1-9	270	20.6	1.11	15.5	292	0.703800	0.000021	35
Puu2-1-18a	337	26.7	1.06	2.8	53	0.703885	0.000049	191
Puu2-1-19	338	24.1	1.18	1.1	97	0.703899	0.000024	104
Puu2-1-39	215	15.8	1.14	7.3	1038	0.703935	0.000010	10
Puu7-1-20	438	24.9	1.48	1.2	177	0.703867	0.000034	57
Puu7-2-15	-	-	-	3.7	527	0.703845	0.000018	19
Puu7-2-92b	-	-	-	0.9	78	0.703917	0.000065	129
Puu7-2-80	324	20.3	1.34	9.2	817	0.703953	0.000016	12
Puu7-2-19	325	35.2	0.78	18.9	2690	0.703838	0.00008	4
Puu7-1-29	330	25.2	1.10	1.2	168	0.703804	0.000038	61
Puu7-3-19a	314	21.9	1.21	17.5	329	0.703916	0.000009	31
Puu7-3-31	265	19.1	1.16	28.8	542	0.703700	0.000023	19
Puu7-1-14	279	28.0	0.84	3.6	67	0.703947	0.000021	149
Puu7-1-30	325	31.6	0.87	2.5	46	0.703896	0.000013	218
Puu7-1-36	338	25.9	1.10	23.5	441	0.703770	0.000014	23
Puu7-2-62	334	24.3	1.16	3.1	444	0.703887	0.000030	23
Puu7-1-41	330	22.8	1.21	7.7	682	0.703977	0.000023	15
Puu7-2-6	327	25.8	1.07	9.8	1400	0.703811	0.000010	7
Puu7-2-40a	280	19.2	1.22	9.6	855	0.703770	0.000021	12
Puu7-2-42	336	22.9	1.24	5.3	469	0.703986	0.000029	21
Puu7-2-7a	311	22.5	1.16	3.1	445	0.703942	0.000040	23
Puu7-1-10	355	26.3	1.14	13.5	1923	0.703949	0.000008	5
Puu7-2-52	345	23.0	1.26	5.0	447	0.703882	0.000035	23
host glass ^c	319	25.9	1.04			0.703912	0.000008	
Mauna Loa ML-20	12-27							
MI 2012-27 2	202	15.2	1 56	10.0	1027	0 702001	0.000012	10
NIL-2012-27_2	203	10.0	1.01	10.0	1027	0.703001	0.000013	10
MI 2012-27_5a	437	50.1 24.1	1.01	10.2	200	0.704010	0.000016	10
NIL 2012 27 192	441	54.1 2E Q	1.09	2.9	500	0.703007	0.000010	54 1 <i>11</i>
NIL-2012-27_10d	420	55.6 24.4	1.00	0.7	101	0.704059	0.000030	144
NIL-2012-27_21a	457	34.4 20.2	1.07	1.0	101	0.704009	0.000043	55
IVIL-2012-27_25	411	50.2	1.15	2.1	215	0.703965	0.000021	40
NIL-2012-27_190	780	9.1	1.24	1.4	74	0.703828	0.000045	137
	344	27.0	1.05			0.703909	0.000006	
Koolau - K89-6								
K89-6_52	-	-	-	3.5	363	0.704401	0.000031	26
K89-6_53	260	16.2	1.35	0.5	25	0.703654	0.000079	416
K89-6_55	244	14.1	1.45	10.5	1495	0.703960	0.000027	7
K89-6_65	327	20.0	1.38	1.4	86	0.703864	0.000026	118
K89-6_76a	267	17.6	1.27	0.6	65	0.703906	0.000041	155
host glass	366	23.3	1.32			0.703965	0.000006	
Loihi - 6K490-1								
6K490-1_51	-	-	-	0.9	52	0.703284	0.000093	211
	459	36.1	1.07	0.3	31	0.703665	0.000090	339
host glass	460	42.1	0.92			0.703586	0.000006	
Loihi - 6K490-2								
6K490-2 79a	507	44.5	0.96	1.1	63	0.703363	0.000033	173
host glass	412	35.0	0.99			0.703574	0.000006	

Note: The Sr and Ce fractionation-corrected concentrations are reported here, but not for partially crystalline inclusions.

^a ⁸⁷Sr/⁸⁶Sr values have been blank corrected and corrected for the offset between the preferred (0.710240) and the measured ⁸⁷Sr/⁸⁶Sr of NBS987. ^b The change in ⁸⁷Sr/⁸⁶Sr ratio due to the blank correction (ppm) is calculated as (⁸⁷Sr/⁸⁶Sr_{meas-norm987}-⁸⁷Sr/⁸⁶Sr_{blank-corrected})/⁸⁷Sr/⁸⁶Sr_{meas-norm987}*10⁶.

^c Sr concentration and ⁸⁷Sr/⁸⁶Sr of Pu'u Wahi glass is from Reinhard et al. (2018).

	K _d (plag-melt) ^a	D _{plag} (1200°C,	Plagioclase	Initial	Output data (concentrations in ppm)							
		m² s ⁻¹) ^b	melt ^c	$melt^d$								
			(ppm)	(ppm)	1 year	5 years	10 years	20 years	50 years	100 years	200 years	400 years
De	0.220	0.00F 10	100	4.60	4.90	F F 2	6.25	7.00	12.02	20.57	25.02	CO 10
ва	0.230	8.09E -19	198	4.69	4.80	5.52	0.35	7.99	12.82	20.57	35.03	60.18
In	0.050	3.10E -21	1.37	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.07	0.07
U	0.024	3.10E -21	0.47	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03
Nb	0.030	3.10E -21	20.4	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24	1.24
К	0.171	4.38E -16	7222	408	2364	5618	6368	6491	6494	6494	6494	6494
La	0.106	3.87E -19	21	1.60	1.60	1.62	1.64	1.67	1.78	1.96	2.32	3.01
Ce	0.081	3.87E -19	53.5	4.57	4.58	4.61	4.64	4.71	4.92	5.28	5.97	7.32
Pb	0.511	1.41E -17	1.69	0.15	0.23	0.52	0.80	1.17	1.55	1.63	1.63	1.63
Nd	0.084	2.92E -19	36	4.76	4.77	4.78	4.80	4.83	4.94	5.11	5.46	6.14
Sr	1.813	3.37E -17	689	63.5	217	534	647	680	682	682	682	682
Zr	0.004	3.10E -21	264	41.5	41.5	41.5	41.5	41.5	41.5	41.5	41.5	41.5
Hf	0.015	3.10E -21	6.33	1.27	1.27	1.27	1.27	1.27	1.27	1.27	1.27	1.27
Sm	0.056	2.92E -19	9.3	1.88	1.88	1.88	1.88	1.89	1.90	1.93	1.99	2.10
Eu	0.534	2.92E -19	3.06	0.74	0.83	1.13	1.39	1.67	1.88	1.94	2.01	2.15
Ті	0.076	3.10E -21	21049	5310	5310	5311	5311	5311	5311	5312	5314	5317
Dy	0.019	3.24E -19	6.77	3.37	3.58	3.58	3.58	3.58	3.58	3.58	3.58	3.58
Ýb	0.015	5.06E -19	2.09	2.04	2.04	2.04	2.04	2.04	2.04	2.04	2.04	2.04
⁸⁷ Sr/ ⁸⁶ Sr			0.703872	0.702569	0.703585	0.703833	0.703863	0.703870	0.703871	0.703871	0.703871	0.703871

Table 2. Model parameters and output data for the diffusive interaction model based on the model of Peterson et al. (2014).

For the diffusion model we assume that the plagioclase grain size is 0.001 m, there is 1% melt, and 50% of the solid is plagioclase.

^aBédard (2006) for plagioclase of An₈₀ at 1200°C

^bDiffusion coefficients for plagioclase at 1200°C from Cherniak and Watson (1994), Giletti and Casserly (1994), Cherniak (1995, 2002, 2003) and Giletti and Shanahan (1997). However, some trace elements do not have experimentally determined diffusion coefficients. For trace elements without this data, Peterson et al. (2014) estimated based on cation size and charge (Behrens et al., 1990; Saal and Van Orman, 2004). Peterson et al. (2014) assumed that the diffusion coefficients for Nb, Zr, and Ti are similar to those of Th and U. Like Peterson et al. (2014), we assumed that Eu is 50% Eu²⁺, and that the diffusion coefficient for Eu²⁺ is similar to that of Sr. ^c While Peterson et al. (2014) used a Galapagos lava to calculate an equilibrium plagioclase composition, we use a Mauna Loa lava (Mauna Loa sample J2-15-13; Wanless et al., 2006). The plagioclase composition in equilibrium with this melt is calculated using the partition coefficients in the table.

^dLike Peterson et al. (2014), the initial melt is a 12% aggregated fractional melt of DMM composition from Workman and Hart (2005), and the isotopic value is the average of the East Pacific Rise Ridge segment averages from Gale et al. (2013).