Extreme isotopic heterogeneity in Samoan clinopyroxenes constrains sediment recycling

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Lavas erupted at hotspot volcanoes provide evidence of mantle heterogeneity. Samoan Island lavas with high $^{87}\text{Sr}/^{86}\text{Sr}$ (>0.706) typify a mantle source incorporating ancient subducted sediments. To further characterize this source, we target a single high $^{87}\text{Sr}/^{86}\text{Sr}$ lava from Savai'i Island, Samoa for detailed analyses of $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ isotopes and major and trace elements on individual magmatic clinopyroxenes. We show the clinopyroxenes exhibit a remarkable range of $^{87}\text{Sr}/^{86}\text{Sr}$—including the highest observed in an oceanic hotspot lava—encompassing ~30% of the oceanic mantle's total variability. These new isotopic data, data from other Samoan lavas, and magma mixing calculations are consistent with clinopyroxene $^{87}\text{Sr}/^{86}\text{Sr}$ variability resulting from magma mixing between a high silica, high $^{87}\text{Sr}/^{86}\text{Sr}$ (up to 0.7316) magma, and a low silica, low $^{87}\text{Sr}/^{86}\text{Sr}$ magma. Results provide insight into the composition of magmas derived from a sediment-infiltrated mantle source and document the fate of sediment recycled into Earth's mantle.
Subduction zones are the largest recycling systems on Earth where sediments, oceanic crust, mantle lithosphere, volatiles, and possibly blocks of continental crust return to the mantle. It is generally accepted that mantle plumes ‘sample’ such recycled materials leading to a diverse range of geochemical signatures reflected in ocean island basalts (OIBs)\(^1-3\). Mantle isotopic heterogeneities are inferred by examining global OIB lavas, but the scale, origin, and compositions of the sources and/or derivative melts remain under intense investigation. Many studies that have evaluated isotopic heterogeneity in OIBs have done so from the analysis of whole rock or bulk mineral separates\(^4-8\), but these types of measurements obscure possible broader isotopic heterogeneity present within individual crystals or melt inclusions within a single lava. Previous work has shown \(87^{\text{Sr}}/86^{\text{Sr}}\) and \(143^{\text{Nd}}/144^{\text{Nd}}\) isotopic heterogeneity in lower oceanic crust clinopyroxene and plagioclase cumulate crystals, and another work has reported \(87^{\text{Sr}}/86^{\text{Sr}}\) heterogeneity in olivine-hosted melt inclusions from a single lava sample—these are a couple of examples among many studies showing isotopic heterogeneity in individual crystals or melt inclusions\(^9-14\). Related to this study, recent work demonstrated intra- and inter-crystal \(87^{\text{Sr}}/86^{\text{Sr}}\) heterogeneity in plagioclase in Samoan lavas with an enriched mantle 2 (EM2)\(^15\) signature\(^16\). Collectively, these studies suggest that multiple isotopically distinct melts can contribute to a single lava, and thus magma mixing\(^13\) is an essential part of igneous petrogenesis.

Subduction recycling of terrigenous sediments into the mantle has been cited to explain the EM2 geochemical signature recorded in lavas erupted at ocean island volcanoes\(^16,17\). Lavas with elevated \(87^{\text{Sr}}/86^{\text{Sr}}\) \((-0.706)^18\) paired with low \(143^{\text{Nd}}/144^{\text{Nd}}\) sampling an EM2 component, are relatively common among the volcanoes of the Samoan Islands in the South Pacific Ocean. Lavas erupted off the submarine flanks of Savai’i Island, western Samoa (Supplementary Fig. 1), have whole rock \(87^{\text{Sr}}/86^{\text{Sr}}\) values up to 0.720469 (ref. 7), plagioclase up to 0.7224 ± 0.0003 (2SE), and plagioclase isotopically distinct melts in oceanic lava is consistent with magma mixing of at least two types of melts\(^15\). The addition of a continental crust-like sediment in large low shear wave velocity provinces (LLSVPs)\(^19,20\). Constraints help shed light on the origin of terrigenous sediment within the Samoan plume and, thereby constraining the petrogenesis of EM2 lavas.

In \(87^{\text{Sr}}/86^{\text{Sr}}\) and \(143^{\text{Nd}}/144^{\text{Nd}}\) space, addition of clinopyroxene Sr and Nd isotopes from lava sample ALIA-D115-18 of this study, together with major and trace element and isotopic constraints from other Samoan whole rock lavas from the same submarine dredge as ALIA-D115-18 (referred to as the ALIA-D115 dredge), are used to constrain the composition of the magma mixing endmembers—including the extreme EM2 endmember—contribution to the isotopic variability observed in clinopyroxenes from ALIA-D115-18. This permits us to better define the origin and makeup of the high \(87^{\text{Sr}}/86^{\text{Sr}}\) EM2 endmember in the Samoan plume, thereby constraining the petrogenesis of EM2 lavas.

Results and discussion

An extreme EM2 sample from Samoa. The studied sample is a 5 Ma submarine lava\(^21\) ALIA-D115-18, dredged off the flanks of Savai’i Island, western Samoa (see Supplementary Fig. 1). This sample is a nepheline-normative trachyandesite with a bimodal crystal population. The larger crystal population is \(\geq 25\) \(\mu\)m in diameter and includes porphyrocrysts of clinopyroxene, plagioclase, and ilmenite as well as porphyroclasts consisting of the same three phases with modal abundances corresponding to a microgabbro. This study focuses on clinopyroxene porphyrocrysts, which were analyzed for major (electron microprobe) and trace elements (laser ablation inductively coupled mass spectrometry (LA-ICP-MS)) prior to dissolution and isotopic analysis by thermal ionization mass spectrometry (TIMS). Seventeen individual clinopyroxene crystals were analyzed for \(87^{\text{Sr}}/86^{\text{Sr}}\) with resulting ratios ranging from 0.716833 (±0.000015, 2SE) to 0.723888 (±0.000015, 2SE), the highest value ever reported in an oceanic lava. Nine of the 17 crystals were also targeted for \(^{143}\text{Nd}/^{144}\text{Nd}\) analysis, and single-crystal values range from 0.512238 (±0.000013, 2SE; \(\epsilon_{\text{Nd}} = -7.6\) calculated using \(^{143}\text{Nd}/^{144}\text{Nd}_{\text{chondrite}} = 0.512630\) (ref. 22)) to 0.512357 (±0.000013, 2SE; \(\epsilon_{\text{Nd}} = -5.3\) (Supplementary Table 1)). Together, the clinopyroxene Sr and Nd isotopes plot along an extension of the mantle array formed by global OIB, and including the new clinopyroxene data of this study, the \(87^{\text{Sr}}/86^{\text{Sr}}\) variability in this single lava sample spans ~35% of the variability observed in all Samoan lavas, and ~30% of the entire oceanic mantle variability (Fig. 1).

Generation of isotopic heterogeneity. Data from clinopyroxene Sr and Nd isotopes from lava sample ALIA-D115-18 of this study, together with major and trace element and isotopic constraints from other Samoan whole rock lavas from the same submarine dredge as ALIA-D115-18 (referred to as the ALIA-D115 dredge), used to constrain the composition of the magma mixing endmembers—including the extreme EM2 endmember—contribution to the isotopic variability observed in clinopyroxenes from ALIA-D115-18. This permits us to better define the origin and makeup of the high \(87^{\text{Sr}}/86^{\text{Sr}}\) EM2 endmember in the Samoan plume, thereby constraining the petrogenesis of EM2 lavas.

In \(87^{\text{Sr}}/86^{\text{Sr}}\) and \(143^{\text{Nd}}/144^{\text{Nd}}\) space, addition of clinopyroxene data obtained in this study to the existing Samoan whole rock data requires a mixing line that exhibits more significant curvature (i.e. \(r < 1\), where \(r = [\text{Nd}]_{\text{S}}/[\text{Sr}]_{\text{M}}\)) where M [mafic] and S [silicic] represent the low- and high- \(87^{\text{Sr}}/86^{\text{Sr}}\) endmembers, respectively\(^21\), than previous suggestions based on whole rock data alone (see ref. 4, Fig. 1). This implies that the Sr and Nd concentrations as well as the \(87^{\text{Sr}}/86^{\text{Sr}}\) and \(143^{\text{Nd}}/144^{\text{Nd}}\) values in the two endmembers need be re-evaluated. It should be noted, however, that the proposed mixing discussed here is not mixing between mantle sources, but mixing between magmas, and therefore differs from the approach in Jackson et al.\(^7\). Employing an inverse model approach to existing whole rock data from Samoa (see “Methods”), we argue for an EM2-derived mixing endmember that is not only high in \(87^{\text{Sr}}/86^{\text{Sr}}\), but also trachytic in composition. Mixing of the EM2 trachytic magma with more primitive Samoan basalt-like compositions reproduces the mixing trends observed in ALIA-D115 whole rock lavas (Fig. 1) as well as the range of \(87^{\text{Sr}}/86^{\text{Sr}}\) and \(143^{\text{Nd}}/144^{\text{Nd}}\) in Samoan clinopyroxene.

The genesis of this high \(87^{\text{Sr}}/86^{\text{Sr}}\), trachytic magma is an outstanding question and is further explored in the discussion and Supplementary Text. This aside, results of this study are consistent with the observation of Jackson et al.\(^7\) who noted a distinct positive correlation between \(87^{\text{Sr}}/86^{\text{Sr}}\) and SiO\(_2\) concentration among Samoan whole rocks from the ALIA-D115 dredge haul, suggesting two-endmember mixing between a highly evolved (high silica, low MgO) endmember with high \(87^{\text{Sr}}/86^{\text{Sr}}\).
and a less evolved (basaltic) endmember with low 87Sr/86Sr. In addition, Edwards et al. 16 noted a negative correlation between 87Sr/86Sr and Sr concentration in plagioclase crystals suggesting the high 87Sr/86Sr (the “EM2” component) endmember likely experienced significant loss of Sr due to plagioclase fractionation prior to mixing, consistent with the “EM2” component being highly evolved prior to mixing. If extreme plagioclase fractionation of the magma with radiogenic 87Sr/86Sr did occur prior to mixing, then a negative correlation should also be observed in the clinopyroxene space (Fig. 3), and in major and trace element space (Supplementary Figs. 2 and 3). Spanning compositions varying from trachybasaltic to trachyandesitic in a single dredge: curved lines define mixing in the ALIA-D115 lavas, while the rest of the Samoan lavas show zero-slope fractional crystallization trends. The Sr, Nd, and major element oxide concentrations, as well as the 87Sr/86Sr and 143Nd/144Nd of the two endmembers that anchor the ALIA-D115 mixing array are reconstructed using a mixing analysis. Note that we do not use the clinopyroxenes for reconstructing the endmembers because they show ill-defined mixing trends with respect to major and trace elements. This is likely due to (1) fractionation of trace elements during crystal growth and (2) the clinopyroxenes being zoned such that averaged spot analyses of trace element concentrations (determined by averaging multiple LA-ICP-MS spots on each crystal; see “Methods”) will not correspond directly with grain surface major elements (obtained by extracting average major element concentrations from EPMA maps; see “Methods”) and will thus produce spurious trends, especially when compared to radiogenic isotopes (which were measured on dissolved crystals). However, these issues will not affect the isotopes, which is why the mixing

Figure 3 shows geochemical data from the ALIA-D115 dredge whole rock lavas (red symbols) in addition to shield lavas (orange symbols) and pillow glasses (purple symbols) from neighboring Samoan islands and seamounts. Mixing is clearly defined by the ALIA-D115 dredge lavas in 87Sr/86Sr versus element concentration space (Fig. 3), and in major and trace element space (Supplementary Figs. 2 and 3). Spanning compositions varying from trachybasaltic to trachyandesitic in a single dredge: curved lines define mixing in the ALIA-D115 lavas, while the rest of the Samoan lavas show zero-slope fractional crystallization trends. The Sr, Nd, and major element oxide concentrations, as well as the 87Sr/86Sr and 143Nd/144Nd of the two endmembers that anchor the ALIA-D115 mixing array are reconstructed using a mixing analysis. Note that we do not use the clinopyroxenes for reconstructing the endmembers because they show ill-defined mixing trend with respect to major and trace elements. This is likely due to (1) fractionation of trace elements during crystal growth and (2) the clinopyroxenes being zoned such that averaged spot analyses of trace element concentrations (determined by averaging multiple LA-ICP-MS spots on each crystal; see “Methods”) will not correspond directly with grain surface major elements (obtained by extracting average major element concentrations from EPMA maps; see “Methods”) and will thus produce spurious trends, especially when compared to radiogenic isotopes (which were measured on dissolved crystals). However, these issues will not affect the isotopes, which is why the mixing

Figure 1 143Nd/144Nd versus 87Sr/86Sr of new ALIA-D115-18 single clinopyroxene data and prior data on the whole rock and bulk (100s of grains of) clinopyroxene. Fields for Samoan shield lavas (excluding lavas from Savai’i Island), and global OIB and mid-ocean ridge basalts, are shown in orange and gray fields, respectively. Other Samoan shield lavas from the ALIA dataset (i.e., submarine shield lavas from Savai’i Island) are shown for reference as well (i.e. ALIA-D114: orange triangles, ALIA-D115: red squares, ALIA-D118: orange circles, and ALIA-D128: lavas: orange diamonds, and the new clinopyroxene data, denoted as yellow crossed squares, collected in this study)7. The inset shows a zoomed view of the clinopyroxene data in addition to the bulk clinopyroxenes analyzed (representing 100s of pooled grains; gray squares with an “x”) and the ALIA-D115-18 whole rock (red square) published previously7,8. All error bars smaller than symbols except where shown and represent 2σ standard errors. Note that only nine of the 17 clinopyroxenes analyzed for 87Sr/86Sr also have 143Nd/144Nd data, so not all clinopyroxenes analyzed for 87Sr/86Sr can be shown on this figure. However, the nine clinopyroxenes analyzed for both Sr and Nd isotopes encompass the full range of 87Sr/86Sr observed in this study. Samoan shield lavas, ALIA lavas, and Savai’i xenolith compositions are from elsewhere5-7,69. The black dashed line is a theoretical magma mixing line derived in this study using binary mixing theory; mixing endmember compositions are provided in Supplementary Table 2, and methods for calculating endmembers are discussed in the text. Each hatch is a 10% increment except for the first two hatches, which are in 5% increments. The green star denotes the EM2-derived, high 87Sr/86Sr, trachytic mixing endmember calculated in this study (see Supplementary Table 2).
plagioclase from the same sample is consistent with magma mixing modeling using MELTS24 Ciem compositions in and linear regression analysis of the ALIA whole rock bulk requires initial estimates for the MgO content of the endmembers, trends (i.e., to reconstruct the composition of both endmembers), Table 2 for calculated endmember compositions). In addition, as Supplementary Figs. 4 and 5 for regressions, and Supplementary comparing SiO2 versus MgO, for example (see Supplementary mixing array formed by the ALIA-D115 lavas as observed when endmembers given the limited variability at both ends of the ab initio, because it can be best constrained in both mixing

Given an initial assumption of 10 wt% and 0.5 wt% MgO in the low 87Sr/86Sr (mafic) and high 87Sr/86Sr (silicic) endmembers, respectively (see “Methods” for justification), we calculate 380 p.p.m. Sr and 63 p.p.m. Nd in the high 87Sr/86Sr EM2 endmember magma (see Supplementary Table 2 for composition of two other high 87Sr/86Sr endmember calculations with different starting MgO concentrations) and retrieve values of 87Sr/86Sr = 0.73158 and 143Nd/144Nd = 0.51213 (εNd = −9.8); the calculated major element composition of this EM2 endmember magma, which is trachytic, is provided in Supplementary Table 2. We calculate 712 p.p.m. Sr and 48 p.p.m. Nd in the low 87Sr/86Sr endmember and retrieve values of 87Sr/86Sr = 0.70610 and 143Nd/144Nd = 0.51280 (see Supplementary Table 2). Figures 1 and 3 illustrate mixing curves (black lines) using these values showing good agreement with the ALIA-D115 lavas and the clinopyroxenes of this study.

The percentage of mixing observed in 87Sr/86Sr versus 143Nd/144Nd space (Fig. 1), consistent with the other major element and isotope plots (Fig. 3 and Supplementary Figs. 2 and 3), suggests that the most silica-rich ALIA-D115 lava (sample ALIA-D115-21) is composed of ~70–80% of the calculated EM2 endmember melt and ~20–30% of the mafic melt endmember.

Preservation of isotopic heterogeneity in the magma chamber. Recent work has shown zoning in pyroxene to be a reliable indicator of magma mixing events29,30. Here, Mg and Fe zoning in clinopyroxene is used to assess magmatic residence times post magma mixing (where magmatic residence time corresponds to the time needed to eradicate anomalous compositional lamella in

trend is evident for 87Sr/86Sr and 143Nd/144Nd. The method for defining the endmember compositions using the ALIA-D115 lava trends (i.e., to reconstruct the composition of both endmembers), in terms of major elements and Sr and Nd concentrations, requires initial estimates for the MgO content of the endmembers and linear regression analysis of the ALIA whole rock bulk compositions in C/MgO versus 1/MgO space (where C is the concentration of a major element oxide or a trace element; see detailed description in the “Methods” section). MgO is estimated, ab initio, because it can be best constrained in both mixing endmembers given the limited variability at both ends of the mixing array formed by the ALIA-D115 lavas as observed when comparing SiO2 versus MgO, for example (see Supplementary Fig. 2). In comparing SiO2 versus MgO concentrations, the ALIA-D115 lavas show a linear mixing array that extends from a group of mafic Samoan shield lavas to a hypothetical endmember extending toward 0 wt% MgO. An averaged Samoan shield lava value is used for the MgO concentration of the mafic mixing endmember and we test three different MgO concentrations for the silicic mixing endmember from the lowest value observed in ALIA-D115 lavas (3.8 wt% MgO) to 0.5 wt% MgO (see “Methods” for more details). Further, according to mixing theory, 87Sr/86Sr versus Nd/Sr concentration and 143Nd/144Nd versus Sr/ Nd concentration (companion plots to the hyperbolic mixing relation in 87Sr/86Sr versus 143Nd/144Nd space25) should give rise to linear arrays in a system described by two-component mixing. Thus, linear regressions were performed using these linear arrays, and given the derived Sr and Nd concentrations in each endmember, estimates of 87Sr/86Sr and 143Nd/144Nd were obtained for both endmembers (see “Methods” for more detail, Supplementary Figs. 4 and 5 for regressions, and Supplementary Table 2 for calculated endmember compositions). In addition, as proof of concept, this linear regression method was applied to two other select trace elements (Nb, Th) pertinent to generating a ratio useful for tracking source mantle characteristics (Nb/Th28, see Supplementary Fig. 8 and Supplementary Table 2 for concentrations in the endmembers).
clinopyroxene), given relevant temperatures and diffusivities. The eradication of compositional anomalies in the form of thin growth lamella can be computed following a standard one-dimensional diffusion analysis \(31\) (details are presented in the “Methods” section). Attention is focused on the most mobile elements in clinopyroxene that exhibit zonation and for which diffusivity data exist at the inferred temperature range relevant to their petrogenesis. Fe/Mg inter-diffusion in clinopyroxene is ideally suited in this analysis, and we select two magmatic temperatures for analysis: 1200 °C \((\log_{10} \frac{D_{Mg/Fe}}{D_{Fl/Fe}})_{\text{inter-diffusion}} = -18 \text{ m}^2/\text{s}^{32}\), and 1150 °C \((\log_{10} \frac{D_{Mg/Fe}}{D_{Fl/Fe}})_{\text{inter-diffusion}} = -19.4 \text{ m}^2/\text{s}^{33}\). The thinnest Fe and Mg lamellae observed in clinopyroxene is about 15 μm thick (see Supplementary Fig. 6). The preservation of Fe and Mg lamellae in clinopyroxene suggest magmatic residence times of <45 years at a temperature of 1150 °C to <2 years at 1200 °C. Although there are uncertainties in the assumed temperature and in the experimental values of Fe/Mg inter-diffusion in clinopyroxene, we argue these estimates are accurate to within an order of magnitude, and potentially within 20–30%, based on experimentally determined activation energies and reasonable estimates for magma temperatures as constrained by phase equilibria \(34\). The elemental concentration field observed in the clinopyroxenes exhibit an irregular zoning pattern, and along with the \(^{87}\text{Sr}/^{86}\text{Sr}\) zoning in plagioclase from Edwards et al.\(^{16}\), are suggestive of at least one magma mixing event. The preservation of elemental zoning in clinopyroxene suggests that the eruption occurred relatively soon after magma mixing. The short timescales between mixing of geochemically distinct melts and the eruption of the mixture was sufficiently short for preservation of the heterogeneous \(^{87}\text{Sr}/^{86}\text{Sr}\) and \(^{143}\text{Nd}/^{144}\text{Nd}\) in the magmatic clinopyroxenes. Hence, the observed isotopic heterogeneity reflects the heterogeneity represented by the two magmas that contributed to the mixture.

### Thermodynamic modeling of the evolution ALIA-D115 lavas

Using the two endmember compositions described above—the mafic, low \(^{87}\text{Sr}/^{86}\text{Sr}\) and the trachytic, high \(^{87}\text{Sr}/^{86}\text{Sr}\) endmembers—thermodynamically constrained mixing simulations using MELTS\(^{24–26}\) and the MCS\(^{27}\) at ~20 MPa best capture the major element and isotope characteristics of the Samoan lavas (see Supplementary Figs. 2 and 3) and validate the mixing endmembers proposed in this study. In addition, the mixing calculations approximately reproduce the dominant mineralogy of clinopyroxene (Mg\(_{71–80}\)) and plagioclase (~An\(_{45}\); natural plagioclase range from An\(_{50–60}\) (ref. \(^8\)) as observed in the ALIA-D115-18 lava (see “Methods” for model parameterizations). It is important to note that the MCS mixing lines plotted in Supplementary Figs. 2 and 3 (plotted in gray) are not expected to follow the same path in compositional space as the closed-system binary mixing models, nor end at the “EM2”-derived endmember (see “Methods”). This is because the former tracks the predicted melt composition whereas the latter tracks the bulk composition of the mixture. It is expected that mixing of two (or more) magmas would produce crystal populations with heterogeneous major, trace, and \(^{87}\text{Sr}/^{86}\text{Sr}\) and \(^{143}\text{Nd}/^{144}\text{Nd}\) compositions since the hybridization (complete mixing or homogenization of two or more magmas) of the mixing endmember magmas is not instantaneous and most likely incomplete, even after decades of magma co-mingling\(^{13}\) (where co-mingling refers to incomplete mixing between two or more magmas producing discrete compositional bands in the actively mixing magmas).

### Origin of EM2 and derived melts

The petrogenetic modeling presented here shows that a silicic endmember EM2 magma is...
consistent with the geochemical array formed by the ALIA-D115 lavas from Samoa. An important outcome of this effort is the determination that the extreme EM2 magma endmember in Samoa is trachytic in composition. The origin of the EM2 endmember is still an important question to be answered. Previous work has shown that even though EM2 has high $^{87}\text{Sr}/^{86}\text{Sr}$ and EM2 lavas at Samoa are relatively evolved, EM2 is not the result of shallow contamination of crustal material or sediments. Oceanic crust has insufficiently radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$ to produce the $^{87}\text{Sr}/^{86}\text{Sr}$ observed in the ALIA-D115 lavas, and furthermore, Pb isotopic compositions of modern marine sediments from the Samoan region compared with the ALIA-D115 lavas form a non-overlapping, diverging trend such that the highest $^{87}\text{Sr}/^{86}\text{Sr}$ lavas are furthest from the marine sediment field16. Supplementary Fig. 8 shows that Nb/Th of the ALIA-D115 lavas decreases with increasing $^{87}\text{Sr}/^{86}\text{Sr}$, consistent with magma mixing between a mafic, low $^{87}\text{Sr}/^{86}\text{Sr}$ (high Nb/Th) source and one that has high $^{87}\text{Sr}/^{86}\text{Sr}$ and low Nb/Th ratios (Nb/Th = 1.8 for the low MgO EM2-derived endmember of this study) similar to upper continental crust (Nb/Th = 1.1 (ref. 35)) or average sediments (Nb/Th = 1.2 (ref. 36)). However, as stated above, neither of which are modern materials, but rather ancient subducted materials.

While the presence of this trachytic endmember (with $^{87}\text{Sr}/^{86}\text{Sr}$ up to 0.73158) is inferred from the mixing calculations presented in this study, it has (not yet) been found in pure form in lavas examined from the ALIA-D115 dataset. However, geochemical interrogation of the highest $^{87}\text{Sr}/^{86}\text{Sr}$ Samoan lavas—ALIA-D115-18 and ALIA-D115-21 (with whole rock $^{87}\text{Sr}/^{86}\text{Sr}$ of 0.718592 and 0.720469, respectively)—reveals that single clinopyroxene crystals ($^{87}\text{Sr}/^{86}\text{Sr}$ up to 0.723888; this study, Supplementary Table 1) and zones in plagioclase ($^{87}\text{Sr}/^{86}\text{Sr}$ up to 0.7224) sample even more extreme EM compositions than the host whole rocks and trend toward the endmember EM2 melt calculated here. More intriguingly, reports of high-SiO$_2$ (65 to 69 wt%) trachytic melt inclusions51, hosted in the same ALIA-D115-18 clinopyroxene grains examined in this study raise the possibility that the EM2 endmember may be preserved in relatively pure form as glassy melt inclusions in the clinopyroxenes from a high $^{87}\text{Sr}/^{86}\text{Sr}$ lava (see Supplementary Table 2 for example melt inclusion compositions). Unfortunately, these melt inclusions are too small for analysis of $^{87}\text{Sr}/^{86}\text{Sr}$. Nonetheless, the presence of trachytic melts representing the EM2 endmember composition raises important questions about the origin of melts sampling this source. The fact that the model presented here indicates ~20% of a mafic magma mixed with ~80% trachytic EM2 endmember magma suggests that the potential conditions of mixing of silicic and mafic magmas may be due to an initially isolated silicic magma body that was recharged by a more mafic magma, where clinopyroxene and plagioclase crystallization (and melt inclusion entrapment) occurs during the mixing process. This scenario is consistent with the highly variable nature of $^{87}\text{Sr}/^{86}\text{Sr}$ observed in the clinopyroxene and plagioclase crystals. The question still remains, what is the origin of these silicic, trachytic melts?

Previous studies provide evidence of silicic melts in the mantle derived by various processes, such as low-degree partial melting of anomalous mantle, reaction between CO$_2$-rich fluids/melts or basaltic to silicic melts and peridotite, or as the result of extensive fractional crystallization5,38-40. One hypothesis tested in this study is partial melting of geochemically enriched (with high $^{87}\text{Sr}/^{86}\text{Sr}$) mantle compositions followed by crystal fractionation to produce trachytic melts. Considering a scenario in which ancient subducted sediment has infiltrated and mixed with ambient peridotite, partial melting of this source could give rise to silicic melts with radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$. Results (in detail in the Supplementary text) show that partial melts (at a range of pressures) of various mixed sediment ± peridotite sources are consistently basanitic (high-degree partial melts) to phonolitic (low-degree melts) and subsequent fractionation of these melts does not produce trachytes. However, we acknowledge that we have not undertaken an exhaustive search of parameter space (e.g., different sediment compositions, different combinations of sediment ± recycled oceanic crust ± peridotite, various H$_2$O and CO$_2$ source concentrations, and a full range of fO$_2$, pressures, and temperatures), and future work will focus on melting and fractional crystallization processes in the petrogenesis of compositions similar to the EM2 trachytes presented here.

A scenario in which trachytic melts are formed directly through partial melting of a sediment-like source and subsequently mixed with more mafic Samoan melts to form the ALIA-D115 mixing trend is also possible. However, silicic melts are highly reactive with ultramafic bulk compositions and a transport scenario in which large volumes of this melt travel through 10s of kilometers of ocean mantle lithosphere to shallow magma chambers and remain isolated, requires further study (see Adams et al.49, for a further discussion).

Trachytic melts have been shown to be common products of metasomatic processes such as reaction of ambient peridotite mantle with CO$_2$-rich fluids or silicic melts45,43,47,48,30-52. Many workers have also suggested that partial melting of mantle already modified by metasomatic processes can produce disequilibrium silicic liquids that might vary substantially in composition depending on the source47,48,53. Evidence of carbonatitic and silicic melt metasomatism in Samoan xenoliths4,5,54,55 exists, and a trachytic glass found in one highly carbonatite metasomatized peridotite mantle xenolith from Savai’i Island—the same island from which the lavas of this study are derived—exhibits radiogenic $^{87}\text{Sr}/^{86}\text{Sr}$, but is still much lower than the predicted $^{87}\text{Sr}/^{86}\text{Sr}$ of this study for the EM2-derived endmember. Further, as noted earlier, it remains to be determined how low melt fractions of trachyte melt observed in a xenolith can be scaled up to larger volumes and additionally, it is not clear how to transport this trachyte melt through the mantle, from the melt source to the magma mixing region. This type of petrogenetic mechanism, which invokes carbonatite metasomatism of peridotite to generate the trachytic melt source, may be a robust avenue for future research into the origin of EM2 that could be tested using thermodynamic phase equilibria modeling and/or experimental analysis (see Adams et al.49, for further discussion).

The origin of EM2 and why it manifests in extreme form in Samoa remain important problems: EM2 is identified in lavas from Society and Marquesas Islands17,56, for example, but the magnitude of the geochemical enrichment at these localities is less pronounced than at Samoa. Nonetheless, the new radiogenic isotopic compositions derived in this study support models advocating for a large fraction of recycled terrigenous sediment into the Samoan plume7,16. This further suggests that, since EM mantle domains have been found to correspond geographically with the LLSVPs, sediment-rich domains can survive in the LLSVPs over geologic timescales to be sampled by the Samoan plume.

Methods

RMS analysis

Sample preparation. Seventeen clinopyroxene grains lacking visible attached groundmass or surface alteration were plucked from indium mounts. The clinopyroxenes were then leached in concentrated HF$\alpha$ at 90 °C for 2 min and then washed once with milli-Q (18.2 × 10$^6$ Ω cm) water; this process was repeated twice to ensure that any indium remaining on the grains was removed. The clinopyroxenes were then leached in 6N HCl at 90 °C for 2 min to remove any remaining surface contamination, then they were rinsed three times with milli-Q water. Seventeen clinopyroxene samples, together with two aliquots of USGS Reference
Material BCR2 hosting 10.8 ng Sr and 0.88 ng Nd (the first aliquot), and 5.6 ng Sr (the second aliquot, but Nd was not analyzed), respectively, were individually transferred to separate vials containing 150 µl of concentrated HF for dissolution. The samples were then set on a 120 °C hotplate for 2 days for dissolution. All clinopyroxenes and BCR2 aliquots were spiked with 84Sr and 149Nd for determination of the total amount of analyte in the sample by isotope dilution. Samples were then dried down and later brought up in a 500 µl concentrated HNO3 solution and placed on a 120 °C hotplate for 24 h to eliminate fluorides. Following dry-down, the samples were brought up in 1 ml of 3 M HNO3 to load on to columns for chemical separation of Sr and Nd. Column chemistry follows Koornneef et al. Total procedural blanks were processed through all steps of sample dissolution (beginning with dissolution in HNO3, HF, column separations, and mass spectrometry). Total procedural blanks (including all stages of wet chemistry, e.g., sample dissolution, column chemistry, etc., and loading on TIMS filaments) varied from 22 to 60 pg for Sr (and averaged 45 pg in one session and 37 pg in the other), and 2.2 to 2.7 pg for Nd. The Sr and Nd blanks are dwarfed by the total amount of Sr (~5.6 to 58.7 ng) and Nd (~0.88 to 25.6 ng), respectively, in each clinopyroxene grain and the two BCR2 aliquots. The [Sr]sample/[Sr]blank and [Nd]sample/[Nd]blank ratios for each sample are provided in Supplementary Table 1. Thus, while blanks corrections are applied to the samples and the BCR2 aliquots (assuming a lab blank 87Sr/86Sr = 0.711 and 143Nd/144Nd blank of 0.5118, obtained by pooling multiple blanks), blank corrections to the 87Sr/86Sr and 143Nd/144Nd ratios are negligible.

87Sr/86Sr and 143Nd/144Nd isotope analyses. All of the clinopyroxene grains were processed through the same batch of column chemistry with a BCR2 and two total procedural blanks; two batches of chemistry were required for Sr isotopes (so two BCR2 aliquots were analyzed), and one batch of chemistry was required for Nd isotopes (so one BCR2 analysis for Nd is reported). The clinopyroxene grains and the BCR2 aliquots were analyzed with 87Sr/86Sr and 143Nd/144Nd over two analytical sessions. The separated, dried Sr samples were brought up in 1 ml of HNO3, and each loaded onto outgassed, zone-refined rhenium (99.999%) filaments (H Cross, USA) along with a 1 µl TcO2, emitter solution. For each analytical session, two total procedural blanks were also loaded onto rhenium filaments in addition to a BCR2, all of which passed through the same column chemistry as the clinopyroxene and BCR2 unknowns. In addition, eight filaments with 1 ng NBS987 were analyzed together with the clinopyroxenes and BCR2 aliquots: average 87Sr/86Sr = 0.702975 ± 0.000016 (2 SD, N = 8). All samples were analyzed by static Faraday collection without amplifier rotation on UCSC’s Triton Plus employing 1013 Ω amplifiers and using a 3.3 picoamp gainboard, and gains were measured every barrel. Samples were corrected for the offset between the preferred (0.70240) and the average measured NBS987 87Sr/86Sr from the same analytical session (i.e., the same batch of chemistry); Sr isotopes were corrected for mass bias assuming an exponential law and using canonical 87Sr/86Sr ratio of 0.1194, and isobaric interferences from Rb were corrected by monitoring mass 85, but changes to the 87Sr/86Sr ratios due to this correction were negligible.

Only nine of the above 17 clinopyroxene grains were analyzed for 143Nd/144Nd; these were selected after 87Sr/86Sr was analyzed and were chosen because they spanned the range of measured 87Sr/86Sr values. The dried, separated Nd samples were brought up in 4 µl of 1 M HNO3 and loaded onto outgassed, zone-refined (99.999%), H Cross, USA) double rhenium filaments. Two blanks, six 1 ng NBS987’s (average 0.512106 ± 0.000033, 2 SD), one 500 ng NBS987 (0.512112 ± 0.000028, 2SE), and a BCR2 (run through the columns) were also loaded onto rhenium filaments and run during the same analytical session (i.e., the same barrel) as the clinopyroxene samples. Samples were corrected for the offset between the preferred (0.512099) and the average measured NBS987 143Nd/144Nd from the same analytical session. All samples were analyzed by static Faraday collection without amplifier rotation. Plus employing 1013 Ω amplifiers and using a 3.3 picoamp gainboard (gains are measured once, at the start of the new barrel). Nd isotopes were corrected for mass bias assuming an exponential law and using a canonical 143Nd/144Nd ratio of 0.7219. Isobaric interferences from Sm were negligible as they were filtered out of the dataset prior to averaging.

Clinopyroxene trace element concentrations were collected at the University of California Santa Barbara (Supplementary Table 1) by LA-ICP-MS using a Photon Machines Excite 193 laser illuminator coupled to an Agilent 7700 quadrupole ICP-MS. A 15-µm spot diameter was used. Unknowns were corrected relative to reference material NIST612 analyzed every 8–10 unknown analyses; analyses and the 2RSID on each element can be found in Supplementary Table 4. Each trace element measurement reported in Supplementary Table 1 is an average of three to four spot analyses spread across each clinopyroxene grain.

Residence times from diffusion. To obtain an estimate of the residence time, defined as the time interval between crystal growth and lava eruption (quenching), of the clinopyroxene porphyroclasts, the diffusive characteristic of lamellar compositional bands is considered. Examination of compositional profiles revealed that thin lamella of locally higher MgFeg were preserved (Supplementary Fig. 6). Based on Fe-Mg inter-diffusion, we used the thickness of these lamella to obtain an estimate of the maximum residence time of the crystal. That is, if diffusion was active for a period of time greater than the residence, the compositional anomaly would have been erased. In this one-dimensional diffusion model, a lamella of thickness 5.6 ng Nd aliquots spiked (0.50 to 0.88 ng), 0.50 to 0.88 ng aliquots spiked with 10.8 ng Sr oxides, trace elements, isotopes), the mixing equations may be recast in the ratio-normalization form. Examination of compositional pro-

\[ N = \frac{k}{\partial t} \]

where the non-dimensional variables are defined as:

\[ C = \frac{C_2 - C_1}{C_2}, \quad x = \frac{x}{b}, \quad \tau = \frac{D}{b^2} \]

Based on this solution, we computed the time necessary to eradicate 90% of the compositional anomaly at the centerline of the lamella (see ref. 18, pg. 101 for graphical illustration) of the thinnest lamella in the clinopyroxene: 15 µm (b = 7.5 µm). Diffusion calculations were performed at 1150 °C (log10 D MgFe inter-diffusion = −19.4 m2/s33) and 1200 °C (log10 D MgFe inter-diffusion = −18 m2/s32). The effect of varying temperature ±50 K is modest, increasing or decreasing residence times by about 20–30%. The greatest uncertainty is in the laboratory determination of diffusivities.

Retrieval of magma mixing endmembers. The principles of magma mixing are well established and have been used for decades to decipher magma mixing from assimilation and/or fractionation processes in natural magmatic systems.23,25–36. The Samoan ALIA whole rock lavas discussed in this study are very likely a result of magma mixing as noted by examination of Fig. 3. The mixing trends can be used to estimate the composition of the mixing endmembers given the MgO content of the siliceous (S; high 87Sr/86Sr) and mafic (M, low 87Sr/86Sr) mixing endmembers. Based on binary mixing theory,23,35,36, for any two chemical species (major oxides, trace elements, isotopes), the mixing equations may be recast in the ratio-re.

\[ C'_M = A + B \]

where \( C'_M \) and \( C'_F \) are concentrations of chemical species 1 and 2 in the mixed (hybrid) magma, respectively, and A and B are constants related to the chemical
species concentrations in endmembers M and S. The constants A and B in Eq. (3) are defined from mixing theory to be:

\[ A = \frac{C_1^M - C_2^S}{C_1^M - C_2^M} \]  

and

\[ B = \frac{C_1^S - C_2^M}{C_1^S - C_2^S} \]  

where \( C_1^M \) and \( C_2^M \) are concentrations of chemical species 1 and 2 in the mafic (M) endmember and \( C_1^S \) and \( C_2^S \) are the concentrations in the silicic (S) endmember. The linear regression of the petrochemical data for the ALIA-D115 lavas gives the intercept and slope that corresponds to A and B. As A and B are two equations with 4 unknowns, thus if the concentration of chemical species 1 is known, or can be confidently estimated, in both endmembers, then the concentration of chemical species 2 in both endmembers can be calculated by simple rearrangement of Eqs. (4) and (5). The quality of the computed parameters depends on the quality of the fit of the data to Eq. (3). We have chosen to initially fix values of MgO in both M and S because there is relatively little variability in MgO concentrations in both endmembers based on the array defined by the ALIA-D115 lavas. In other words, a plot of Samoan lavas in SiO2 versus MgO space (see Supplementary Fig. 2), shows the linearity expected for mixing, as the ALIA-D115 lavas form a trend with the M endmember, sitting in the cloud of data points of “other Samoan shield lavas” and the S endmember extending toward 0 wt% MgO, thus, the MgO in the S mixing endmember must lie between the value of the most evolved high 87Sr/86Sr and the silicic endmember was 712 p.p.m. and 380 p.p.m. in the silicic endmember. If we had instead performed the same calculation with Sr/MgO versus 1/MgO, the correlation coefficient is 0.85 and the Sr concentration in the mafic endmember would be 726 p.p.m. and the silicic endmember would be 360 p.p.m. These differences are comparable to analytical uncertainties for real measurements.

MCS mixing modeling. Using the calculated mafic (M; low 87Sr/86Sr) and silicic (S; high 87Sr/86Sr) mixing endmembers from the above procedure, MELTS14,25 and the MCS27 were used to simultaneously model the effects of magmatic recharge and fractional crystallization processes on Samoan lavas (RFC in MCS jargon). The metric used to gauge model acceptability was how well the phase equilibria and geochemistry matches the petrology and petrochemistry of the observed Samoan ALIA-D115 lava suite. In the magma mixing calculations, the M endmember composition was the host magma and the S endmember composition, based upon an assumed MgO content of 0.5 wt% (see Supplementary Table 2 for details), was the recharge magma. The H2O content of both M and S endmembers was set at 0.1 wt% along with 1% K2O and 1% magnetite-fayalite buffer establishing the ferro-ferrous iron ratio in each endmember. Since the porphyroclastic clinopyroxenes of this study are in isotopic disequilibrium with the whole rock, clinopyroxene-liquid thermobarometry is not meaningful. Because an independent pressure estimate could not be made, MCS simulations were performed in the range 0.1 to 1 GPa. Upon mixing at low pressures (<0.5 GPa), the stable fractionating mineral assemblage consists of olivine, clinopyroxene, plagioclase, and spinel. Simulations were run for three cases where the high 87Sr/86Sr recharge magma (S endmember) was initially either: (1) 100% liquid, (2) possessed a crystallinity of about 15% (15% An21Ab60Or15, anorthoclase feldspar, modally) and (3) possessed a crystallinity of 10% An50Ab40Or10, allanite feldspar, 23% An23Ab76Or1, plagioclase feldspar, 5% spinel, and 2% orthopyroxene, modally) upon addition to the host magma (M endmember). At constant pressure, the difference between these three initial recharge magma thermodynamic states is that as the crystallinity of the recharge magma increases the initial anorthite content and Mg# of the plagioclase and clinopyroxene, respectively, that crystalize from the host magma increases upon the initiation of mixing. As an example, at 0.2 GPa, when the recharge magma was 100% liquid, plagioclase (An50) was stabilized at 1107 °C whereas when the recharge magma was 60% crystalline, An29plagioclase was stabilized at ~1119 °C. In the case of the ALIA-D115 lavas, the latter case more closely corresponds to observations in plagioclase within the ALIA-D115 lavas. With increasing pressure, the first plagioclase crystalizes from the magma decreased in An content and there was increased stabilization of orthopyroxene in the assemblage over clinopyroxene, which is not observed in the ALIA-D115 lava suite. Thus, MCS simulations favor lower pressures of magma mixing and a partly crystalline recharge magma at the time of mixing. Overall, the AlIA-D115 lavas modeling results in Supplementary Fig. 1 in Eq. (3) yield a line based on closed system binary mixing. In any regard, the MCS mixing lines plotted in Supplementary Figs. 2 and 3 are the evolutionary paths of the melt as a result of magma mixing and crystallization (crystals are removed from the system as soon as they form), so the bulk composition is not conserved and thus, one would not expect the mixing lines to end at the EM2-derived endmember, much different than the conservative mixing line based on closed system binary mixing. In addition, the MCS models are thermodynamically constrained models and so the terminus of the mixing line is dictated by where the simulation terminated. In any regard, the ALIA-D115 lavas can be explained reasonably well given the ubiquitous geological uncertainties in any modeling exercise.

Data availability
All data generated in this study are available in the Supplementary data files of this article and can also be found on the IEDA EarthChem online database at the following link: https://doi.org/10.26022/IEDA/111769. Source data for figures can be found within the Supplementary Data files.

Code availability
The current Magma Chamber Simulator (MCS) source code, details on how to operate MCS, and numerous applied examples can be found at https://mcs.geol.ucsb.edu/. Also see Bohrson et al.27, Bohrson et al.29, and Hennonen et al.38 for more details on MCS operations and applied examples.

Received: 26 March 2020; Accepted: 21 January 2021; Published online: 23 February 2021

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**Acknowledgements**

J.V.A. acknowledges the GSA graduate student research grant that partially funded the TIMS isotope analysis. M.G.J. acknowledges NSF grants OCE-1736984, EAR-1429648, EAR-1900652. We thank Stan Hart and Hubert Staudigel for contributing sample material.

**Author contributions**

J.V.A. performed the theoretical modeling, mixing calculations, much of the analytical work, and wrote the manuscript. M.G.J. provided the samples and conceived the project idea. F.J.S. helped formulate the binary mixing and diffusion calculations. M.G.J. and F.J.S. contributed significant additions and edits to the manuscript. Clinopyroxene sample preparation and TIMS analyses were performed by J.V.A., A.A.P. and B.L.B. Clinopyroxene EPMA major element maps and data analysis was performed by G.S. and J.V.A. Clinopyroxene trace element analyses were performed by J.V.A. with data reduction and analysis help by J.M.C. All authors participated in discussion and/or interpretation of results and gave valuable feedback on the manuscript.

**Competing interests**

The authors declare no competing interests.

**Additional information**

Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s41467-021-21416-9.

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**Peer review information** *Nature Communications* thanks David Graham and Janne Koorneef for their contribution to the peer review of this work. Peer reviewer reports are available.

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