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Preservation of terrestrial organic carbon in marine sediments off shore Taiwan: mountain building and atmospheric carbon dioxide sequestration

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Abstract

Geological sequestration of atmospheric carbon dioxide (CO_2) can be achieved by the erosion of organic carbon (OC) from the terrestrial biosphere and its burial in longlived marine sediments. Rivers on mountain islands of Oceania in the western Pacific have very high rates of OC export to the ocean, yet its preservation offshore remains 5 poorly constrained. Here we use the OC content (C_{org}, %), radiocarbon ($\Delta^{14}C_{org}$) and stable isotope ($\delta^{13}C_{org}$) composition of sediments offshore Taiwan to assess the fate of terrestrial OC. We account for rock-derived fossil OC to assess the preservation of OC eroded from the terrestrial biosphere (non-fossil OC) during flood discharges (hyperpycnal river plumes) and when river inputs are dispersed more widely (hypopy-10 cnal). The C_{org} , $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ of marine sediment traps and cores indicate that during flood discharges, terrestrial OC is transferred efficiently to the deep ocean and accumulates offshore with little evidence for terrestrial OC loss. In marine sediments fed by dispersive river inputs, the $C_{org},\,\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ are consistent with mixing of marine OC and terrestrial OC and suggest that efficient preservation of terrestrial 15 OC (>70%) is also associated with hypopycnal delivery. Re-burial of fossil OC is pervasive. Our findings from Taiwan suggest that erosion and marine burial of terrestrial non-fossil OC may sequester > 8 TgC yr⁻¹ across Oceania, a significant geological CO_2 sink which requires better constraint. We postulate that mountain islands of Oceania provide strong link between tectonic uplift and the carbon cycle, one moderated by the 20 climatic variability that controls terrestrial OC delivery to the ocean.

1 Introduction

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Photosynthesis sequesters CO_2 within living matter as OC. If a fraction of this productivity escapes respiratory consumption and oxidation, it represents a carbon-sink that will reduce greenhouse gas concentrations and influence Earth's radiation energy balance (Sundquist, 1993; Stallard, 1998; Berner, 2006). On geological timescales, the



burial of OC in marine sediments is the second largest sink of atmospheric CO_2 after carbonate deposition formed from the products of continental silicate weathering (Gaillardet et al., 1999; Hayes et al., 1999). The erosion of terrestrial OC and its delivery by rivers to the ocean along with clastic sediments is thought to contribute approximately

- ⁵ half of this oceanic OC burial flux (Schlunz and Schneider, 2000; Burdige, 2005; Blair and Aller, 2012) in part because the efficiency of OC burial is closely related to the accumulation rate of the accompanying sediment (Canfield, 1994; Burdige, 2005; Galy et al., 2007a). Therefore CO₂-sequestration by OC burial may be sensitive to changes in tectonic and climatic boundary conditions which regulate the erosion and transfer of clastic sediment and terrestrial OC by rivers (Dadson et al., 2003; Hilton et al., 2008a,
- 2012; Milliman and Farnsworth, 2011), giving rise to feedbacks in the global carbon cycle (cf. West et al., 2005) which are not represented in models of the carbon cycle (Berner, 2006).
- The Himalayan orogeny is thought to exert tectonic forcing on the carbon cycle (Gaillardet and Galy, 2008), sequestering 3.7 ± 0.4 TgC yr⁻¹ through erosion of OC from terrestrial biomass (OC_{non-fossil}) and its preservation and burial in the distant Bengal Fan (France-Lanord and Derry, 1997; Galy et al., 2007a). On mountain islands of Oceania (Taiwan, Philippines, Indonesia, Papua New Guinea and New Zealand), where landocean linkages are strong, small mountain rivers drain a larger combined source area
- ²⁰ than the Himalayas (~ 2.7×10^{6} km² cf. 1.6×10^{6} km²). These rivers transport ~ 7 Pg yr⁻¹ of clastic sediment (Milliman and Farnsworth, 2011) and an estimated 20–40% of the global particulate OC flux to the oceans (Lyons et al., 2002). There, convergent plate margins have steep, high standing topography where erosion of OC_{non-fossil} occurs at very high rates (up to ~ 70 MgC km⁻² yr⁻¹) and rivers can deliver particulate materi-
- ²⁵ als rapidly to the ocean across short floodplains (Dadson et al., 2003, 2005; Hilton et al., 2008a, 2012; Bass et al., 2011). These conditions should be conducive to high rates of OC burial but, unlike the Himalayan system, our understanding of the fate of OC_{non-fossil} offshore and the resultant CO₂ sequestration around these ocean islands remains incomplete (Eglinton, 2008).



Firstly, this reflects the challenge of accounting for fossil OC from sedimentary rock (OC_{fossil}) in terrestrial and marine sediments (Blair et al., 2003) which can contribute significantly to the particulate load of mountain rivers (Kao and Liu, 1996; Komada et al., 2004; Leithold et al., 2006; Hilton et al., 2010; Clark et al., 2013). OC_{fossil} transfer and re-burial lengthens the residence time of OC in the lithosphere (Galy et al., 2008; Hilton et al., 2011) however it does not represent recent atmospheric CO.

- 2008; Hilton et al., 2011), however it does not represent recent atmospheric CO_2 and so must be quantified separately. Secondly, it reflects the difficultly of assessing the range of delivery mechanisms to the ocean by mountain rivers. During floods, high suspended sediment concentrations (> 40 g L⁻¹) can cause the density of the river outflow
- to surpass that of ambient seawater (hyperpycnal) and result in density currents transporting sediment down submarine canyons into the deep ocean (Mulder and Syvitski, 1995). Previous work has postulated that hyperpycnal discharges are essential for the efficient transfer of terrestrial OC into marine deposits offshore mountain islands (Kao et al., 2006; Hilton et al., 2008). However, large amounts of terrestrial OC and clastic sediment are also delivered to the surface ocean by rivers in hypopycnal plumes
- tic sediment are also delivered to the surface ocean by rivers in hypopychal plumes with density lower than seawater. Such plumes disperse fluvial materials over a larger region (Mulder and Syvitski, 1995; Dadson et al., 2005; Kao and Milliman, 2008) and may have a lower terrestrial OC burial efficiency.

In order to shed some light on the fate of OC_{non-fossil} eroded from high standing ocean islands, we consider the mountain island of Taiwan (Fig. 1). In Taiwan, where rapid convergence of the Philippine Sea Plate and the Eurasian continental margin combines with a climate characterised by frequent tropical cyclones to drive high rates of fluvial sediment export to the ocean (Dadson et al., 2003; Kao and Milliman, 2008). Findings from Taiwan are of wider relevance because the steep mountain rivers drain-

ing this island are common throughout Oceania (Milliman and Farnsworth, 2011). They have short transit times (Hilton et al., 2008) and deliver most of their sediment loads (~60–70%) under hypopycnal conditions (Dadson et al., 2005; Kao and Milliman, 2008). However, Taiwan's rivers can also produce hyperpycnal plumes (Mulder and Syviskti, 1995; Dadson et al., 2005) allowing us to study terrestrial OC transfer and



preservation associated with both modes of fluvial delivery. To assess the offshore transfer of terrestrial OC and its preservation in marine sediments, we have collected seafloor sediments, material from sediment traps and cores from the Gaoping Canyon off Southwest Taiwan, which is prone to hyperpycnal inputs. In addition, we examine marine sediments from the Okinawa Trough, Taiwan Strait and Gaoping Shelf where hyperpycnal inputs are thought to be more important (Fig. 1). Employing an established

- hypopycnal inputs are thought to be more important (Fig. 1). Employing an established approach, we have measured the OC content, stable OC isotopes and radiocarbon content of OC to determine sources of OC in the sediments and to assess the preservation of fossil and non-fossil terrestrial OC offshore (e.g. Blair et al., 2004; Komada 10 et al., 2004; Leithold et al., 2006; Galy et al., 2007a, 2008; Hilton et al., 2008, 2010;
- Blair et al., 2004; Leithold et al., 2006; Galy et al., 2007a, 2008; Hilton et al., 2008, 2010; Blair et al., 2010; Clark et al., 2013). The findings from Taiwan are placed in a regional context, and their implications for the global carbon cycle are discussed.

2 Materials and methods

2.1 River suspended sediment samples

- ¹⁵ To characterise the composition of OC input to the ocean by Taiwanese rivers, suspended sediment samples were collected for this study from the primary rivers (Fig. 1) under common flow conditions as well as during tropical cyclone-induced floods, covering water discharges ranging from <1 to ~40 times the long-term average (Table S1) and complemented with published data (Kao and Liu, 1996; Hilton et al., 2008). For each sample, a known volume (between 250 mL and 1 L) of river water was collected from the surface of the main river channel in a wide-mouthed plastic bottle thoroughly rinsed with river water. The sample was then filtered through 0.7 μm GF/F membrane filters and the content were dried at 60 °C, weighed to determine total suspended sediment concentration (SSC, gL⁻¹) and stored in sealed glass dishes. Water discharge
- $_{25}$ (Q_{w} , m³ s⁻¹) was measured by the Water Resources Agency, Taiwan, and reported here where available (Table S1).



2.2 Marine sediment samples

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To assess the fate of terrestrial OC delivered to the ocean by hyperpychal discharges, a sediment trap mooring was deployed at 650 m water depth in the submarine Gaoping Canyon, fed by the Gaoping River (Fig. 1). Full details of the collection methods
⁵ can be found elsewhere (Huh et al., 2009; Liu et al., 2012, 2013). Briefly, the sediment trap mooring and an upward-facing long-range acoustic Doppler profiler were moored in the canyon during the 2008 typhoon season. The mooring was configured with a non-sequential sediment trap consisting of a collecting funnel and core-liner, and an intervalometer timer capable of inserting Teflon discs into the collected sediment as
¹⁰ embedded time-markers (Xu et al., 2010). During deployment, Typhoon Kalmaegi impacted Taiwan (17 July 2008) and the Telfon discs inserted before its landfall and after the flood waters had ceased allowed us to constrain sediment associated with the typhoon event. To assess longer-term preservation, a box core was collected from the thalweg of the Gaoping Canyon by R/V *Ocean Researcher-1* in September and Oc-

tober 2009 (Fig. 1), K1 at a water depth 160 m and sub-sampled at different depths (Table S3). The sediments are thought to represent deposits associated with hyperpy-cnal river discharge during Typhoon Morakot in August 2009 (Sparkes, 2012; Liu et al., 2013), whose exceptionally heavy rainfall in Taiwan triggered a very large number of landslides (West et al., 2011) and high rates of sediment delivery offshore. Together,
these marine sediments allow us to assess the transfer and deposition of terrestrial materials to the deep ocean by river hyperpycnal flows.

To assess the fate of terrestrial OC delivered by more dispersive events, hypopycnal discharges, sediments collected from marine trap moorings at 760 m and 940 m in the Southern Okinawa Trough (Fig. 1) were used where the hyperpycnal river discharges are less common (Dadson et al., 2005; Hsu et al., 2006; Kao and Milliman, 2008).

In addition, seafloor sediment samples collected between 1994 and 2009 with a box corer on R/V *Ocean Researcher-1* and *-2* from the Gaoping Shelf, Southern Okinawa Trough and Taiwan Strait were selected (Table S4). Samples were collected from the



top 2 cm of these cores using a stainless steel spatula and freeze-dried. We have also examined Holocene sediments in the long piston core MD012403 collected by R/V *Marion Dufresne* in 2001 from a water depth of 1420 m (Kao et al., 2008). The depositional age of these sediments was determined by analysis of the radiocarbon $_{5}$ content of planktonic foraminfera; so the $\Delta^{14}C_{org}$ at time of deposition can be estimated (Kao et al., 2008).

2.3 Geochemical methods

All marine samples were rinsed with deionised water (>18M Ω) to remove salts. All dried sediment samples were homogenized in an agate mortar. Prior to measurement of the OC concentration (Corg, %) and analysis of the stable isotopes of OC 10 $(\delta^{13}C_{org}, \infty)$, samples were treated with 1 N HCl for 16 h to remove carbonate (Galy et al., 2007b), the residue was centrifuged and freeze-dried (Kao et al., 2008). $\delta^{13}C_{ord}$ analysis was carried out using Carlo-Erba 2100 elemental analyzer connected to a Thermo Finnigan Deltaplus Advantage isotope ratio mass spectrometer and reported in δ notation with respect to the PDB standard and renormalized based on working standards (USGS 40 and acetanilide), with reproducibility better than 0.2 ‰. Radiocarbon (¹⁴C) was measured on total organic carbon by accelerator mass spectrometry after carbonate removal and graphitization at Woods Hole Oceanographic Institute, USA, Institute of Geological and Nuclear Sciences, New Zealand and Keck-Carbon Cycle AMS Facility at University of California at Irvine, USA. ¹⁴C values are given after correction for ¹³C fractionation (normalization to a δ^{13} C value of –25%), and expressed as percent modern carbon (pMC) comparative to 95% of the ¹⁴C activity of the NBS oxalic acid and Δ^{14} C based on established protocols (Stuiver and Polach, 1977), with precision typically better than 10%. Samples from the Liwu River in 2004 were analyzed by similar methods described elsewhere (Hilton et al., 2008). Inorganic carbon 25 removal by HCI leaching was preferred over HCI vapour to ensure complete removal of dolomite (Galy et al., 2007b) which may be present in Taiwanese bedrock and river



sediments (Hilton et al., 2010). The deviation between these two methods for terrestrial materials, $\delta^{13}C_{org} \sim \pm 0.2\%$ and $\Delta^{14}C_{org} \sim \pm 10\%$ (Komada et al., 2008), was similar to the precision of the analyses.

3 Results

5 3.1 Composition of terrestrial OC exported to the ocean

Particulate OC in suspended sediments from the major rivers in Taiwan (Fig. 1) had an average $C_{org} = 0.4 \pm 0.2 \%$ (\pm S.D. Table S2) which is at the lower end of values measured in rivers worldwide (Meybeck, 1982; Stallard, 1998) but is consistent with measurements from mountain rivers elsewhere (Komada et al., 2004; Leithold et al., 2006; Clark et al., 2013; Smith et al., 2013) and previous measurements on Taiwanese rivers (Kao and Liu, 1996, 2000; Hilton et al., 2008, 2010). The particulate OC was radiocarbon depleted and ¹³C-depleted, with a mean $\Delta^{14}C_{org} = -660 \pm 250 \%$ and

- $\delta^{13}C_{org} = -24.9 \pm 0.9 \%$ for the Taiwanese rivers studied here (Table S2). The range in isotopic composition of terrestrial OC define a triangular domain between $\Delta^{14}C_{org}$ and
- 15 $\delta^{13}C_{org}$ (Fig. 2a) and the measured values are consistent with previous measurements on suspended sediments from Taiwan (Kao and Liu, 2000; Hilton et al., 2008, 2010).

3.2 Composition of marine OC

The sediments collected from the trap in the Gaoping Canyon accumulated during Typhoon Kalmaegi (17 July 2008) as constrained by the timing discs deployed by the sediment trap. At that time, SSC in the Gaoping River reached > 20 g L⁻¹ the day after the flood peak. Based on past records of Q_w and SSC (Dadson et al., 2005; Kao and Milliman, 2008) it is highly likely that the Gaoping River surpassed SSC = 40 g L⁻¹ necessary for hyperpycnal discharge in this region during Typhoon Kalmaegi. This is consistent with the very high throughput of sediment in the canyon during the event (Liu



et al., 2012). Particulate OC samples from the trap have an average $C_{org} = 0.5 \pm 0.3 \%$ (n = 12), which is within a standard deviation of the mean of western river samples ($C_{org} = 0.4 \pm 0.2 \%$, n = 20) and similar to previous measurements in the canyon (Kao et al., 2006). Along with the core samples collected from the canyon following Typhoon Morakot, the marine sediments from the Gaoping Canyon have a mean $C_{org} = 0.6 \pm 0.4 \%$ (n = 15) which is only slightly higher than the terrestrial OC (Fig. 3b). Both sets of Gaoping Canyon samples were radiocarbon-depleted (Table S3) and their $\delta^{13}C_{org}$ and $\Delta^{14}C_{org}$ values fall into the triangular domain defined by terrestrial OC carried by Taiwanese rivers (Fig. 3a).

Away from direct hyperpycnal river inputs, marine particulates collected from sed-10 iment traps in the Okinawa Trough (Fig. 1) all had higher $\delta^{13}C_{org}$, $\Delta^{14}C_{org}$ and C_{org} values than terrestrial OC and Gaoping Canyon samples (Fig. 4, Table S4). When plotted with seafloor sediments collected from the Okinawa Trough, Taiwan Strait and the Gaoping Shelf (Fig. 1), the samples reveal a significant (P < 0.0001, n = 26) positive, linear correlation between $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ (Fig. 4a). Samples collected from below 15 the sediment water interface on the Gaoping Shelf (Table S4) also plot on this trend, as do Holocene sediments from the Okinawa Trough (Kao et al., 2008). These seafloor sediments and trap samples also define a significant negative correlation (P = 0.003, n = 26) between 1/C_{org} and Δ^{14} C_{org} (Fig. 5a). A linear trend between isotopes can result from mixing of two dominant sources (a binary mixture), or may reflect a process 20 by which both sets of isotopes are modified (for example by preferential loss of one component of organic matter). A linear trend between the inverse of concentration and isotope composition may also result from binary mixing, but organic matter loss can result in linear trend which is distinct from mixing.



4 Discussion

4.1 Erosion and transfer of terrestrial OC

Physical erosion processes occur at very high rates in mountain landscapes. In Taiwan, suspended sediment yields reach > $10\,000\,Mg\,km^{-2}\,yr^{-1}$ (Dadson et al., 2003)

- due to a combination of steep slopes and intense precipitation during tropical cyclones. These factors mean that bedrock landslides are common, delivering clastic sediment to mountain rivers (Hovius et al., 2000). Landslides also erode organic matter from the terrestrial biosphere and supply OC, mixed with clastic sediment, to rivers (Hilton et al., 2008; West et al., 2011). In addition, the high runoff intensity promotes mobilisation of soil organic matter by overland flow processes (Hilton et al., 2012). The combination of
- these geomorphic processes results in rates of OC_{non-fossil} transfer that rank amongst the highest in the world (Stallard, 1998; Kao and Liu, 2000; Hilton et al., 2008, 2012; Smith et al., 2013).

The geomorphic processes occurring in Taiwan can explain the triangular domain in $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ values due to mixing of OC_{fossil} and OC_{non-fossil} (Hilton et al., 15 2010). Radiocarbon depletion in the samples can be explained by the input of OC_{fossil} from bedrock. Due to its geological age (> 50 ka), OC_{fossil} from Mesozoic-Cenozoic sedimentary rocks has no measureable ¹⁴C and so has $\Delta^{14}C_{ora} = -1000$ ‰. In addition, OC_{fossil} input can account for the range in $\delta^{13}C_{org}$ values at low $\Delta^{14}C_{org}$ in Taiwan (Fig. 2a), where meta-sedimentary bedrocks have $\delta^{13}C_{org}$ values between ~ -25%. 20 and $\sim -20\%$ (Hilton et al., 2010). Hilton et al. (2010) reported that rivers draining the eastern flank of Taiwan can have higher $\delta^{13}C_{org}$ values than those draining the west due to different bedrock geology leading to variable OC_{fossil} composition. The ¹⁴C-depleted samples are consistent with this observation (Fig. 2a). In contrast, when values of $\Delta^{14}C_{org}$ are high (¹⁴C-enriched) the stable isotope composition is much less 25 variable (Fig. 2a). The $\delta^{13}C_{org}$ values are similar to those of C3 biomass and soil in Taiwanese mountain forest (Kao and Liu, 2000; Hilton et al., 2013) and support previ-



ous work showing that surface soil horizons in Taiwan are young, with a C_{org} -weighted average $\Delta^{14}C_{org} \sim 0 \%$ (Hilton et al., 2008). This reflects the high rates of geomorphic processes in the source area (Hilton et al., 2012).

- The rapid transit times of sediment in Taiwanese rivers, combined with young soils, ⁵ mean that the ¹⁴C content of particulate OC provides a proxy for OC_{fossil} input to the suspended load, in analogy with work from other small mountain river systems (e.g. Komada et al., 2004; Leithold et al., 2006; Clark et al., 2013). Taking $\Delta^{14}C_{org}$ values for bedrock (-1000‰) and $OC_{non-fossil}$ (0‰), we have used an end member mixing model to quantify the fraction of non-fossil OC (Hilton et al., 2008). Results indicate
- ¹⁰ that the flux of particulate $OC_{non-fossil}$ (g s⁻¹) increases with water discharge (Fig. 2b). This confirms a strong climatic control on the erosion and fluvial transfer of $OC_{non-fossil}$ highlighted in several Taiwanese catchments using a slightly different method for quantifying the mixing of OC_{fossil} and $OC_{non-fossil}$ (Hilton et al., 2010, 2012). It reflects the activation of erosion processes (overland flow, gully incision, landsliding) during heavy
- precipitation and supply of OC_{non-fossil} and sediment to rivers when their transport capacity is high (Hilton et al., 2008, 2012). This mechanism is not unique to Taiwan, and has been observed elsewhere when flux and OC source data are both available in tropical (Bass et al., 2011; Clark et al., 2013; Lloret et al., 2013) and temperate mountain forests (Hatten et al., 2012; Smith et al., 2013). A consequence of this behaviour
- is that flood events dominate the particulate flux, with 80–90% of the decadal transfer of OC_{non-fossil} by the Liwu River found to occur during cyclonic storms with return times > 1 yr (Hilton et al., 2008). The importance of storm-triggered floods for particulate transfer is a common feature of small mountain rivers (Townsend-Small et al., 2008; Bass et al., 2011; Milliman and Farnsworth, 2011; Lloret et al., 2013).
- The suspended sediment samples provide good constraint on the compositional range of terrestrial OC delivered directly to the deep ocean by hyperpychal plumes. In addition, because our sample set captures particulate OC from across the mountain belt (Fig. 1) we can assess the likely composition of hypopychal inputs, which may be expected to be a mixture of sediments sourced from individual river catchments. The



average of all river samples (Table S2) is $C_{org} = 0.4 \pm 0.2 \%$, $\delta^{13}C_{org} = -24.9 \pm 0.9 \%$, $\Delta^{14}C_{org} = -660 \pm 250 \%$. The $\Delta^{14}C_{org}$ value suggests that $OC_{non-fossil}$ contributes ~ 30 % of the total particulate OC on average, with OC_{fossil} making up the remaining fraction in agreement with previous estimates from Taiwan (Kao and Liu, 2000; Hilton $_{5}$ et al., 2008, 2010).

4.2 Fate of terrestrial OC offshore Taiwan

To constrain the transfer of terrestrial OC to the marine environment we can use the isotopic and elemental composition of samples from marine sediment traps offshore Taiwan. To assess the longer-term preservation, the composition of seafloor sediments and longer cores can be used. The $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ values of terrestrial OC exported by rivers from Taiwan (Fig. 2a) have been characterised more thoroughly than any previous study (Kao and Liu, 1996; Hilton et al., 2008). With these values in hand, recent marine OC can be distinguished from terrestrial OC due to its higher $\delta^{13}C_{org}$ and

 $\Delta^{14}C_{org}$ (Hsu et al., 2006). However, the assessment of OC provenance is not the same

- ¹⁵ as quantifying preservation. To do that we have used C_{org} values of the sediments as C_{org} values are sensitive to changes in the association of OC with clastic particles and can track OC loss (e.g. Galy et al., 2007a; Blair and Aller, 2012; Cathalot et al., 2013). We note that our investigation of marine samples does not extend to the very deep ocean waters offshore the east coast of Taiwan (Fig. 1). However, O₂ concentrations in
- the un-sampled region are low and comparable to those in the Bay of Bengal at 2000 m water depth (Garcia et al., 2010) and so OC preservation may be higher there due to the lower oxidation potential of these deep waters (e.g. Cai and Sayles, 1996; Galy et al., 2007a) than the sites which form the focus of our study (Fig. 1). As a result, our estimates of terrestrial OC preservation efficiency are likely to be conservative.
- To assess the fate of terrestrial OC delivered to the ocean by rivers during hyperpycnal discharges, we have examined OC collected by the sediment trap moored in the channel thalweg of the Gaoping Canyon, fed by the Gaoping River (Fig. 1). The



sediments trapped during the passage of Typhoon Kalmaegi (17 July 2008) had a range in $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ values consistent with the mixture of terrestrial OC_{fossil} and OC_{non-fossil} observed in river sediments (Fig. 3a), with an average C_{org} very close to the river samples (Fig. 3b). These observations suggest that loss of terrestrial OC during transfer to deep waters ($\sim 600 \,\mathrm{m}$) during this hyperpycnal delivery event has been negligible. The trapped sediment included an organic rich sub-sample (Fig. 3b) with visible woody debris ($C_{org} = 1.6 \%$, $\Delta^{14}C_{org} = -112 \%$). Samples collected from the floor of the Gaoping Canyon after Typhoon Morakot also lie within the terrestrial mixing domain (Fig. 3a). Their Corg values also imply little evidence for terrestrial OC loss (Fig. 3b). Thus, we have found that contrary to a previous study (Kao et al., 2006), 10 loss of OC_{non-fossil} (which would systematically lower $\Delta^{14}C_{org}$ and C_{org}) is not consistent with the data. Together, the trap and core samples suggest efficient transfer and preservation (~100%) of terrestrial OC (both OC_{non-fossil} and OC_{fossil}) in a submarine canyon fed by hyperpycnal flows. Moreover, it appears that the natural buoyancy of some macro-particles of OC_{non-fossil} can be overcome during flood discharges, as ob-15 served in modern source-to-sink settings elsewhere (Leithold and Hope, 1999) and in the geological record (e.g. Saller et al., 2006). This suggests that the density of the turbid river plume may be high enough to effectively sequester woody debris and/or that water logging can occur prior to entrainment or during transport. This observation warrents further investigation. 20

The fate of terrestrial OC away from direct hyperpycnal inputs can be examined using marine samples collected from a wider region around Taiwan (Fig. 1). The core and trap samples from the Okinawa Trough, Taiwan Strait and the Gaoping Shelf displayed significant trends between $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ (Fig. 4a), which distinguish them from the Gaoping Canyon samples (Fig. 3a). However, compared to recent marine OC (OC_{marine}) from the western Pacific (Hsu et al., 2006) they were variably depleted in both ¹³C and ¹⁴C (Fig. 4a). The values cannot be explained by aging and re-suspension of OC_{marine}, because this ¹⁴C-depletion only results in $\Delta^{14}C_{org}$ values of approximately –50 to –100‰ in this setting (Hwang et al., 2010). Instead, the linear trend between



 $\Delta^{14}C_{ora}$ and $\delta^{13}C_{ora}$ values may be indicative of binary mixing between end members with distinct compositions (Komada et al., 2004; Clark et al., 2013). The best fit to the data intersects the average of measured riverine OC (itself a mixture of OC_{fossil} and OC_{non-fossil}) and the values expected for recent OC_{marine} (Fig. 4a). Thus, the first order pattern in the samples collected away from direct hyperpycnal inputs can be explained by mixing OC_{marine} with terrestrial OC.

However, loss of terrestrial OC during marine transfer and deposition may have caused $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ values to evolve towards the composition of OC_{marine}. The linear trend suggests that if this loss has occurred, it has done so in a relatively short period of time, because otherwise $\Delta^{14}C_{org}$ would vary with time and produce a 10 non-linear relationship with $\delta^{13}C_{org}$. To assess the possible loss of terrestrial OC in the marine realm, we model a scenario of instantaneous loss (see Supplement). The results indicate that preferential loss of OC_{non-fossil} (e.g. Kao et al., 2006; Cathalot et al., 2013) produces a negative, linear trend between $\Delta^{14}C_{ora}$ and $\delta^{13}C_{ora}$ (Fig. 4b) which is not consistent with the data. On the other hand, bulk loss of terrestrial OC 15 (both OC_{non-fossil} and OC_{fossil}) can produce the observed positive, linear trend between $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ (Fig. 4b).

To constrain whether mixing or loss is the dominant control on the isotopic composition of the marine samples, we have turned to Corg. The model of bulk terrestrial OC loss, which can explain the $\Delta^{14}C_{org}$ and $\delta^{13}C_{org}$ values (Fig. 4b), cannot repro-20 duce the negative linear relationship between $1/C_{\text{org}}$ and $\Delta^{14}C_{\text{org}}.$ The modelled bulk terrestrial OC loss results in a trend which is perpendicular to that observed in the samples (Fig. 5b) and so the patterns in the data are not consistent with terrestrial OC loss, nor selective (OC_{non-fossil}, Fig. 4b) or pervasive (OC_{non-fossil} and OC_{fossil} Fig. 5b). The scatter around the linear trends in the data means that some terrestrial OC loss 25 may have occurred, but the model outputs suggest that this is <30% of the total (i.e. preservation efficiency > 70%). Instead, the only way to account for the measured isotopic and elemental composition of OC ($\delta^{13}C_{ora}$, $\Delta^{14}C_{ora}$ and C_{ora}) in the offshore

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Discussion

sediments is through a mixture of OC_{marine} and riverine OC (itself a mixture of OC_{fossil} and $OC_{non-fossil}$) (Figs. 4 and 5).

Efficient preservation of terrestrial OC_{non-fossil} and OC_{fossil} in both hyperpycnal and hypopycnal marine sediments (Figs. 3, 4 and 5) is consistent with the high sedi-5 mentation rates in the ocean basins surrounding Taiwan, which result from the tectonic and climate setting. Sedimentation rates in the Southern Okinawa Trough reach > 1 mm vr⁻¹, sustained by fluvial sediment delivery (Hsu et al., 2006). Indeed, if the total suspended sediment flux of \sim 380 Tg yr⁻¹ (Dadson et al., 2003) is mostly deposited within $\sim 100 \,\mathrm{km}$ of the coast ($\sim 150\,000 \,\mathrm{km}^2$), then the average sedimentation rate would be $\sim 2 \text{ mm yr}^{-1}$ (density of 2.2 g cm⁻³). The rapid accumulation of clastic sed-10 iment limits the time over which OC is exposed to oxygen (Burdige, 2005; Galy et al., 2007a). Since the O_2 penetration depth in muddy marine sediments is typically of the order 1-10 mm (Hedges and Keil, 1995; Cai and Sayles, 1996), OC_{non-fossil} deposited offshore Taiwan is probably exposed to O₂ for only a matter of years. Our findings are consistent with oxic marine sediments undergoing rapid accumulation elsewhere, with 15 OC preservation efficiencies of 70-100%, exceeding common preservation rates in other depositional settings (Burdige, 2005; Galy et al., 2007a; Blair and Aller, 2012).

4.3 Implications for the global carbon cycle

After accounting for addition of terrestrial OC_{fossil} and OC_{marine} to the offshore sediments (e.g. Fig. 4), we can now assess how the erosion of terrestrial biomass (OC_{non-fossil}) drives sequestration of atmospheric CO₂. Our findings suggest that in the Taiwan sediment routing system, rivers deliver sediments which contain on average 0.15% of terrestrial OC_{non-fossil} (average C_{org} × average fraction of non-fossil) in hypo- and hyperpycnal river plumes. Given the suspended sediment flux from Taiwan to the ocean of 384 Tg yr⁻¹ (Dadson et al., 2003, 2005), this abundance of OC_{non-fossil}

²⁵ to the ocean of 384 Tg yr⁻¹ (Dadson et al., 2003, 2005), this abundance of OC_{non-fossil} and the estimated preservation efficiencies of terrestrial OC of >70% (see Sect. 4.2) we calculate an OC_{non-fossil} burial flux of 0.5–0.6 TgC yr⁻¹ in basin fills derived from Taiwan. This may be a lower bound if material coarser than ~ 500 µm (i.e. coarser than



most suspended load) contributes importantly to OC_{non-fossil} transfer and burial (West et al., 2011). Normalized over Taiwan's mountain island surface area (35 980 km²), this estimated flux represents an OC_{non-fossil} burial yield of 13–16 MgC km⁻² yr⁻¹. CO₂ sequestration associated with physical erosion of Taiwan appears to be seven times
⁵ more efficient per km² than the Himalayan erosion system, which has a burial yield of ~2 MgC km⁻² yr⁻¹ (Galy et al., 2007a). Our analysis suggests that the OC_{fossil} reburial flux from Taiwan is 0.9–1.1 TgC yr⁻¹, similar to the total OC_{fossil} buried annually in the Bay of Bengal (Galy et al., 2008). OC_{non-fossil} burial from this single mountain island represents up to ~1% of the estimated total annual OC burial in the oceans of between 48 TgC yr⁻¹ (Galy et al., 2007a) and ~100 TgC yr⁻¹ (Schlunz and Schneider, 2000) from only 0.02% of Earth's continental surface.

The islands of Oceania are sediment production hotspots, with suspended sediment yields typically > 1000 Mg km⁻² yr⁻¹ (Milliman and Farnsworth, 2011). The transfer of OC_{non-fossil} together with clastic sediment may enhance the OC burial efficiency (Canfield, 1994; Burdige, 2005; Blair and Aller, 2012) even when materials are delivered by hypopycnal river plumes (Fig. 5). As in Taiwan, terrestrial productivity is high across tropical Oceania and mountain forests contain large stores of OC_{non-fossil} in standing biomass and soil (Dixon et al., 1994). This permits us to extrapolate our observations to provide a tentative estimate of the CO₂-sink associated with the oceanic burial of terrestrial OC_{non-fossil}. Assuming a linear relationship between sediment yield and OC_{non-fossil} yield (e.g. Hilton et al., 2012), the terrestrial OC_{non-fossil} content from Taiwan can be combined with the Oceania sediment export of 7 Pg yr⁻¹ (Dadson et al., 2003; Milliman and Farnsworth, 2011) and the range of preservation efficiencies obtained here (70–100 %) to estimate a burial flux of 8–11 TgC yr⁻¹ from the mountain

²⁵ islands of Oceania. This estimate may is likely to be conservative because the high sediment yields in Taiwan of ~ 9000 Mg km⁻² yr⁻¹ result in a lower percent of OC_{non-fossil} in sediments when compared to other mountain rivers (Leithold et al., 2006; Hilton et al., 2012; Clark et al., 2013; Smith et al., 2013). Alternatively, it could be assumed that the OC_{non-fossil} burial yield from Taiwan (13–16 MgC km⁻² yr⁻¹) holds over the Oceania



area $(2.7 \times 10^{6} \text{ km}^{2})$, giving a burial flux of 35–40 TgC yr⁻¹. This value could be viewed as an upper bound, since the erosion rate of OC_{non-fossil} from Taiwan may be toward the high end of global values (Hilton et al., 2012). The lower conservative estimate of OC_{non-fossil} burial by the erosion of Oceania is globally significant. It represents be-

- tween ~ 10-20 % of previous estimates of the total OC burial in clastic sediments in the oceans (Schlunz and Schneider, 2000; Galy et al., 2007a; Blair and Aller, 2012). Adjustment of these global estimates is warranted and requires further observational constraint on the processes and magnitude of this significant, but underappreciated flux in the global carbon cycle.
- ¹⁰ Our findings suggest that mountain building in Oceania can result in a globally important geological CO₂-sink through erosion of the terrestrial biosphere, OC_{non-fossil} transport by mountain rivers and preservation in marine sediments from hyperpycnal but, importantly, also hypopycnal delivery events (Figs. 4 and 5). This region provides a strong link between tectonic uplift and the C cycle which should influence atmospheric
- ¹⁵ CO₂ concentrations on geological timescales. Importantly, the CO₂ sequestration associated with erosion of OC_{non-fossil} should be sensitive to the coverage of terrestrial biomass in the tropics, which is in part moderated by the available supply of CO₂ for productivity (Norby et al., 2005). In addition, the amount and variability of runoff control the erosion and export of terrestrial OC (Fig. 2b) and clastic sediment by small moun-
- tain rivers (Dadson et al., 2003; Hilton et al., 2008). Therefore, islands of Oceania have the potential to introduce stabilizing feedbacks in the carbon cycle on geological timescales which are presently not considered in Earth System models (Berner, 2006; Archer et al., 2010) nor invoked to explain the evolution of atmospheric CO₂ levels in the Cenozoic (e.g. Pagani et al., 2009). One aspect of this may be the link between warm-
- ²⁵ ing ocean temperatures and the occurrence of extreme tropical cyclones in the western Pacific (Elsner et al., 2008) which deliver terrestrial OC and sediment efficiently to the ocean, as previously hypothesised (Hilton et al., 2008). However, a wider response across Oceania may be felt due to CO₂-fertilization of tropical forests (Norby et al., 2005) while ocean warming also increases runoff and runoff variability in the tropics



away from tropical cyclone influence (Milly et al., 2005). The corresponding increase in terrestrial $OC_{non-fossil}$ export to the oceans from tropical islands may act to mitigate the increase in greenhouse gas concentration, with enhanced CO_2 -sequestration by terrestrial $OC_{non-fossil}$ burial in the ocean. These feedbacks may play a significant role in linking tectonics and climate and their impact on the global carbon cycle that deserves further attention.

Supplementary material related to this article is available online at http://www.earth-surf-dynam-discuss.net/1/177/2013/ esurfd-1-177-2013-supplement.pdf.

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Fig. 1. Location of terrestrial and marine samples from Taiwan and the surrounding ocean used in this study. River sediments were collected during typhoon floods across the island (black circles). Marine samples fed by dispersive fluvial inputs (white symbols) were obtained from box core surface sediments, with the location of the longer piston core MD012403 indicated. Sediments were also acquired from within the Gaoping Canyon (black triangles) which is fed by hyperpycnal river plumes. Sediment traps (indicated by squares within symbols) were deployed in the Okinawa Trough (depths provided) and at 650 m in the channel of the Gaoping Canyon.





Fig. 2. Isotopic compositions and flux of particulate organic carbon for Taiwan's rivers. **(a)** Stable and radioactive isotopic compositions of organic carbon ($\delta^{13}C_{org}$ and $\Delta^{14}C_{org}$, ‰) of suspended sediments from rivers in Taiwan. Analytical errors are smaller than the point size. Samples from catchments draining the two flanks of the mountain belt define a mixing domain between organic carbon from the terrestrial biosphere ($OC_{non-fossil}$) and fossil organic carbon (OC_{fossil}) from bedrocks. **(b)** Instantaneous particulate flux of $OC_{non-fossil}$ (gs^{-1}) as a function of water discharge during floods in the sampled catchments demonstrating a strong hydrological control on $OC_{non-fossil}$ export.





Fig. 3. Composition of marine sediments in the Gaoping Canyon (Fig. 1) fed by periodic hyperpycnal flows. **(a)** Stable and radioactive isotopic compositions of organic carbon ($\delta^{13}C_{org}$ and $\Delta^{14}C_{org}$, ‰). **(b)** Organic carbon concentration (C_{org} , %), with mean ± standard deviation shown by large symbol and whiskers.





Fig. 4. Stable and radioactive isotopic compositions of organic carbon ($\delta^{13}C_{org}$ and $\Delta^{14}C_{org}$, ‰) in marine sediments offshore Taiwan fed by dispersive terrestrial inputs (Fig. 1). (a) White dots denote samples from the trap moorings. The mean terrestrial OC composition delivered by rivers (green circle, whiskers ± S.D.) and the expected composition of recent marine OC (blue box) are shown. The samples display a positive linear relationship (black line, with 95% confidence intervals in grey). (b) Linear relationship displayed in the samples along with the isotopic composition of OC predicted by: (i) mixing marine OC and terrestrial OC (black line and dashes with fraction of terrestrial OC); (ii) loss of terrestrial OC_{non-fossil} starting at fraction terrestrial OC = 0.8 (circles with % loss); (iii) bulk terrestrial OC loss (squares with % loss).





Fig. 5. Radioactive isotopic compositions of organic carbon ($\Delta^{14}C_{org}$, ‰) versus the inverse of OC concentration ($1/C_{org}$) in marine sediments offshore Taiwan fed by dispersive terrestrial inputs (Fig. 1) with symbols as in Fig. 4. (a) All samples show a negative relationship between the variables (r = -0.6; P = 0.003) with a linear fit to all samples apart from those in brackets shown by the blank line ($r^2 = 0.7$; P = 0.0001, grey line is the 95% confidence interval). (b) Linear relationship displayed in the samples along with the OC content predicted by: (i) mixing marine OC and terrestrial OC (black line and dashes with fraction of terrestrial OC); (ii) loss of OC_{non-fossil} (starting at fraction terrestrial OC = 0.8, circles with % loss); (iii) bulk terrestrial OC loss (squares with % loss).



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