Supplemental material

1. Detailed methods

1.1 Sediment analyses

Sediment samples were obtained from archived DSDP, ODP, and piston cores from the Lamont-Doherty Earth Observatory (LDEO) core archive. Samples were dried at 50 °C, and sediment aliquots for Total Organic C (TOC) analysis were acidified using 1 N HCl for at least 12 hours or until effervescence ceased. Following acidification, samples were centrifuged and washed 3x with DI water and re-dried before homogenization with an agate mortar and pestle. Approximately 100 mg was combusted with Cu/CuOx in an evacuated quartz tube at 910 °C overnight. Resulting CO2 was purified and collected on a gas line. Sample aliquots for carbonate analysis were acidified with 100% phosphoric acid at 70 °C in a Finnigan Gas Bench CO2 handling system. For all samples, CO2 was quantified using manometry, and all isotope measurements were conducted on a Finnigan MAT 252 mass spectrometer operated in dual inlet mode. All δ13C values are reported relative to VPDB C isotope standard. For measurements of C isotopic composition of carbonate within Altered Oceanic Crust (AOC), we used a microdrill to obtain samples of the carbonate veins and breccia matrix. These samples were then analyzed in an identical fashion to sedimentary C.

1.2 Sediment and unit thicknesses
Archived 3.5 kHz single channel seismic lines from the Lamont-Doherty Earth Observatory (LDEO) collection were used to determine overall sediment thickness and the thicknesses of geochemically distinct units outboard of the Sunda margin. Archived profiles from the following cruise tracks were used:

- RC 1107
- RC 1402
- RC 1403
- V 1909
- V 2009
- V 2410
- V 2819
- V 2901
- V 3503
- V 3616

Seafloor, basement, and unit boundary picks, made using the GeoMap App program for these profiles, were augmented with additional constraints from more recent published multichannel lines (Kopp and Kukowski, 2003; Lüschen et al., 2011; Kopp et al., 2006; Planert et al., 2010; Kopp, 2013; Kopp et al., 2009; McNeill et al., 2016; Dean et al., 2010). Geochemical unit boundaries (Nicobar Fan, terrigenous trenchfill, pelagic material, and calcareous turbidites) were defined by picks following the seismic interpretations in the Initial Report volumes of the appropriate DSDP/ODP sites and subsequent seismic studies. The terrigenous trenchfill unit is defined by reflectors that converge moving away from the trench, and the top of the Nicobar Fan was inferred to coincide with the base of this unit as reported elsewhere (e.g. Dean et al., 2010).
The base of the Nicobar Fan lacks clear seismic reflectors, and as such, we assumed a uniform thickness of 200 m for the underlying pelagic unit (which is intermediate between results from DSDP 211 and IODP 1480) rather than using dubious seismic picks to constrain the thickness of the Nicobar Fan unit (Expedition 22 Scientific Party, 1974; McNeill et al., 2017). The lower extent of the unit of calcareous turbidites from the Exmouth Plateau/Australian margin is defined by a distinct set of reflectors (Ludden and Gradstein, 1990), and since this unit is uppermost at Sites 261 and 765 where it is sampled, we infer that overlying sediment is likely to be quantitatively minimal throughout the margin. Therefore, constraining this unit’s thickness – as well as that of the underlying pelagic sediments – was straightforward. The lack of a clear seismic boundary between the calcareous turbidite unit and the trenchfill wedge in seismic profiles along the southeastern portion of the margin led us to assign a thickness for the trenchfill unit by subtracting the thickness of the other units outboard of the trench from the overall sediment thickness at the trench. This approach, while necessary to constrain overall sediment flux to the trench, is unlikely to substantially affect our estimates of subducting C because the entirety of the trench wedge appears to be off-scrapped through much of the margin. While the Roo Rise is inducing localized subduction erosion along eastern Java, sediment cover on the rise is minimal, and sediments trenchward of it are difficult to interpret from seismic profiles. Indeed, East Java stands out as a region of low sediment cover (Figures DR2 and DR3), so while our estimates consider the subduction of the thin sediment veneer currently present, significantly more sediment and sedimentary C may have subducted in the recent past when the Roo Rise first impinged upon the margin.

When seismic velocity profiles reported two-way travel time rather than sediment thickness and sediment velocity models were not available, sediment velocities of 2 and 2.5 km/s...
were used to construct upper and lower bounds on sediment thickness respectively. While these
velocities are greater than those determined for surficial sediments (Dean et al., 2010), they
encapsulate the range of velocities expected for the deeper sediments that escape off-scraping at
the deformation front along much of the margin. Therefore, these seismic velocity bounds are
reasonable for estimating subducting C flux and may slightly overestimate the amount of
sediment – and hence C – present at the trench. A total of ~8000 picks were made, and
additional unit and sediment thickness constraints from DSDP Sites 211, 213, 260, 261, ODP
Site 765, and IODP Site 1480 were also incorporated. Using these data, surfaces were
constructed to model unit and overall sediment thicknesses and extrapolate these from core sites
to the trench. With these models and results from sediment analyses, we estimated the amount
and types of C within the sediment column at the trench.

Plate motion models of the Indoaustralian plate relative to the stable Sunda core from
McNeill et al. (2014) were used to convert C content into a C flux to the trench, calculated at
points along the trench that correspond to the MORVEL plate boundary between the
Indoaustralian and Sunda plates (Argus et al., 2011). Single and multichannel seismic profiles
as well as velocity models across the trench (Lüschen et al., 2011; Kopp et al., 2009, 2006; Kopp
and Kukowski, 2003; Kopp et al., 2001; Planert et al., 2010) were used to estimate the fraction of
the overall sediment and C flux bypassing the deformation front and subducting past ~20 km, the
maximum seismically-resolved depth. These estimates were combined with those from McNeill
et al. (2014) to generate upper and lower bounds on the sediment subducting past the
deformation front. This range in possible subduction channel thickness leads to the range in
estimates for subducted C amount.

2. Comparing shipboard and shore-based organic C analyses
The tendency for disagreement between shipboard estimates of Total Organic C (TOC) made during coring expeditions and shorebased measurements has been previously documented (Olivarez Lyle and Lyle, 2006). Shipboard TOC is typically not measured directly but calculated as the difference between total C (measured by combustion elemental analysis) and carbonate C (measured by coulometry). However, this method can lead to substantial uncertainty. Several hundred replicate shipboard analyses conducted on a total of 5 samples from IODP Site 1480 give one-sigma uncertainties of 35 to 180% (McNeill et al., 2017). Furthermore, comparing the pool of shipboard data from DSDP Sites 211, 260, 261, 262, and ODP Site 765 (Ludden and Gradstein, 1990; Plank and Ludden, 1992; Bode, 1974; Pimm, 1974) with our shore-based TOC results shows that we are unable to reproduce the highest shipboard TOC values and that the distribution of values is substantially different between ship- and shore-based analyses (Figure DR1). While these analyses were not conducted on precisely the same samples, the large number of data compiled suggests a true difference between the two distributions, and indeed a one-tailed non-parametric unpaired t-test reveals that the mean of shipboard data is higher than that of shorebased data at a statistically significant level ($p < 10^{-10}$). This suggests that using shipboard data is likely to overestimate of the abundance of organic C and to overemphasize its importance in C-cycling. Future studies of organic C cycling at convergent margins should therefore rely on shorebased reanalysis of shipboard samples.

3. Caveats in determining provenance of arc CO$_2$

The three endmember mixing model of Sano and Marty (1995) has been used previously to establish the provenance of volcanic and hydrothermal CO$_2$ released along Indonesia (Halldórsson et al., 2013; Figure DR3). In this model, the $\delta^{13}$C and CO$_2$/$^3$He composition of
volcano or hydrothermal CO₂ are used to resolve contributions from subducting carbonate and organic C as well as C from the mantle wedge. While the δ¹³C of organic C can be readily measured, the meaning of a CO₂/³He ratio for organic C is less clear. A value of ~10¹⁴ is typically used for this ratio, as well as for the CO₂/³He ratio of sedimentary carbonate (termed “limestone” in previous work; Marty et al., 1989; Hilton et al., 2002; Halldórsson et al., 2013). Because typical volcanic gases have a CO₂/³He ratio of ~10¹⁰, CO₂ provenance results from this mixing model are relatively insensitive to changes in these endmember compositions. However, the CO₂/³He composition of the mantle wedge endmember is also required for the mixing model, and small changes in this value can have enormous effects on inferred CO₂ provenance.

A value of ~2.7 x 10⁹ is usually used for the CO₂/³He ratio of the mantle wedge (e.g. Hilton et al., 2002), but this value reflects the average of measurements of volatile concentrations in mid-ocean ridge glasses, which show significant variation (e.g. Graham, 2002), an may not represent the composition of the extensively flux-melted mantle wedge. When the CO₂/³He ratio in volcanic gas samples is relatively low, the precise CO₂/³He ratio used for the mantle wedge endmember becomes very important. Using data from Varekamp et al. (1992) for Sirung volcano, even a 25% deviation from 2.7 x 10⁹ leads to a ~10-fold change in the ratio of CO₂ from carbonate to CO₂ from organic C inferred from the mixing model. Therefore, results from this mixing model should be used with caution, which is why we consider the simple balance between the δ¹³C of the subducting sedimentary C and the δ¹³C of volcanic CO₂ to infer that an additional ¹³C rich reservoir external to sedimentary C is likely contributing to the arc-released CO₂. We note that one of the recent studies invoking significant CO₂ contributions from carbonates in the overlying crust relied almost entirely on this mixing model to argue for the global importance of this CO₂ source (Mason et al., 2017). It is worth noting that both the Sunda
and Central American subduction margins, which this model infers to have substantial contributions from crustal carbonate, are regions in which a significant amount of carbonate-rich sediments are subducting.

Additionally, this mixing model does not permit differentiation of carbonate CO₂ source (i.e. sedimentary carbonate, carbonate in altered oceanic crust, or carbonate in the overriding plate) due to their indistinguishable δ¹³C values. Likewise, our method of directly comparing the δ¹³C of subducting sediments with that of volcanic CO₂ reveals only that an additional carbonate source is needed to raise the δ¹³C of subducting C (~ -13‰) to that of arc CO₂ (~ -4 ‰). Thick limestone sequences in southern Sumatra and Java have been explored as potential hydrocarbon reservoirs, and because of this, they may contribute a substantial quantity of CO₂ to volcanic emissions (e.g. Deegan et al., 2010). However, we find little evidence for extensive carbonate deposits in northern Sumatra, particularly in the regions of greatest volcanic activity and hence densest sampling (Wilson, 2002). This section of the margin also shows the greatest potential for subduction of organic-rich Nicobar Fan sediments, and even if the décollement is below the fan deposits as has been hypothesized (Hüpers et al., 2017), the amount of organic and inorganic C subducting in sediments will be approximately equal, and an additional C source will be required to balance the δ¹³C of the sedimentary influx with that of volcanic CO₂. Carbonates in the overlying crust could be a significant factor in closing the C cycling budget, but we also consider carbonate in altered oceanic crust to be a likely contributor in the northern section of the margin, and therefore a possible C source both throughout this margin and others.

Other insight, potentially significant, regarding the mixing of C from multiple sources comes from study of deeply subducted metasedimentary rocks containing varying proportions of C in carbonate (oxidized) and C in carbonaceous matter (reduced). Cook-Kollars et al. (2014)
and Kraft et al. (2017; manuscript in preparation) demonstrated extensive C isotope exchange between C in the two reservoirs as they attempted equilibration particularly at the higher temperatures represented in the suite. This exchange is most significant in pelite-carbonate mixed rocks inferred by Cook-Kollars et al. (2014) to have experienced the most C loss by decarbonation. Both the oxidized and the reduced C reservoirs were shifted in $\delta^{13}C$ from their starting/oceanic compositions toward the “mantle value” of -6‰. Depending on the relative abundances of carbonaceous matter and carbonate in the samples, the reduced C is shifted from $\delta^{13}C$ values near -22‰ for low-grade rocks in which there is little re-equilibration with carbonate near 0‰ to values as high as -8‰ at the higher grades (also see Kraft et al., 2017). Carbonate C is shifted from $\delta^{13}C$ values near +1‰ for low-grade rocks in which there is little re-equilibration with carbonate near 0‰ to values as low as -6‰ at the higher grades. Shift in the C isotope compositions of the two subducting C reservoirs would obviously have implications for the application of the three-component mixing model of Sano and Marty (1995). Yet unknown are the relative degrees of C release from each of these reservoirs during the devolatilization and partial melting beneath arcs at the subduction interface and in subducting slabs.
References


Figure DR3
Figure DR1. Histograms showing distributions of shipboard (a) and shore-based (b) Total Organic Carbon (TOC) measurements generated for this study from DSDP 211, 213, 260, 261, 262 and ODP 765. Normalized counts represent the fraction of samples that fall within a given range of TOC values. Shipboard values are calculated as the difference between total C, measured by combustion elemental analysis, and carbonate C, measured by coulometry whereas shore-based values represent direct measurements of TOC following acidification to remove carbonate phases. While we were not able to analyze splits of precisely the same samples used for shipboard analyses, the pronounced difference in the distributions and number of data points strongly suggests that shipboard analyses overestimate the true TOC. Indeed, we were unable to reproduce TOC values over 0.86 wt% C with shore-based analyses.

Figure DR2. Model of overall sediment thickness model based on seismic picks (small red points) of seafloor and acoustic basement depth. Points outside the interpolated region represent regions where acoustic basement was not reliably imaged and picks could only be made to constrain unit thicknesses. Black points indicate the DSDP/ODP/IODP sites that are labeled in Fig. 1. The color scale saturates at 3 km sediment thickness. Material from the Nicobar Fan (to the northwest) and calcareous turbidites from the Australian margin (to the southeast) are separated by the Christmas Island Seamount Province.
Figure DR3. The three-endmember mixing model of Sano and Marty (1995) can be used to infer the provenance of volcanic CO₂ from measured δ¹³C and CO₂/³He ratios in volcanic gases (Halldórsson et al., 2013; Varekamp et al., 1992). Solving a set of linear equations for the fraction of CO₂ from sedimentary carbonate (termed “limestone” in the original reference and represented the L endmember), organic C (termed “sedimentary” and denoted by S), and mantle wedge (M) sources. Open, unshaded circles represent global averages for the S and L endmembers that were used previously, and yellow circles represent values measured for this study. The CO₂/³He ratio of the mantle wedge is inferred from measurements of Mid-Ocean Ridge Basalt (MORB), but whether these values accurately represent the mantle wedge composition is unclear. For some volcanoes, the results of this mixing model are exceedingly sensitive to the exact CO₂/³He used.

Table DR1. Concentrations and isotopic compositions of carbonate and organic C

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