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This paper was submitted to *Nature Communications* on May 3, 2024, and is currently under review. Later versions of this manuscript may have slightly different content. If accepted, the final version will be available via the 'Peerreviewed Publication DOI' link on the right-hand side of this page.

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## **Seamounts control subducted carbonate recycling in Central America – evidence from stable Sr isotopes**

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## **Seamounts control subducted carbonate recycling in Central America – evidence from stable Sr isotopes**

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#### **Abstract**

 **Accurate estimates of carbon recycling efficiency at subduction zones are crucial for understanding the long-term evolution of the planet's climate. Estimating subducting carbon recycling efficiency is difficult, however, due to magmatic degassing deep within the crust. Stable strontium (δ<sup>88</sup> Sr) isotopes have the potential to serve as an additional proxy for carbonate recycling due to carbonate's high Sr concentrations and unique δ <sup>88</sup> Sr composition. New data from Nicaraguan lavas and sediments verify that Sr can indeed be used to track subducting carbon. When interpreted in conjunction with other magmatic trace element abundances, these data reveal systematic variations that are best understood as the effects of seamount subduction. Seamounts erupting at the same time carbonate-dominated sedimentation ceased on the Cocos plate could potentially 'cap' the carbonate layer and control the degree to which carbonate material is transported to the arc. This model can also be used to quantify the flux of carbonate sediment from the slab to the arc and thus provides an independent method to estimate the magmatic carbon recycling efficiency for the Nicaraguan Arc. Our model shows, absent seamounts, 26% to 52% of carbon from subducted carbonate is returned to the arc, an estimate consistent with recent estimates made from volcanic gases.**

18 **Introduction**

19 Carbon cycling in subduction zones has played a key role in maintaining the planet's 20 climate and long-term habitability<sup>1-5</sup>. Some fraction of the carbon initially subducted is quickly 21 returned to the surface through volcanic arcs, with the remainder transported to the deep 22 mantle. The proportion of initially subducted carbon that is returned from the slab to arc is 23 often referred to as the "carbon recycling efficiency." Though accurate estimates of the carbon 24 recycling efficiencies of volcanic arcs are essential to our understanding of Earth's planetary 25 evolution, recent estimates range widely, from  $14\%$  to  $100\%$ <sup>1,6-19</sup>. This range persists largely 26 because arc magmas lose most of their initial carbon budget to degassing deep in the crust. 27 Even melt-inclusions hosted in primitive olivines likely sample liquids that have lost  $CO<sub>2</sub>$  prior to 28 entrapment<sup>20</sup>. As such, direct sampling of the primary arc magma carbon concentrations or 29 isotope ratios may not be possible in most cases. 30 Most estimates of carbon recycling efficiency are instead derived from measurements 31 of CO<sub>2</sub> abundances and/or isotope ratios of volcanic gases<sup>9-11,13</sup>, which also have limitations. For 32 example, carbon fluxes at volcanic arcs are spatially and temporally variable, and the extent of 33 diffuse degassing is not usually well constrained<sup>10-12,19</sup>. Carbon isotopes can also fractionate 34 during slab devolatilization and volcanic degassing $^{21}$ , crustal contamination, and shallow 35 processes such as calcite precipitation in the forearc $^{11}$ .

36 Here, we investigate the potential of Sr stable isotope ratios as a tracer of carbonate 37 recycling from a subducting slab to a volcanic arc using high-precision double-spike stable Sr 38 isotope (Sr isotope data are reported as <sup>88</sup>Sr/<sup>86</sup>Sr in standard delta notation, relative to NIST 39 SRM-987 in units of permil as: δ<sup>88</sup>Sr (‰)= [(<sup>88</sup>Sr/<sup>86</sup>Sr)<sub>sample</sub>/(<sup>88</sup>Sr/<sup>86</sup>Sr)<sub>SRM987</sub> −1]x 1000) and

40 radiogenic <sup>87</sup>Sr/<sup>86</sup>Sr measurements of Cocos plate sediments and arc lavas from Telica, Cerro Negro, and Masaya volcanoes in the Nicaraguan segment of the Central American Volcanic Arc. These Sr isotope data, in conjunction with traditional radiogenic isotopes and trace element abundances, provide a novel way to quantify carbon recycling efficiency. Our results also motivate a re-evaluation of the origins of geochemical variability present among Central American arc lavas, which appears to be importantly regulated by subducting seamounts. The Central American Volcanic Arc (Fig. 1a) extends ~1400 km from the Mexico- Guatemala boarder to Panama, where the Cocos Plate subducts underneath the Caribbean at a 48 rate of 5.6 - 6.4 cm/yr<sup>22</sup>. The subducting plate consists of 20 Ma oceanic crust produced at the East Pacific Rise and carries numerous seamounts with elevations up to 2000 m above the 50 seafloor and diameters of up to 14  $km^{23}$ . The ocean floor offshore of Central America is covered by two sediment packages of similar thickness: an upper hemiplegic unit and a lower unit 52 dominated by pelagic carbonate (Fig. 2a and b) $^{24-28}$ . The mid-Miocene carbonate crash led to a 53 rapid shift in sediment composition between these layers<sup>29</sup>. Cores from Deep Sea Drilling Program site 495 off Guatemala and Ocean Drilling Program site 1039 off the Nicoya Peninsula of Costa Rica (Fig. 1a) show that the sediments remain uniform in thickness and distribution 56 along the entire length of the arc<sup>26,30</sup>.

 As with all arc-front stratovolcano lavas, the volcanic rocks of the Central American volcanic front inherited the distinct geochemical signatures of subducted materials. Studies of 59 the Northern Guatemala-El Salvador<sup>28</sup> and Southern Costa Rica-Panama<sup>31,32</sup> segments have also identified isotope compositions that appear to originate from the lithospheric mantle and the Galapagos hotspots, respectively. The volcanic rocks of Nicaraguan segment, by contrast,

62 mostly reflect a depleted MORB mantle (DMM) source overprinted with the incompatible trace 63 element pattern of a typical arc lava (e.g. enrichment of Rb, Ba, Th, U, Pb, and Sr and depletion 64 in Nb, Ta)<sup>26</sup>. The upper plate of the Nicaraguan segment is thin and mostly oceanic in origin 65 with little evidence for crustal assimilation<sup>33</sup>. A comparison between thermo-mechanical 66 models of the slab beneath Nicaragua and experimentally determined solidus temperatures<sup>34,35</sup> 67 (Fig. 1b) indicates that at least the upper 1 km of the slab reaches temperatures that exceed the 68 H<sub>2</sub>O-saturated solidi for carbonate and pelitic sediments<sup>35-37</sup>, altered oceanic crust (AOC)<sup>38</sup>, and 69 unaltered MORB<sup>39</sup>. A similar comparison shows that progressive lawsonite dehydration in the 70 lower slab layers should provide a continuous source of water to the slab top (Fig. 1b)<sup>35</sup>. This 71 suggests that hydrous slab melting is the dominant process by which subducting materials are 72 transferred to the mantle wedge in the Nicaraguan segment, and that carbonate melting may 73 mediate the  $CO<sub>2</sub>$  flux from the slab to the arc.

74 Our current understanding of carbon fluxes out of the Central American arc are mostly 75 derived from measurements of volcanic gases and fluid seeps. Most estimates of arc  $CO<sub>2</sub>$  fluxes 76 utilize direct measurements of fumarole  $CO<sub>2</sub>/<sup>3</sup>$ He or  $CO<sub>2</sub>/SO<sub>2</sub>$  alongside an assumed <sup>3</sup>He flux or 77 an independently measured  $SO<sub>2</sub>$  flux. Using this approach, Shaw et al. (2003) estimated that the 78 carbon flux from Costa Rican and Nicaraguan arc segments accounted for only ~15% of the total 79 subducting carbon budget. Carbon fluxes estimated by Mather et al. (2006) and Aiuppa (2014) 80 are similar to that of Shaw (2003), while de Moor et al. (2017)'s study of carbon outgassing 81 from 2015-2016 are much higher, indicating carbon recycling efficiencies as high as ~76%. The 82 increased carbon recycling efficiency estimate of de Moor et al. (2017) could be due to 83 improved methodology and much denser sampling, but also coincided with a net increase in

84 volcanic activity during their study period. This suggests that arc  $CO<sub>2</sub>$  fluxes can be highly variable across years-long timescales. Recent work by Barry et al. (2019) measured deeply sourced hot springs along two across-arc transects in Costa Rica and estimated that an 87 additional ~19% of subducted carbon may be lost from the slab beneath the forearc. Their total 88 slab-derived carbon outputs are consistent with recent global carbon recycling efficiency 89 estimates of 46%-65%<sup>8,15</sup>.

 Helium and carbon isotope ratios in gas samples can also be used to determine the relative contributions of subducting materials to the overall carbon budget of arcs. Isotope studies of Central American gases largely agree that most of the carbon emitted through the 93 arc originates from subducting carbonate<sup>9,40,41</sup>, consistent with the fact that the sedimentary carbonate layer dominates the carbon budget of the subducting slab.

 Trace element abundances in Central American lavas serve as additional proxies for 96 carbonate and hemipelagic sediment recycling from the slab to the arc<sup>26</sup>. This is possible because the hemipelagic sediment has high concentrations of most incompatible trace elements, while the carbonate layer is diluted in most trace elements aside from Sr, Ba, La, and Pb (Fig. 2a), leading to distinct downcore variations in trace element ratios such as Ba/Th and U/La (Fig. 2b). Central American arc lavas exhibit correlated variations in Ba/Th and U/La, presumably due to varying recycling efficiencies of these sediment layers. In some arcs, these ratios could be controlled by the addition of aqueous fluids from the slab, which fractionate 103 "fluid mobile' elements such as Ba and U from 'fluid immobile' elements such as Th and  $La^{42}$ . The negative correlation between these ratios among Nicaraguan lavas (Fig. 3) is inconsistent with aqueous fluid control for the Nicaraguan arc, however, and accords well with variance

 expected for contributions from the different sediments. The hypothesis that the incompatible trace element compositions of these lavas are controlled by variable subducting sediment 108 compositions is further confirmed by a correlation<sup>43</sup> between U/La and  $^{10}$ Be/ $^{9}$ Be<sup>33,44</sup>.

 These trace element proxies show that the sediment recycled from the slab to the arc must be highly variable even though core samples all reflect similar offshore sediment profiles. Patino et al. (2000) postulated that this could be due either to off-scraping of the upper hemipelagic sediments from horsts to grabens or to a more complicated multi-stage slab melting process. Others have proposed mechanisms relating to slab dip, differing extents of lithospheric serpentinization, varying extents of fore-arc erosion, or temporal variability in the 115 spatial distribution of the carbonate sediment layer $10,26,29,45$ .

 While the trace element approach has produced important qualitative constraints on sediment recycling in Central America, these proxies cannot be used to accurately quantify the absolute recycling efficiency of either sediment package. For U/La, this is because U decreases significantly down the entire sediment core (Fig. 2b), including within the hemipelagic layer, as 120 the proportion of preserved organic-rich sediments decreases<sup>26,29</sup>. The relationship between Ba/Th in sediments vs lavas is complicated by the fact that Ba concentrations in pore fluids and 122 sediments offshore of Costa Rica indicate that upwards of ~60% of the incoming bulk sediment 123 Ba may be lost to the forearc<sup>46</sup>. Uncertainties such as these make it difficult to directly estimate the recycling efficiencies of the individual sediment layers from trace element data alone.

 The high Sr concentrations and distinct Sr isotope compositions of carbonates make Sr stable isotopes a promising proxy for recycled carbonate that is free from the uncertainties 127 inherent in gas data or trace element ratios.  $Sr^{2+}$  can readily substitute for Ca<sup>2+</sup>, leading to

128 elevated Sr abundances in most carbonate-rich marine sediments. Seawater inherits 129 terrigenous Sr with high  $\delta^{88}$ Sr (0.39‰)<sup>47,48</sup>, whereas biogenic marine carbonates are typically a 130 mix of foraminifera and coccoliths with much lower  $\delta^{88}$ Sr (0.14‰ to 0.27‰; Fig.4)<sup>47,48</sup>. High-131 precision TIMS measurements of  $\delta^{88}$ Sr in typical MORB samples are limited, but thus far cluster 132 around  $0.28\pm 0.05\%^{49}$ . Experimental work shows that high-temperature hydrothermal 133 alteration of basalt leads to elevated  $\delta^{88}$ Sr<sup>50</sup>, and while actual AOC  $\delta^{88}$ Sr measurements are also 134 limited, existing data support this general process<sup>49</sup>. High-temperature magmatic processes, on 135 the other hand, have little effect on Sr isotope fractionation in systems for which Sr behaves as 136 an incompatible element. Plagioclase fractionation may have a small impact on magmatic 137  $8^{88}$ Sr<sup>51</sup>. The application of Sr to carbonate recycling during subduction also benefits from the 138 fact that Sr and C partition similarly between carbonate and melt<sup>52,53</sup>. Together, these studies 139 show that subducting materials, and marine carbonates in particular, exhibit distinct  $\delta^{88}$ Sr 140 compositions that should be reflected as recycled components in mafic arc lavas. This proxy has 141 the potential to constrain carbon recycling from measurements of lava samples without the 142 complications that can arise from early carbon degassing and the various processes that can 143 fractionate trace element ratios. Stable Sr isotopic compositions thus could provide an 144 alternative method to estimate long-term carbon fluxes at subduction zones. 145 Lavas from the Nicaraguan arc were chosen for this study due to their strong sediment 146 signature and lack of evidence for crustal assimilation or influence from the Galapagos 147 plume<sup>28,29,54,55</sup>. Nine samples were selected to encompass the full range of U/La and Ba/Th 148 found among Nicaraguan lavas (Fig. 3). The samples include lavas from Cerro Negro, Masaya, 149 and Telica volcanoes that were obtained from the Rutgers CAVA sample repository<sup>56</sup> and have

been previously analyzed for major elements, trace elements, and radiogenic isotopes (Pb, Nd,

151 and some  $Sr$ <sup>26,57</sup>. These prior studies showed that the selected samples exhibit compositional

variations that largely reflect slab and mantle processes.

- 
- **Results**

## **Variation in <sup>87</sup>Sr/<sup>86</sup>Sr and <sup>88</sup> Sr in Central American lavas and sediments**

156 Sr/ $86$ Sr and  $88$ <sup>88</sup>Sr data are presented in Table 1 and shown in Figure 5. External 157 precision, reported as the average 2-SE for  $\delta^{88}$ Sr values is <0.02‰. Carbonate sediment  $\delta^{88}$ Sr ranges from 0.170±0.016‰ to 0.219±0.016‰, consistent with the calcareous Aegean 159 sediments ( $\delta^{88}$ Sr=0.195‰ to 0.213‰) measured by Klaver et al. (2020). Surprisingly, the 160 hemipelagic sediment sample is isotopically lighter ( $\delta^{88}$ Sr=0.146‰±0.021‰) than the carbonate, which suggests that the Sr budget of the hemipelagic sediments is also dominated 162 by biogenic material. Both sediments have Sr/ $86$ Sr ratios near that of modern seawater 163 (~0.709), consistent with Patino et al. (2000). Nicaraguan arc lavas ( $\delta^{88}$ Sr=0.212±0.013‰ to 164 0.322±0.015‰) exhibit a greater relative variation in  $\delta^{88}$ Sr than the sediments. Samples from 165 Masaya and Cerro Negro define heavy and light  $\delta^{88}$ Sr end members, respectively, while Telica  $\delta^{88}$ Sr varies between the two (0.243±0.014‰ to 0.279±0.019‰). Arc lava  $87$ Sr/ $86$ Sr ratios (0.7040 to 0.70419) are uniform relative to the range of subducting materials and have compositions closer to DMM or AOC than sediments (Fig. 5). The altered ocean crust composite 169 samples ( $\delta^{88}$ Sr=0.247±0.008‰ to 0.263±0.014‰;  ${}^{87}$ Sr/ ${}^{86}$ Sr = 0.70330 to 0.70679) are consistent within uncertainty to measurements of the same samples by Klaver et al. (2020) but are

 unlikely to represent the compositional range of actual subducting ocean crust as discussed in greater detail in the Online Supplement.

## **<sup>88</sup> Sr provides new insights into Nicaraguan subduction dynamics**

 The primary goal of this study was to assess stable Sr isotopic variability as a proxy for carbonate recycling at subduction zones, but these data also shed new light on important subduction zone systematics that directly influence the geochemical variability of Nicaraguan 178 arc magmas. Because the hemipelagic and carbonate sediment packages both have low  $\delta^{88}$ Sr, 179 and because the arc data unexpectedly span a larger range in  $\delta^{88}$ Sr than the subducting sediments, a robust estimate of sediment recycling efficiencies requires interpretations of Sr isotope data alongside other trace element compositions. To quantify the provenance of Sr erupted in the arc, we first employ a simple Sr mass balance utilizing trace element abundances 183 and Sr/ $86$ Sr data. This approach can determine the general proportion of Sr in the arc lavas 184 originating from all sediments, AOC, and mantle. Next, we incorporate the  $\delta^{88}$ Sr data to distinguish between the inputs of the carbonate vs hemipelagic sediment. These constraints 186 indicate that Nicaraguan arc lavas require both a high- $\delta^{88}$ Sr altered ocean crust component and 187 an unaltered crustal component with low- $\delta^{88}$ Sr. We propose that these combined signals may result from the subduction of seamounts along the Cocos plate, and present an internally consistent, quantitative model that supports this hypothesis. This model can also be used to quantify the flux of carbonate sediment from the slab to the arc and thus provides an independent method to estimate the magmatic carbon recycling efficiency for the Nicaraguan Arc.

## **Quantifying strontium sources from sediment, AOC, and the ambient mantle**



214 melt, while the amount of AOC melt is between 6% and 13% (Fig. 6a). This mass balance shows 215 that only  $\sim$ 10% of the Sr in the arc lavas could have originated from the ambient mantle.

216 Because very little of the Sr in erupted arc lavas is derived from the ambient mantle, the 217 ambient mantle source has a negligible influence on  $\delta^{88}$ Sr. An alternative low- $\delta^{88}$ Sr, low-218 <sup>87</sup>Sr/<sup>86</sup>Sr reservoir is thus required from the slab. This is depicted quantitively by the two mixing 219 triangles on Figure 6b. The first (blue) mixing triangle spans between hemipelagic sediment, 220 carbonate sediment, and a hypothetical AOC melt composition with  $\delta^{88}$ Sr =0.36‰ (consistent 221 with experimental data<sup>50</sup>, and discussed in more detailed in the Online Supplement), which is 222 the minimum AOC value capable of producing a mixing triangle that overlaps the composition 223 of Cerro Negro, the high  $\delta^{88}$ Sr arc end member. For the second (gray) mixing triangle, the pure 224 AOC melt end member is replaced by a mixture of 94% DMM and 6% AOC melt, which is the 225 minimum possible AOC melt addition inferred from figure 6a. Varying the influence of the DMM 226 within this allowable range on figure 6b does not significantly shift the position of the mixing 227 triangle, and neither triangle overlaps the remainder of the arc data. To account for the full 228 range of the arc data, there must instead be an additional slab component with both low 229  $\frac{87}{5}$ r/ $\frac{86}{5}$ r and low  $\delta^{88}$ Sr. Figure 6c shows that a second ocean crust component with a  $\delta^{88}$ Sr 230 equal to average unaltered MORB ( $\delta^{88}$ Sr =0.28± 0.05‰)<sup>49</sup> could plausibly serve as this 231 additional slab component. In this case, the high  $\delta^{88}$ Sr composition of Cerro Negro would 232 reflect a large contribution of AOC melt, while the low- $\delta^{88}$ Sr composition of Masaya would be 233 dominated by a melt of unaltered ocean crust.

234 To summarize, the arc lava Sr budget is dominated by low-<sup>87</sup>Sr/<sup>86</sup>Sr sources that must 235 have variable  $\delta^{88}$ Sr. Mass balance shows that the low- $\delta^{88}$ Sr, low- $^{87}$ Sr/ $^{86}$ Sr source cannot be the

236 DMM. Instead, two distinct ocean crust components are needed to account for the full range of 237 arc lava  $\delta^{88}$ Sr. The Cerro Negro lava end member requires an AOC melt with a  $\delta^{88}$ Sr >0.36‰ 238 whereas the Masaya end member must have an ocean crust component with  $\delta^{88}$ Sr <0.28‰. 239 We propose that this second low-<sup>87</sup>Sr<sup>/86</sup>Sr, low-δ<sup>88</sup>Sr component represents oceanic crust with 240 an unaltered MORB composition.

241

### 242 **Evidence for co-variation of ocean crust and sediment components**

243 A surprising result is that the proportions of high-δ<sup>88</sup>Sr vs low-δ<sup>88</sup>Sr ocean crust appear 244 to co-vary with the proportions of hemipelagic and carbonate sediment. Figure 6c shows that a 245 low-δ<sup>88</sup>Sr, DMM-like, ocean crust component (gray hexagon) produces an ocean crust-sediment 246 mixing triangle that overlaps the compositions of the Telica (gray triangles) and Masaya (red 247 diamond) samples but does not encompass the composition of the Cerro Negro (blue circle). An 248 alternative mixing triangle starting from a high- $\delta^{88}$ Sr AOC is required to account for the Cerro 249 Negro composition, but it does not encompass any other samples. The Cerro Negro 250 composition is reproduced within uncertainty by a mixture of high-δ<sup>88</sup>Sr AOC and a range of 251 different sediment proportions, though other trace element constraints (e.g., Fig. 3) suggest 252 that a dominantly hemipelagic sediment component is implausible for this volcano. In contrast, 253 the composition of the Masaya end member can only be reproduced using the low- $\delta^{88}$ Sr ocean 254 crust component and a sediment that is almost entirely dominated by the hemipelagic 255 component. The Masaya composition thus requires a dominantly hemipelagic sediment and 256 less altered oceanic crust, while the Cerro Negro composition requires high-δ<sup>88</sup>Sr altered 257 oceanic crust and a greater contribution from carbonate sediment.



279 which post-date the "carbonate crash"<sup>29</sup> – could potentially 'cap' the carbonate layer and prevent the transfer of carbonate material from the slab to the mantle wedge.

 The seamounts provide a reasonable explanation for why samples that mix to the low- $\delta^{88}$ Sr ocean crust component also require a smaller contribution from subducting carbonate (e.g., Fig. 6c-d). Absent seamounts, the down-going slab surface consists of typical AOC overlain by a carbonate and hemipelagic sediment (Fig. 7a), in which case melts of the upper slab would be a mixture of typical AOC and carbonate+hemipelagic sediment in proportions comparable to the total subducting sediment masses. This scenario accounts best for the end-member composition of Cerro Negro (Mixing Scenario A, Fig. 6d). If a seamount erupted following the carbonate crash, however, it would cap the carbonate layer and become blanketed in hemipelagic sediment prior to subduction (Fig. 7b). In this case, the slab melts would be a mixture of depleted and less-altered seamount material combined with hemipelagic sediment, consistent with the end-member composition of Masaya (Mixing Scenario B, Fig. 6d). Intermediate scenarios are also possible. Older seamounts erupted prior to the carbonate crash could produce slab melts composed of depleted ocean crust and varying proportions of hemipelagic and carbonate sediment (Fig. 7c). Alternatively, where the subducting plate is capped by thinner outlying seamount lava flows, melting might affect both 296 high- and low-δ<sup>88</sup>Sr ocean crust alongside both sediment units (Fig. 7d). Either of these intermediate scenarios might account for the range of compositions observed among the Telica samples.

#### **Testing the seamount hypothesis with quantitative trace element modeling**

 If the Sr isotope variations of the arc lavas reflect changing sediment proportions due to the presence of seamounts on the subducting plate, then systematic compositional differences in a variety of trace element abundances should also be observed. To evaluate this hypothesis, we first consider the Th/Nd ratio, which is sensitive to hemipelagic sediment input. Mixing model component compositions in Figure 8 are based on the same sources as those in Figure 6. Figures 8a-b show that the mixing scenarios determined from Sr isotope constraints are also successful for Th/Nd: Mixing Scenario A, which includes a bulk sediment mixture comparable to total subducting sediment along with typical AOC, produces a sediment mixing curve that passes through the lower Th/Nd of Cerro Negro. Mixing Scenario B, which is dominated by hemipelagic sediment and melt from unaltered ocean crust, produces a mixing curve that passes through the higher Th/Nd composition of Masaya. The same parameters used in the preferred Masaya and Cerro Negro quantitative models based on Sr isotopes alone (Fig. 6) also reproduce these volcano Th/Nd compositions (Fig. 8a-b). Though it is difficult to distinguish between the two ocean-crust end member

314 components on figure 8a and b, Th/Nd ratios clearly show that the high-δ<sup>88</sup>Sr and low-δ<sup>88</sup>Sr lavas of Cerro Negro and Masaya, respectively, require significantly different sediment inputs consistent with the proportions determined by Sr isotope constraints (Fig. 6d). Masaya lavas 317 require little to no carbonate input and a low-δ<sup>88</sup>Sr, DMM-like, unaltered ocean crust whereas Cerro Negro can be explained by mixing between a sediment melt with a ~70% carbonate 319 component and a high-δ<sup>88</sup>Sr AOC.

 A further test of this conceptual model can be conducted using the full array of commonly measured incompatible trace element abundances. Figure 9 shows that after accounting for differences in fractional crystallization and extents of mantle melting, the end member mixing scenarios developed using only Sr isotope ratios (Fig. 6) and Th/Nd (Fig. 8) also reproduce the full array of trace element abundances for both Masaya and Cerro Negro. All elements that are strongly controlled by the slab exhibit a close match in our model except for Ba (dotted lines and open symbols in Fig. 9). A possible mechanism to reconcile the Ba mismatch is the partial loss of Ba from the sediments into the forearc before reaching sub arc depths, as observed in Costa Rica by Solomon and Kastner (2012). The Ba abundances of both Masaya and Cerro Negro can both be reproduced by the model if 50% to 60% of Ba is lost from the hemipelagic sediment prior to subduction (solid lines and closed symbols in Fig. 9). Model results consistent with measured arc lava trace element abundances support our proposed seamount subduction model based on the Sr stable isotope data. This new geochemical framework provides a first-order explanation for the longstanding observation of variable sediment recycling in Central America that was previously not well understood. Additionally, our model results suggest that Ba in the sediments must be lost to the forearc prior to melting.

#### **Carbon recycling efficiency estimates**

 The calculated proportions of Sr contributed to the arc can now be used to estimate carbon recycling efficiency. Given ~200 m of subducting carbonate sediment with a density of 341 1.75 g/cm<sup>3</sup> and a convergence rate of 71.5 km/Ma<sup>22</sup>, the subducting  $CO_2$  input into the



## **Discussion**

 Strontium stable isotope measurements of Nicaraguan arc basalts not only enable direct estimates of magmatic carbon flux from the arc but also shed new light on the subduction zone systematics that give rise to the geochemical variability in Central American 363 arc lavas. δ<sup>88</sup>Sr and <sup>87</sup>Sr<sup>/86</sup>Sr constraints dictate that (1) in addition to AOC, a second, unaltered,

364 DMM-like crustal component is required to account for the low- $\delta^{88}$ Sr lava from Masaya lavas; and (2) co-variation of the ocean crust components with the hemipelagic and carbonate 366 sediment components can account for the full  $\delta^{88}$ Sr array of the measured Nicaraguan arc 367 lavas. The low-δ<sup>88</sup>Sr ocean crust component likely comes from subducting seamounts atop the 368 Cocos plate that exhibit mantle-like <sup>87</sup>Sr/<sup>86</sup>Sr. With diameters up to 14 km and elevations up to 2 km above the surrounding seafloor, the seamounts erupted after the carbonate crash plausibly cap the underlying carbonate sediments and AOC producing a "seamount effect" that only allows for the transfer of a hemipelagic+depleted and less-altered seamount material to the arc. End-member mixing proportions for sediment and crustal components are consistent 373 for both Sr isotopes and trace elements. Low- $\delta^{88}$ Sr Masaya lavas require mixing of a pure depleted unradiogenic crustal component with a nearly 100% hemipelagic sediment. The high-375 S<sup>88</sup>Sr Cerro Negro end member requires a pure AOC ocean crust component mixing with a 376 sediment component that is ~70% carbonate material. Telica lava  $\delta^{88}$ Sr values fall between Cerro Negro and Masaya end members and likely were produced by intermediate scenarios that reflect variable proportions of all four components.

 Our conceptual model is supported by quantitative forward model results which reproduce the Cerro Negro and Masaya lava end member Sr isotope compositions and most trace element abundances within uncertainty. Using our quantified carbonate flux determined from Sr stable isotope data we estimate that ~26% to ~52% of the carbon from the subducting carbonate sediments is recycled back out of the arc. These estimates are consistent with recent carbon recycling efficiencies from Barry et al. (2019) and Bekaert et al. (2020). The results of this study provide a new quantitative framework to explain the local variations in arc lava

 geochemistry in Nicaragua and highlight the power of stable Sr isotope compositions as a tool to understand subduction components and carbonate fluxes at arcs. Strontium stable isotopic data have enormous potential to serve as a robust proxy for carbon transfer across subduction 389 zones alongside traditional  $CO<sub>2</sub>$  gas and carbon isotope measurements.

#### **Methods**

#### **Sample selection**

393 Chly samples with Eu\*> 0.95 (Eu/Eu\* = Eu<sub>cn</sub>/[Sm<sub>cn</sub> \* Gd<sub>cn</sub>]<sup>1/2</sup>) were selected to minimize the potential influence of plagioclase fractionation. Hemipelagic and carbonate sediments from 124 m, 238 m and 362 m core depths recovered from DSDP site 495 were also analyzed to constrain the Sr isotope compositions of the subducting Cocos plate sediments. Two AOC 397 composite powders from Kelly et al. (2003) previously measured for  $\delta^{88}$ Sr by Klaver et al. (2020) were also analyzed.

### **Strontium double spike TIMS analysis and sample preparation**

 Strontium isotope analyses were conducted via Thermal Ionization Mass Spectroscopy 402 (TIMS) using a Sr double-spike to correct for instrumental mass fractionation. Two  $\sim$ 10 mg aliquots of each sample – one containing the Sr double spike and one unspiked – were digested 404 in a 4:1 solution of 16N Aristar Plus trace metal analysis grade HNO<sub>3</sub> and 48N ultra-pure HF in 405 Savillex beakers at 150° C for 48 hours in clean labs at the University of Massachusetts, 406 Amherst. Samples were evaporated to dryness at 100° C then dissolved twice in 20ul of 16N 407 Aristar Plus trace metal analysis grade  $HNO<sub>3</sub>$  and evaporated to dryness at 100 $^{\circ}$  C. Following the



429 Morkman and Hart (2005),  $\delta^{88}$ Sr (~0.28‰) that matches the average N-MORB  $\delta^{88}$ Sr reported



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## **Acknowledgments**

- We thank Isaac Larson for use of clean lab facilities, Stephen Eyles for providing space and technical
- assistance with quadrupole ICP-MS analyses, and Timothy Mock for this technical assistance with TIMS
- analyses. This work was supported by the National Science Foundation (Grant No. 1939080).
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## **Author Contributions**

- S.J.T. and R.P. conceived of the project and acquired funding. All authors contributed to method
- development. A.J.H. performed column calibration experiments and conducted all sample preparation
- of unknowns and conducted TIMS analysis with R.W.C. A.J.H. and S.J.T primarily contributed to data
- interpretation; all authors participated in data discussion. A.J.H. wrote the manuscript with input from
- all authors.
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## **Ethics declarations**

- Competing interests
- The authors declare no competing interests

**Figures**







665 666 **sediments from DSDP Site 495**. **A** DMORB (black dotted line) <sup>62</sup> normalized incompatible element diagram highlighting the Bulk sediment compositions from ref 26. **B** General stratigraphy of DSDP site 495 with U/La and Ba/Th ratios vs depth (see text for explanation). A data point with an anomalously large Ba/Th value has been removed to preserve scale.



669 670 **Figure 3. U/La vs Ba/Th trace element ratios showing how variations in sediment flux and the relative proportion each Cocos plate sediment influence the overall geochemical variations in Nicaraguan arc lavas (open circles**). The decreasing trend of the arc lavas

- indicate that U/La and Ba/Th are controlled by the sediments. Volcanoes selected for this study include: Telica (gray triangles, Cerro Negro (blue circle), and Masaya (red diamond).
- 671 The Blue and yellow filled squares are bulk hemipelagic and carbonate sediment compositions from ref 26, respectively. Gold star is depleted MORB mantle (DMM) of ref 60. Pink pentagon is altered ocean crust (AOC) melt.



673 674 675 **Figure 4. δ <sup>88</sup>Sr of different geochemical reservoirs.** Fractionation of Sr isotopes during carbonate precipitation (yellow diamond) from seawater (blue diamond) produce an average carbonate  $\delta^{88}$ Sr that is significantly lighter than the reported range of  $\delta^{88}$ Sr for altered and unaltered MORB (grey bar). The original  $\delta^{88}$ Sr of each component transferred to the arc should not undergo discernible fractionation during magma petrogenesis therefor highlighting the Sr stable isotope system's potential to serve as a tracer for subduction components. Average seawater and marine carbonate δ<sup>88</sup>Sr from ref 47 and 48. Range of MORB/AOC  $δ<sup>88</sup>Sr$  from ref 49 and 50.



678 679 680 681 **Figure 5. Three isotope plot showing Sr isotope systematics of measured Nicaraguan arc**  lavas and Cocos plate sediments from this study. Arc lava <sup>87</sup>Sr/<sup>86</sup>Sr does not vary and falls closer to the un radiogenic slab components. Gray triangle is possible range of oceanic crustal component Sr isotope compositions based on measured and experimental δ<sup>88</sup>Sr for altered and unaltered N-MORB . The upper bound of the array represents possible AOC component with a  $\delta^{88}$ Sr ~0.37. This value is from experimentally altered MORB from ref 50. Gold star is ambient mantle Sr isotope composition (DMM). DMM Sr isotope values determined using average unaltered MORB  $\delta^{88}$ Sr of 0.279 ± 0.05‰ from ref 49 and  $87$ Sr/ $86$ Sr of 0.7028 measured in back arc basalts from La Providencia Island that have negligible subduction related trace element signatures<sup>61</sup>.

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685 686 687 688 **Figure 6. Plots of <sup>87</sup>Sr/<sup>86</sup>Sr vs trace element ratios and δ <sup>88</sup>Sr for Nicaraguan arc basalts showing binary mixing models and quantitative forward model results for Masaya and Cerro Negro volcanoes. A** Yb/Sr ratio mass balance to determine relative proportion of Sr contributed from each slab component. Even at very small amounts, the mass balance indicates AOC exerts the greatest control on Nicaraguan arc magma Sr content. **B** δ<sup>88</sup>Sr further highlights how AOC dominates Sr contributions. **C** A second, DMM-like, low δ <sup>88</sup>Sr unaltered ocean crust component can account for the low  $\delta^{88}$ Sr Masaya end member lava. **D** Strontium isotope mixing and quantitative model results. Dashed lines are mixing lines for the Cerro Negro and Masaya mixing models, respectively.

689 **End Member Scenarios** Seamount lem Carbonate <u>Carbonate</u> AOC a) Cerro Negro: Hemi+Carb+AOC b) Masaya: Hemi+Seamount **Intermediate Scenarios** c) Hemi+Carb+Seamount d) Hemi+Carb+Seamount+AOC 690 **Figure 7. Conceptual models showing simplified slab surface of down going Cocos plate for** 691 **several Nicaraguan subduction scenarios. A** and **B** are end member scenarios for Cerro Negro and Masaya lavas, respectively. **A** reflects normal subduction in which a mixture of 692 AOC and hemipelagic+carbonate sediments are added from the slab to the arc. In **B** large seamounts on the Cocos plate erupted after the carbonate crash effectively cap the 693 carbonate sediments and underlying AOC. **C** and **D** represent intermediate scenarios in which various proportions of each component could reach the arc. 694 695 696 697 698





 **Figure 8. Plots of <sup>87</sup>Sr/<sup>86</sup>Sr vs Th/Nd for Nicaraguan arc basalts showing mixing models and quantitative forward model results for Masaya and Cerro Negro volcanoes. A** and **B** show Th/Nd ratio binary mixing and quantitative forward model results for Masaya and Cerro Negro end members respectively. These results are consistent with  $\delta^{88}$ Sr constraints shown

 in **Fig. 6 A-D**. Symbols same as in **Fig. 6.** Solid hashed lines are binary mixing lines between ocean crust and sediment components.





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710 **Figure 9. DMORB normalized<sup>62</sup> incompatible element diagram comparing modeled and measured trace element compositions for Cerro Negro and Masaya volcanoes.** Shaded regions are the range of measured trace element compositions for Masya and Cerro Negro

- 711 volcanoes, respectively. Dotted lines with open symbols are model results that do consider Ba loss. Following the results of ref 46 a 60% loss of Ba from the sediments in the forearc can
- 712 account for the Ba mismatch in the trace element abundance model. Nb and Ta are controlled by residual rutile and not a function of slab recycling. Similarly, Zr and Hf do not match for either model, but these values are dependent on our assumption of zircon
- 713 solubility in the slab melts which is not well constrained. See Turner and Langmuir (2022b) for detailed explanation on how partition coefficients for Zr, Hf, Nb, and Ta are determined.

## 716 **Tables**

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**Table 1.** Strontium isotope compositions and selected trace element ratios of Nicaraguan arc basalts, Cocos plate sediments, altered ocean crust, and reference standard JB-2. δ<sup>88</sup>Sr values are reported relative to SRM-987. Complete trace element compositions of for arc lavas and sediments provided in the Online Supplement.



Trace element data for basalts from ref 56



## **Ion exchange chromatography and column chemistry calibration**

 Spiked and un-spiked aliquots were prepared for each unknown sample. For spiked 724 samples, a mixed <sup>87</sup>Sr-<sup>84</sup>Sr solution was added to the aliquot prior to dissolution in order to capture any mass-dependent fractionation during digestion or column chemistry. Strontium 726 was separated using Eichrom Sr Spec resin<sup>1</sup> in house-made Teflon microcolumns. Resin was 727 preconditioned by passing 3 ml of 8N Aristar Plus trace metal analysis grade HNO<sub>3</sub> through the columns. Next, digested sample solutions were loaded into the columns and washed twice with 729 3.25 ml of 8N Aristar Plus trace metal analysis grade HNO<sub>3</sub> to elute matrix elements (the use of 730 8N HNO<sub>3</sub> as opposed to 3N HNO<sub>3</sub> was required to fully separate Sr from Ba). Following the 731 wash, purified Sr was collected by two elutions of 3 ml of Milli-Q H<sub>2</sub>O (18.2 M $\Omega$ ·cm) and evaporated until dry, then redissolved in 10 ul of 8N Aristar Plus trace metal analysis grade 733 HNO<sub>3</sub>. The resulting solution was then dried down once more before a final dissolution in 10 ul 734 of 8N Aristar Plus trace metal analysis grade HNO<sub>3</sub>.

 This column procedure was calibrated to enable efficient separation of Sr from Ba and ensure complete recovery of Sr from our samples, some of which have very high Ba concentrations. The column calibration was verified using a Perkin Elmer Nexion 350D Quadrupole ICP-MS at UMass Amherst. Figure S1 shows the relative Ba and Sr concentrations of eluate collected from the sample wash, and collection phases of our final column calibration

740 procedure using DSDP 495 sediment powder, which contains 2106 ppm Ba and 1444 ppm Sr.

741 The total Sr yield from the collection eluant is 99.9% of the total Sr in the added solution with a

742 negligible amount of Ba (~0.3% of the total Ba in solution passed through the column).



#### 743

**Figure S1**. Sr and Ba concentrations of eluant collected from each step in our successful ion exchange chromatography column chemistry calibration experiment. Samples were measured via solution ICP-MS. Of the total Ba loaded into the column ~5% was lost during the initial column loading step (not measured) and ~95% was eluted during the wash step leaving 0.3% in the collection eluant.

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#### 745 **TIMS analysis and data reduction**

746 Spiked and un-spiked aliquots were analyzed for each unknown sample. For each

- 747 analysis, 1ug of purified Sr was loaded onto degassed Re filaments along with a TaO<sub>2</sub> activator
- 748 and measured using a Thermo Finnigan Triton TIMS at the Carnegie Institution for Science Earth
- 749 and Planets Laboratory in Washington, D.C. <sup>85</sup>Rb was used to monitor for isobaric interference
- 750 from <sup>87</sup>Rb on <sup>87</sup>Sr. In-run <sup>85</sup>Rb/<sup>86</sup>Sr ratios were <  $1x10^{-5}$  and interference corrections to the
- 751 87Sr/86 Sr ratio were accordingly minimal. Data were acquired in 30 blocks of 20 cycles. The spike

 composition was verified via measurements of variable mixtures of the spike solution and SRM987 using ratios of 0:1,1:2, 1:2:,2:1,1:0. To calculate unknown Sr isotope compositions, an exponential fractionation was assumed and spike:sample mixing proportion were left as unknowns. This simple system of equations was then solved by minimizing least squares misfit using MATLAB's fminsearch solver. Results for unknowns were then normalized to fractionation-corrected SRM987 measurements (results of spiked-unspiked pairs) from the same barrel.

# **<sup>88</sup> Sr measurements of ODP 801c composite samples not representative of typical altered ocean crust.**

762 The high- $\delta^{88}$ Sr, low- $87$ Sr/ $86$ Sr end member required to account for the Cerro Negro 763 composition is consistent with altered ocean crust compositions inferred from<sup>2</sup>, though the 764 range of available  $\delta^{88}$ Sr measurements of actual AOC samples, which are all composite samples  $\,$  from ODP 801 $c^3$ , does not encompass the compositional range of AOC end members required to account for the Central American arc data. The 801c site is probably not a good general proxy for AOC, however, because the upper 100 m consists of off-axis lavas rather than typical ocean crust. For example, Pockalny and Larson (2003) suggest that the presence of thick, massive, and laterally extensive off-axis lava flows could seal the original fractured ocean crust from seawater alteration. Moreover, crustal accretion models show that lava flows extending ~2-4 km off-axis can influence crustal subsidence and cause crust to fracture less extensively, 772 decreasing the extent and depth of hydrothermal alteration<sup>4-6</sup>. The unique geology at site 801c may thus explain the lack of heavy  $\delta^{88}$ Sr compositions observed in most of the 801c composite



## **Tables**

	DMM and AOC melt			Sediment melting partition coefficients, bulk sediment				
	compositions			and modeled sediment melt compositions				
	$1$ DMM	2AOC	<sup>2</sup> AOC Melt	$3$ Bulk	$3$ Bulk	<sup>2</sup> Sed D	<sup>4</sup> Sed Melt	<sup>4</sup> Sed Melt
		Melt (CN)	(MS)	Carb	Hemi	values	(CN)	(MS)
<b>Rb</b>	0.05	9.09	6.13	4.28	45.81	0.70	20.71	56.67
Cs	0.00	0.12	0.08	0.15	2.21	0.10	1.40	4.02
Sr	7.66	571.28	414.61	501.37	369.79	0.25	805.38	644.78
Ba	0.56	96.87	65.28	2145.48	2108.18	0.40	3353.39	3312.37
Ti	0.07	0.00	0.00	0.03	0.52	1.00	0.17	0.52
<b>Nb</b>	0.15	0.73	1.04	0.44	5.13	1.33	3.36	4.20
Ta	0.01	0.04	0.07	0.00	0.00	1.33	0.00	0.00
Zr	5.08	62.00	111.00	20.16	117.04	1.80	32.07	76.24
Hf	0.13	2.14	2.68	0.00	0.00	1.20	0.00	0.00
La	0.19	3.71	4.78	8.78	23.34	0.83	14.77	26.22
Ce	0.55	10.57	13.48	2.40	32.64	0.90	12.27	34.91
Nd	0.58	8.46	10.58	6.79	22.90	1.60	8.30	16.36
Sm	0.24	2.01	2.42	1.45	5.23	2.20	1.43	2.89
Eu	0.10	0.47	0.56	0.82	2.45	2.80	0.59	1.10
Gd	0.36	0.96	1.04	1.65	5.21	4.00	0.89	1.71
Dy	0.51	0.35	0.37	1.99	5.98	9.00	0.49	0.92
Er	0.35	0.10	0.10	1.40	3.84	16	0.19	0.34
Yb	0.37	0.06	0.06	1.18	3.40	50	0.05	0.10
Lu	0.06	0.01	0.01	0.19	0.53	75	0.01	0.01
Υ	3.33	0.00	0.00	16.22	39.55	0.00	42.22	71.91
Sc	16.50	0.00	0.00	2.63	15.05	0.00	11.56	27.37
Pb	0.02	1.97	1.54	3.70	11.12	0.50	8.59	16.13
<b>Th</b>	0.01	0.37	0.44	0.16	3.35	0.60	1.50	4.49
U	0.00	0.33	0.33	0.15	3.54	0.40	1.83	5.56
$\delta^{88}$ Sr	0.279	0.36	0.279	0.22	0.15		0.20	0.15
<sup>87</sup> Sr/ <sup>86</sup> Sr	0.7028	0.7032	0.7026	0.70858	0.70763		0.70835	0.70763

**Table S1.** Quantitative forward model end member starting compositions and bulk partition coefficients.

<sup>1</sup> Values from Workman and Hart (2005)

<sup>2</sup> AOC melt and sediment partition coefficients are determined in the model based on temperature input parameters following the methods described by Turner and Langmuir (2022b). Cerro Negro (CN) and Masaya (MS) AOC melt compositions correspond to "moderate" and "hot" values from Turner and Langmuir (2022b), respectively. Sediment partition coefficients correspond to "hot" values from Turner and Langmuir (2022b)

<sup>3</sup> Values from Patino et al. (2000). Bulk hemipelagic sediment values are average composition of the bottom three hemipelagic layers recovered from DSDP 495 (see text for details).

<sup>4</sup>Sed melts calculated using aggregate fractional melting equation, Sed Melt= (D<sub>sed</sub>/F<sub>sed</sub>)\*(1-F<sub>sed</sub>)(1/D), with a melt fraction of F  $= 0.55$ 

forward models.							
Sample	Cerro Negro	Masaya					
Input parameters							
P	1.9	1.9					
F	0.22	0.15					
%Carb	0.7	0					
%Sed	0.0156	0.027					
%AOC	0.09	0.075					
Sed T $(^{\circ}C)$	900	900					
$AOCT(^{\circ}C)$	850	900					
<b>Model Results</b>							
<b>Rb</b>	5.99	16.94					
Cs	0.17	0.96					
<b>Sr</b>	352.96	444.70					
Ba	310.65	789.35					
Nb	1.26	2.68					
Ta	0.06	0.11					
Zr	52.80	119.17					
Hf	1.52	2.49					
La	3.70	10.20					
Ce	8.21	20.06					
Nd	7.03	14.11					
Sm	2.06	3.71					
Eu	0.69	1.26					
Gd	2.12	3.59					
<b>Dy</b>	2.45	3.99					
Er	1.52	2.36					
Yb	1.52	2.32					
Lu	0.23	0.33					
Y	18.37	39.89					
Pb	1.65	4.65					
<b>Th</b>	0.32	1.33					
U	0.31	1.47					
$\delta^{88}$ Sr	0.324	0.237					
87Sr/86Sr	0.7041	0.7042					

**Table S2.** Input parameters and results for Cerro Negro and Masaya quantitative

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