

# Global carbon export from the terrestrial biosphere controlled by erosion

Valier Galy<sup>1</sup>, Bernhard Peucker-Ehrenbrink<sup>1</sup> & Timothy Eglinton<sup>1,2</sup>

**Riverine export of particulate organic carbon (POC) to the ocean affects the atmospheric carbon inventory over a broad range of timescales<sup>1–5</sup>. On geological timescales, the balance between sequestration of POC from the terrestrial biosphere and oxidation of rock-derived (petrogenic) organic carbon sets the magnitude of the atmospheric carbon and oxygen reservoirs<sup>6,7</sup>. Over shorter timescales, variations in the rate of exchange between carbon reservoirs, such as soils and marine sediments, also modulate atmospheric carbon dioxide levels<sup>1</sup>. The respective fluxes of biospheric and petrogenic organic carbon are poorly constrained, however, and mechanisms controlling POC export have remained elusive, limiting our ability to predict POC fluxes quantitatively as a result of climatic or tectonic changes. Here we estimate biospheric and petrogenic POC fluxes for a suite of river systems representative of the natural variability in catchment properties. We show that export yields of both biospheric and petrogenic POC are positively related to the yield of suspended sediment, revealing that POC export is mostly controlled by physical erosion. Using a global compilation of gauged suspended sediment flux, we derive separate estimates of global biospheric and petrogenic POC fluxes of  $157_{-50}^{+74}$  and  $43_{-25}^{+61}$  megatonnes of carbon per year, respectively. We find that biospheric POC export is primarily controlled by the capacity of rivers to mobilize and transport POC, and is largely insensitive to the magnitude of terrestrial primary production. Globally, physical erosion rates affect the rate of biospheric POC burial in marine sediments more strongly than carbon sequestration through silicate weathering. We conclude that burial of biospheric POC in marine sediments becomes the dominant long-term atmospheric carbon dioxide sink under enhanced physical erosion.**

The atmosphere is a small reservoir of carbon in comparison with rocks, soils, the biosphere and the ocean<sup>1</sup>. Its size is therefore sensitive to small imbalances in the exchange of C with and between these larger reservoirs. Over long timescales, the continental biosphere is mostly at equilibrium with the atmosphere, because most C fixed by terrestrial photosynthesis is quickly returned to the atmospheric reservoir through respiration<sup>1</sup>. However, rivers deliver to the oceans a fraction of this net primary production (NPP) as POC and dissolved organic carbon (DOC)<sup>2–5</sup>. Although most DOC is quickly returned to the atmosphere through oxidation in estuaries and the ocean, a significant fraction of riverine POC is buried in marine sediments and stored over long timescales. This 'leakage' of carbon from the biosphere–atmosphere loop represents a net sequestration of atmospheric C (ref. 6). Rivers also transfer POC from the rock reservoir (petrogenic organic carbon, OC) to marine sediments, thereby transferring C between two reservoirs disconnected from the atmosphere<sup>7</sup>. During this transfer, oxidation of petrogenic OC represents another leakage of C, in this case towards the atmosphere<sup>8</sup>. The nature and efficiency of riverine export of POC to the ocean thus fundamentally affect the long-term atmospheric C inventory. Despite its importance, global riverine export of POC to the ocean has until now remained poorly con-

strained<sup>2–5</sup>. In particular, the respective global fluxes of biospheric and petrogenic POC remain largely unconstrained. More importantly, the sensitivities and relative magnitudes of global biospheric and petrogenic POC export are not well defined, impeding our ability to quantitatively predict POC fluxes and their impact on the long-term global C cycle under different forcing scenarios.

Developing accurate constraints on fluvial transfer of biospheric POC requires the quantification of, and correction for, petrogenic OC in river sediment. Although the presence of petrogenic OC in river sediments and river-dominated margin sediments has been inferred for decades, its direct and unambiguous detection is quite recent<sup>7,9,10</sup>. Consequently, few quantitative reconstructions of petrogenic OC fluxes exist. However, those reconstructions encompass such diverse river systems as the Amazon, Taiwanese rivers and the Ganges–Brahmaputra system<sup>7,8,11</sup>. Radiocarbon (<sup>14</sup>C) measurements have provided key constraints on petrogenic OC concentrations and fluxes. Exploiting the absence of <sup>14</sup>C in petrogenic OC and its presence in biospheric OC, <sup>14</sup>C measurements on riverine POC combined with additional constraints on the chemical composition of either constituent allow these two key constituents to be differentiated<sup>7,12–18</sup>. Here we use published POC compositional data (including <sup>14</sup>C measurements) to derive a direct, global estimate of the petrogenic OC flux (Methods). These new results, together with published petrogenic OC fluxes, provide a unique compilation of riverine export of petrogenic and biospheric POC to the ocean from 43 river systems that account for 20% of the sediment discharge to the oceans.

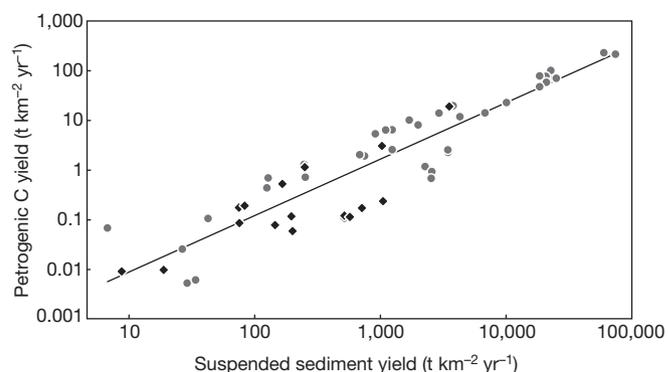
Petrogenic OC is an integral component of sedimentary and other rocks. Its export by river systems is therefore tightly linked to that of sediments. Indeed, both Komada *et al.*<sup>17</sup> and Hilton *et al.*<sup>16</sup> have shown that yields of petrogenic POC (the petrogenic POC flux normalized to catchment area) are positively correlated with corresponding yields of suspended sediment in the Santa Clara River (California) and Taiwanese rivers, respectively, with the latter property serving as a measure of spatially averaged physical erosion rate. Our data set extends this observation to a broad range of river systems, covering more than four orders of magnitude in both catchment size and sediment yield (Supplementary Table 1). These data broadly follow a power-law relationship characterized by a power exponent close to unity ( $1.11 \pm 0.13$ ) (errors are 1 s.d.; see Methods). Petrogenic POC yield thus varies roughly linearly with sediment yield, implying that the behaviour of petrogenic OC during erosion and transport is similar to that of the other mineral phases (Fig. 1). In particular, this finding implies a generally uniform depth-distribution of petrogenic OC in soils and rocks. The average petrogenic POC concentration in river sediments, however, varies considerably (0.02% to 0.6%). This can be explained by variations in the average petrogenic OC content of rocks and/or by variable oxidation of petrogenic OC during sediment transfer to the ocean. Recent studies of large river basins with extensive floodplains (Ganges–Brahmaputra and Amazon) indicate that up to 50% of petrogenic POC initially present in rocks can be oxidized during sediment transport and temporary storage in intermediate reser-

<sup>1</sup>Woods Hole Oceanographic Institution, Department of Marine Chemistry and Geochemistry, 360 Woods Hole Road, Woods Hole, Massachusetts 02543, USA. <sup>2</sup>Geological Institute, Department of Earth Sciences, Sonneggstrasse 5, Eidgenössische Technische Hochschule, 8092 Zürich, Switzerland.

voirs<sup>7,8</sup>. In contrast, petrogenic OC is very efficiently preserved in fluvial systems characterized by rapid sediment transfer to the ocean, such as Taiwanese rivers<sup>19</sup> and the Eel River (California)<sup>20</sup>. Enhanced oxidation of petrogenic OC could explain up to about 50% of the observed order-of-magnitude difference in petrogenic OC concentration. In addition, average petrogenic OC concentrations in rocks have been reported to vary from catchment to catchment by at least an order of magnitude<sup>10,21</sup>. Together, these observations suggest that whereas initial contents in rocks and subsequent oxidation of petrogenic OC during sediment transport together are the dominant controls of petrogenic OC concentration in river sediments, sediment yield—that is, physical erosion rate—is the primary control on petrogenic OC export efficiency.

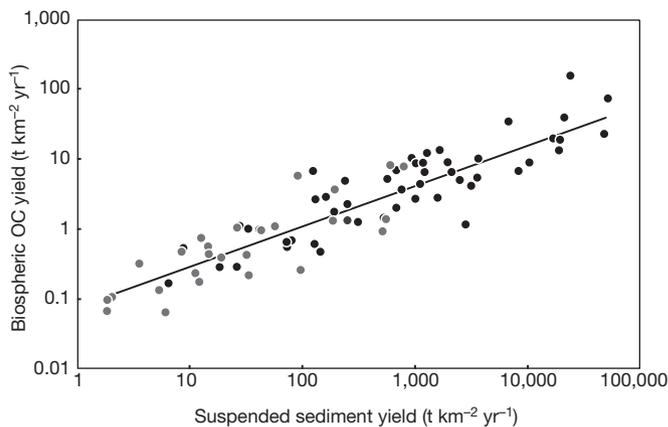
Unlike petrogenic OC, biospheric POC is not an indigenous mineral component of the sediment; instead, it is added during vegetation growth, soil formation and processes associated with the movement of materials from source to sink (for example landsliding or overland flow). The controls on its behaviour could therefore differ from those affecting the mineral load. Because it is the other component of riverine POC, biospheric OC fluxes can be obtained by subtracting petrogenic OC contributions from riverine POC fluxes. Here we use a compilation of riverine POC fluxes from 70 river systems (Supplementary Tables 1 and 2), covering  $42.7 \times 10^6 \text{ km}^2$  (that is, 40% of the total exorheic continental area) and accounting for about 45% of the global freshwater discharge to the ocean. For 27 of these river systems we lack direct estimates of petrogenic OC contributions. We therefore use the relationship between sediment yield and petrogenic OC yield (Fig. 1) to estimate petrogenic contributions to POC in these systems (Methods). This permits the calculation of biospheric POC fluxes and yields for the entire set of 70 river systems. Our data show that biospheric POC yield is positively correlated with suspended sediment yield, following a unique power-law relationship ( $r^2 = 0.78$ ) (Fig. 2). Although relationships between riverine POC and suspended sediment concentrations have been reported previously<sup>2</sup>, we extend this observation to biospheric POC, accounting for its dilution by petrogenic OC at high sediment yield. The singularity of the relationship between biospheric POC and sediment yield is remarkable, considering the very broad range of climate, vegetation, geomorphology and anthropogenic disturbance characterizing the drainage basins considered. It implies that, globally, the rate of biospheric POC export is primarily controlled by sediment export processes. The exponent of the power relationship between biospheric POC and sediment yield is significantly smaller than 1 ( $0.56 \pm 0.03$ ), reflecting an increasing dilution of biospheric POC by mineral phases (that is, decreasing biospheric POC concentrations) at high sediment yield. This reflects the well-documented decrease in OC concentration with depth in soil profiles, which results in an increase in POC stock with depth that globally follows a power law characterized by an exponent of 0.4 (ref. 22). In general, at low sediment yield, erosion proceeds mostly by means of overland flow, exporting surface material (such as soil litter and O horizons) characterized by high biospheric POC concentrations. Conversely, at high sediment yield, erosion proceeds by means of deep gully erosion and/or landslides, thereby lowering the overall biospheric POC concentration of the eroded material<sup>23</sup>.

To further evaluate to what extent productivity and associated soil OC content control biospheric POC export, we used the MOD17 database<sup>24</sup> to extract basin-scale estimates of NPP for 40 systems. Biospheric OC yield and NPP are weakly positively correlated (power law with exponent of 1.16 and  $r^2 = 0.30$ ; Extended Data Fig. 1), suggesting that productivity does not impose a strong control on biospheric OC yield. However, the calculated fractions of the NPP exported from catchments by rivers vary by more than three orders of magnitude (0.01% to 2.1%) and are positively correlated with the suspended sediment yield, following a power-law relationship (Fig. 3) that is characterized by an exponent ( $0.50 \pm 0.05$ ) statistically identical to that of the relationship defined by biospheric POC yield. We there-



**Figure 1 | Relationship between petrogenic OC yield ( $Y_{\text{petro}}$ ) and suspended sediment yield ( $Y_{\text{sed}}$ ).** Catchments larger (black diamonds) or smaller (grey dots) than  $100,000 \text{ km}^2$  plot on the same trend. Most of the variability in petrogenic OC concentration derives from variable initial OC concentrations in rocks and petrogenic OC oxidation during sediment transport. The regression line is  $Y_{\text{petro}} = 0.0007 Y_{\text{sed}}^{1.11}$ ;  $r^2 = 0.82$ ;  $P < 0.001$ .

fore conclude that the rate of biospheric POC export depends primarily on the capacity of rivers to mobilize and transport POC out of catchments, rather than on POC production within the watershed. Our data set also reveals significant secondary variations (Figs 2 and 3), suggesting the existence of additional control mechanisms. Among the possible mechanisms, we postulate that, for a given sediment yield, higher frequency and/or deeper landslides result in decreased biospheric POC yield, illustrating the critical role of physical erosion processes in biospheric POC export. Other possible mechanisms include the sorption of DOC onto mineral phases in sediment-starved rivers (for example the Congo river) that are often characterized by high DOC concentrations. However, the lack of correlation between DOC concentration and biospheric POC yield suggests that sorption of DOC does not exert a significant control on the efficiency of biospheric POC export. The increasing relative variance of biospheric POC yield at a low concentration of suspended sediment—conditions that promote aquatic primary production—suggests that within-river biological productivity could explain part of the secondary variability of biospheric POC yield. Transient non-steady-state erosion of soil and biosphere reservoirs can also introduce some variability in biospheric POC yield that might not be accounted for by either sediment yield or NPP. Finally, human activities such as damming and agriculture currently affect virtually all modern river systems and probably influence biospheric POC yields.

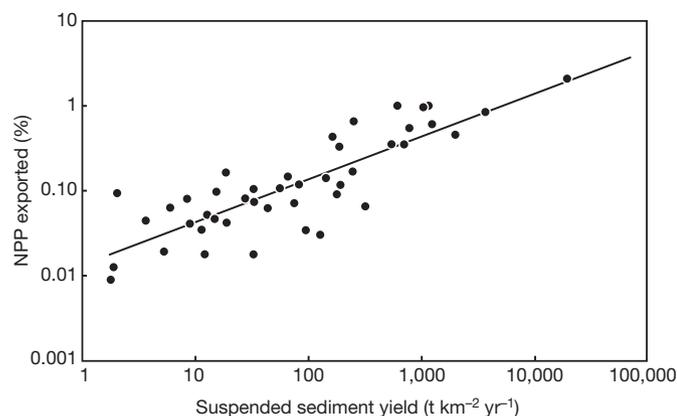


**Figure 2 | Relationship between biospheric POC yield ( $Y_{\text{bios}}$ ) and suspended sediment yield.** Data obtained by subtracting measured petrogenic OC fluxes from riverine POC fluxes (black dots) and those obtained using petrogenic OC fluxes inferred from the relationship shown in Fig. 1 (grey dots) plot on the same trend. The regression line is  $Y_{\text{bios}} = 0.081 Y_{\text{sed}}^{0.56}$ ;  $r^2 = 0.78$ ;  $P < 0.001$ .

The strong relationships between suspended sediment yield and POC yield (both petrogenic and biospheric) allow global POC fluxes to be inferred from the better-constrained global sediment flux. Here we use recent estimates of the suspended sediment flux to the ocean<sup>25–27</sup> to estimate global biospheric and petrogenic POC fluxes. We use a global suspended sediment flux of  $19,000 \pm 500 \text{ Mt yr}^{-1}$  and a corresponding suspended sediment yield of  $176 \pm 8 \text{ t km}^{-2} \text{ yr}^{-1}$  (Methods). In turn, using the overall relationship between suspended sediment yield and biospheric POC yield (Fig. 2), we estimate a global biospheric POC yield of  $1.46^{+0.68}_{-0.47} \text{ t C km}^{-2} \text{ yr}^{-1}$ , which translates into a global biospheric POC flux of  $157^{+74}_{-50} \text{ Mt C yr}^{-1}$ . A similar approach applied to the relationship between suspended sediment yield and the fraction of the NPP exported by rivers (Fig. 3) gives a global value of 0.18% of terrestrial NPP being exported to the ocean. Combined with a mean estimate of the global terrestrial NPP of  $77.6 \text{ Gt C yr}^{-1}$  (ref. 24), this gives a global biospheric POC flux of  $140^{+96}_{-57} \text{ Mt C yr}^{-1}$  that is statistically identical to our estimate based on biospheric POC yield data. Annually, about 0.02% of the total mass of C present in the atmosphere is thus transferred to the ocean as POC, showing that biospheric POC sequestration can affect the size of the atmospheric reservoir over timescales as short as  $10^3$ – $10^4$  years.

The dependence of petrogenic OC yield on OC concentrations in rocks complicates the estimation of the global petrogenic POC flux from our estimate of global suspended sediment yield. Ideally, the distribution of rocks characterized by variable OC concentrations as well as intrinsic geomorphic characteristics (such as the size of the floodplain and the spatial distribution of physical erosion) need to be taken into account, because they both exert strong control on petrogenic OC yields. In the absence of such a model, we can only assume that the 43 river systems that we characterized are representative of the natural variability (that is, rock types and catchment morphology). Using our global suspended sediment yield of  $176 \pm 8 \text{ t km}^{-2} \text{ yr}^{-1}$ , we estimate a global petrogenic POC flux of  $43^{+61}_{-25} \text{ Mt C yr}^{-1}$  (Methods).

Finally, we derive a combined global flux of terrestrial POC to the ocean of  $200^{+135}_{-75} \text{ Mt C yr}^{-1}$ , of which about 80% and 20% are biospheric and petrogenic POC, respectively. This direct estimate of these two fluxes provides an assessment of the magnitude of POC transfer from the terrestrial biosphere to the ocean, and reveals the global significance of petrogenic OC as a component of POC export by rivers to the ocean. However, these fluxes do not take bedload transport into account and must therefore represent a lower bound of actual petrogenic and biospheric POC fluxes to the ocean. Indeed, bedload material can be dominated either by petrogenic OC<sup>7,8</sup> or biospheric POC<sup>28</sup>,



**Figure 3 | Relationship between the proportion of NPP exported annually ( $\text{NPP}_{\text{exp}}$ ) and suspended sediment yield.** Normalization of biospheric POC export to NPP does not remove its dependence on suspended sediment yield, illustrating the overarching control exerted by physical erosion on biospheric POC export. The regression line is  $\text{NPP}_{\text{exp}} = 0.013 Y_{\text{sed}}^{0.50}$ ;  $r^2 = 0.71$ ;  $P < 0.001$ .

implying that bedload transport contributes globally to the fluvial export of both petrogenic and biospheric POC.

On geological timescales, petrogenic and biospheric OC have opposing roles in the global C cycle: the net transfer of C between atmospheric and terrestrial reservoirs is set by the balance between petrogenic OC oxidation and biospheric POC burial. We show that the rate of both petrogenic and biospheric POC export from the continents is controlled primarily by the rate of sediment export; that is, physical erosion. The preservation of both petrogenic and biospheric POC in the ocean is up to three times higher at high physical erosion rates<sup>10,16,19</sup>. Thus, increased physical erosion rates favour efficient transfer and burial of biospheric POC coupled with enhanced preservation (that is, decreased oxidation) of petrogenic POC, both acting to limit the return of carbon to the atmospheric reservoir. The small fraction of NPP exported even at very high erosion rates (a few per cent; Fig. 3) suggests that enhanced export of biospheric POC is sustainable over long timescales, as the terrestrial biospheric OC reservoir is continuously replenished by photosynthetic C fixation. Using available estimates of terrestrial POC burial efficiency, we show that biospheric POC burial yield is positively correlated with sediment yield (Extended Data Fig. 2). Globally, the rate of C sequestration through silicate weathering has a weaker sensitivity to sediment yield (Extended Data Fig. 2) as a result of kinetic limitation of weathering reactions at high physical erosion rates<sup>29</sup>. Biospheric POC burial is thus predicted to become the dominant long-term atmospheric  $\text{CO}_2$  sink under a fourfold increase in global physical erosion rate at constant temperature. We conclude that tectonic and climatic forcing of physical erosion favours biospheric POC sequestration over silicate weathering.

**Online Content** Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

Received 2 September 2014; accepted 9 March 2015.

- Sarmiento, J. & Gruber, N. in *Ocean Biogeochemical Dynamics* (eds Sarmiento, J. & Gruber, N.) 392–453 (Princeton Univ. Press, 2006).
- Ludwig, W., Probst, J.-L. & Kempe, S. Predicting the oceanic input of organic carbon by continental erosion. *Glob. Biogeochem. Cycles* **10**, 23–41 (1996).
- Schlünz, B. & Schneider, R. R. Transport of terrestrial organic carbon to the oceans by rivers: re-estimating flux and burial rates. *Int. J. Earth Sci.* **88**, 599–606 (2000).
- Meybeck, M. in *Interactions of C, N, P and S: Biogeochemical Cycles and Global Change* (eds Wollast, R., Mackenzie, F. T. & Chou, L.) 163–193 (Springer, 1993).
- Degens, E. T., Kempe, S. & Richey, J. E. *Biogeochemistry of Major World Rivers* (Wiley, 1991).
- Berner, R. A. Burial of organic carbon and pyrite sulfur in the modern ocean: its geochemical and environmental significance. *Am. J. Sci.* **282**, 451–473 (1982).
- Galy, V., Beyssac, O., France-Lanord, C. & Eglinton, T. I. Recycling of graphite during Himalayan erosion: a geological stabilization of carbon in the crust. *Science* **322**, 943–945 (2008).
- Bouchez, J. *et al.* Oxidation of petrogenic organic carbon in the Amazon floodplain as a source of atmospheric  $\text{CO}_2$ . *Geology* **38**, 255–258 (2010).
- Blair, N. E., Leithold, E. L. & Aller, R. C. From bedrock to burial: the evolution of particulate organic carbon across coupled watershed–continental margin systems. *Mar. Chem.* **92**, 141–156 (2004).
- Galy, V. *et al.* Efficient organic carbon burial in the Bengal fan sustained by the Himalayan erosional system. *Nature* **450**, 407–410 (2007).
- Hilton, R. G. *et al.* Climatic and geomorphic controls on the erosion of terrestrial biomass from subtropical mountain forest. *Glob. Biogeochem. Cycles* **26**, GB3014 (2012).
- Bouchez, J. *et al.* Source, transport and fluxes of Amazon River particulate organic carbon: insights from river sediment depth-profiles. *Geochim. Cosmochim. Acta* **133**, 280–298 (2014).
- Drenzek, N. *et al.* A new look at old carbon in active margin sediments. *Geology* **37**, 239–242 (2009).
- Galy, V. & Eglinton, T. I. Protracted storage of biospheric carbon in the Ganges–Brahmaputra basin. *Nature Geosci.* **4**, 843–847 (2011).
- Hilton, R. G. *et al.* Tropical-cyclone-driven erosion of the terrestrial biosphere from mountains. *Nature Geosci.* **1**, 759–762 (2008).
- Hilton, R. G., Galy, A., Hovius, N. & Horng, M. J. Efficient transport of fossil organic carbon to the ocean by steep mountain rivers: an orogenic carbon sequestration mechanism. *Geology* **39**, 71–74 (2011).
- Komada, T., Druffel, E. R. M. & Trumbore, S. E. Oceanic export of relict organic carbon by small mountainous rivers. *Geophys. Res. Lett.* **31**, 1–4 (2004).

18. Leithold, E. L., Blair, N. E. & Perkey, D. W. Geomorphologic controls on the age of particulate organic carbon from small mountainous and upland rivers. *Glob. Biogeochem. Cycles* **20**, GB3022 (2006).
19. Kao, S.-J. *et al.* Preservation of terrestrial organic carbon in marine sediments offshore Taiwan: mountain building and atmospheric carbon dioxide sequestration. *Earth Surf. Dyn.* **2**, 127–139 (2014).
20. Blair, N. E. *et al.* The persistence of memory: the fate of ancient sedimentary organic carbon in a modern sedimentary system. *Geochim. Cosmochim. Acta* **67**, 63–73 (2003).
21. Hilton, R. G., Galy, A., Hovius, N., Horng, M. J. & Chen, H. E. The isotopic composition of particulate organic carbon in mountain rivers of Taiwan. *Geochim. Cosmochim. Acta* **74**, 3164–3181 (2010).
22. Jobbagy, E. G. & Jackson, R. B. The vertical distribution of soil organic carbon and its relation to climate and vegetation. *Ecol. Appl.* **10**, 423–436 (2000).
23. Hilton, R. G., Meunier, P., Hovius, N., Bellingham, P. J. & Galy, A. Landslide impact on organic carbon cycling in a temperate montane forest. *Earth Surf. Process. Landf.* **36**, 1670–1679 (2011).
24. Zhao, M., Nemani, Z. & Running, S. (ed. NASA).
25. Milliman, J. D. & Farnsworth, K. *River Discharge to the Coastal Ocean: a Global Synthesis* (Cambridge Univ. Press, 2011).
26. Peucker-Ehrenbrink, B. Land2Sea database of river drainage basin sizes, annual water discharges, and suspended sediment fluxes. *Geochim. Geophys. Geosyst.* **10**, Q06014 (2009).
27. Larsen, I. J., Montgomery, D. R. & Greenberg, H. M. The contribution of mountains to global denudation. *Geology* **42**, 527–530 (2014).
28. Bianchi, T. S., Galler, J. J. & Allison, M. A. Hydrodynamic sorting and transport of terrestrially derived organic carbon in sediments of the Mississippi and Atchafalaya Rivers. *Estuar. Coast. Shelf Sci.* **73**, 211–222 (2007).
29. West, A. J., Galy, A. & Bickle, M. Tectonic and climatic controls on silicate weathering. *Earth Planet. Sci. Lett.* **235**, 211–228 (2005).

**Supplementary Information** is available in the online version of the paper.

**Acknowledgements** We thank Y. Godderis, J. Hemingway and G. Soulet for comments on early versions of the manuscript. G. Fiske generated the NPP data. Support for this project was provided by US National Science Foundation (NSF) grant OCE-0851015 (to B.P.-E., T.E. and V.G.), NSF grant OCE-0928582 (to V.G. and T.E.) and Swiss National Science Foundation grant 200021\_140850 (to T.E.).

**Author Contributions** V.G. designed the study, performed the analysis and drafted the manuscript with inputs from B.P.-E. and T.E.

**Author Information** Reprints and permissions information is available at [www.nature.com/reprints](http://www.nature.com/reprints). The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to V.G. ([vgaly@whoi.edu](mailto:vgaly@whoi.edu)).

## METHODS

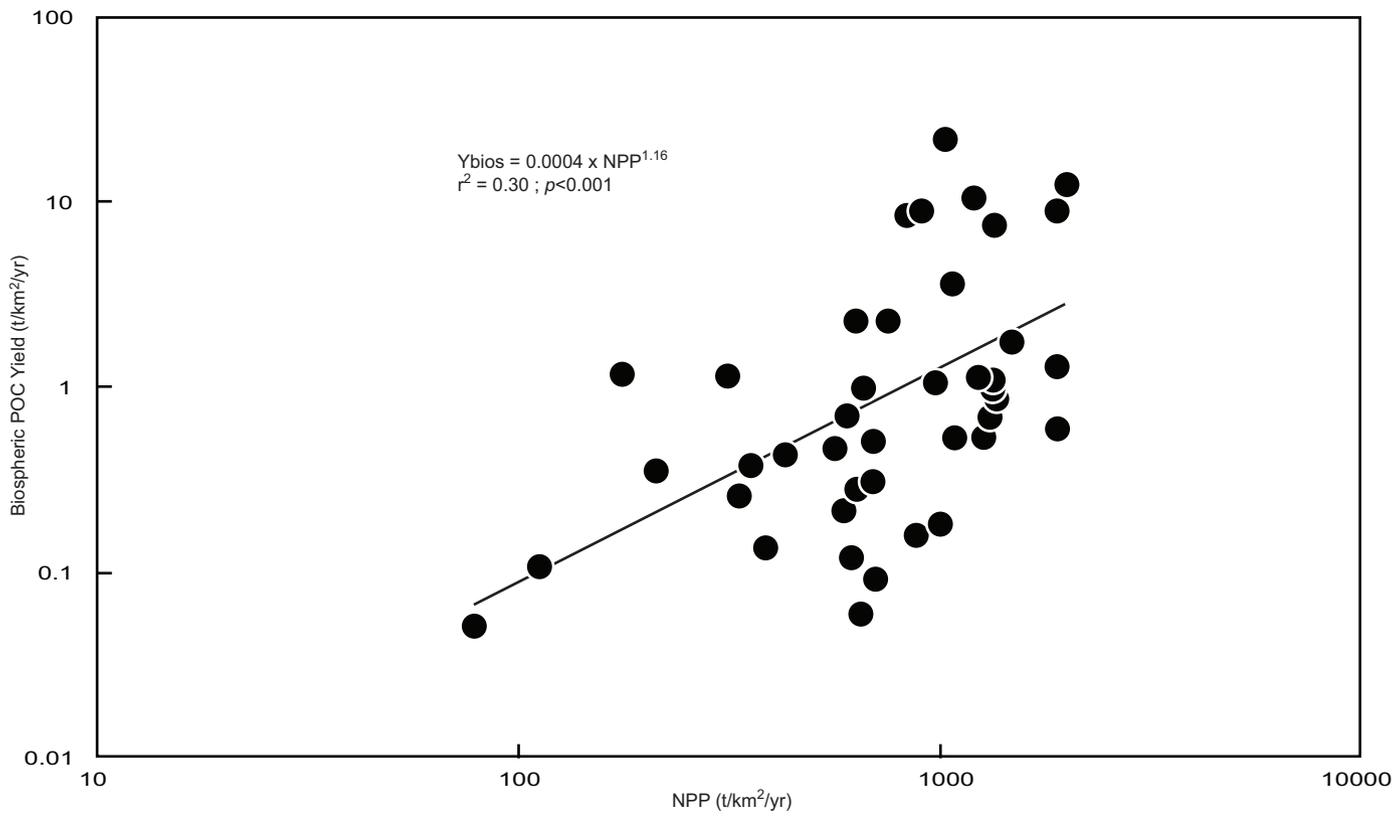
**Quantitative apportionment of biospheric and petrogenic POC in river sediments.** Several methods have recently been used to apportion petrogenic and biospheric POC quantitatively in river sediments. All of these methods are based on the unique property of petrogenic OC that it does not contain any  $^{14}\text{C}$ , whereas biospheric POC does. However, the  $^{14}\text{C}$  content of biospheric POC is difficult to predict a priori, because it depends on a complex array of processes such as physical erosion, soil formation and the dynamics of the biosphere itself. Therefore, in most cases, additional constraints on the chemical composition of biospheric and/or petrogenic OC are needed. Galy *et al.*<sup>12</sup> proposed that the different hydrodynamic properties of petrogenic and biospheric POC in large streams and rivers enables the use of bulk radiocarbon measurements on POC in suspended and bed sediments collected along depth profiles to estimate both petrogenic OC concentration and biospheric POC  $^{14}\text{C}$  content. The underlying assumption is that petrogenic OC is uniformly distributed in the water column (owing to its physical properties and size distribution), whereas the relative concentration of biospheric POC—which is preferentially associated with fine-grained sediments—decreases predictably with depth. This approach has been tested and yields robust results for the Ganges–Brahmaputra<sup>7,14</sup>, Amazon<sup>8,12</sup> and Fraser<sup>30</sup> rivers. Here we reanalyse published  $^{14}\text{C}$  data and show that the approach developed by Galy *et al.*<sup>7</sup> for suspended sediment depth profiles can also be applied to size-fractionated and density-fractionated fluvial sediments as well as to suspended sediments sampled across a wide range of flow conditions. The  $^{14}\text{C}$  content of density-fractionated sediments derived from the Mississippi river<sup>31</sup> and of time-series suspended sediments of the Ishikari river<sup>32</sup> provide excellent examples (Extended Data Figs 3 and 4). This type of binary mixing approach is, however, not always appropriate, either because adequate data may not be available or because of non-systematic behaviour of biospheric POC in the water column. Indeed, biospheric POC often reflects the mixing of several components such as soil and fresh plant debris, which may associate with the mineral load in different ways and have different hydrodynamic properties. Here the Mackenzie river provides a good example. Recently fixed plant-derived biospheric OC and very old permafrost-derived biospheric OC are preferentially associated with coarse and fine sediments, respectively, undermining the binary mixing approach<sup>33</sup>. In these circumstances, additional constraints on the composition of the petrogenic and biospheric end-members are needed. These can be obtained by using bulk (for example N/C,  $\delta^{13}\text{C}$  or  $\delta^{15}\text{N}$ ) or compound-specific ( $^{14}\text{C}$ ) data, as demonstrated for Taiwanese rivers<sup>21</sup>, the Eel River<sup>13</sup> and the Mackenzie river<sup>34</sup>. Once a priori compositions of the petrogenic and different biospheric end-members have been established, simple mixing models can be used to apportion petrogenic and biospheric POC quantitatively. We used a combination of the above-mentioned techniques to evaluate petrogenic OC concentrations in river sediments by using published and newly acquired POC characterizations. Supplementary Table 1 summarizes the sources of the data, the methods employed and the results obtained.

In some cases, POC fluxes have been measured but adequate data to quantify petrogenic and biospheric POC are not available. In these cases (27 rivers; Supplementary Table 2) we estimate the petrogenic OC yield by using the relationship between suspended sediment yield and petrogenic OC yield defined by all rivers for which we could quantify petrogenic and biospheric OC (43 rivers; Fig. 1 and Supplementary Table 1). The significant scatter around this relationship introduces uncertainty in the estimation of petrogenic OC yield from suspended sediment yield. We therefore estimated the uncertainty of calculated petrogenic OC yield on the basis of the uncertainty of the relationship between suspended sediment yield and petrogenic OC yield. Biospheric POC yield is obtained by subtracting inferred petrogenic OC yield from riverine POC yield. The relationship between biospheric POC yield and suspended sediment yield is identical when the two groups of data—that is, data determined from geochemical characterization versus data inferred from the relationship between suspended sediment yield and petrogenic OC yield—are considered separately (Fig. 2). Specifically, the exponents and multiplying terms of the two relationships are statistically identical (within  $\pm 1$  s.d.):  $0.51 \pm 0.04$  and  $0.59 \pm 0.09$  for the exponents, and  $-0.93 \pm 0.13$  and  $-1.19 \pm 0.15$  for the multiplying term. This shows that inferring petrogenic OC yield from the relationship between suspended sediment yield and petrogenic OC yield does not introduce a systematic bias in the determination of biospheric POC yield.

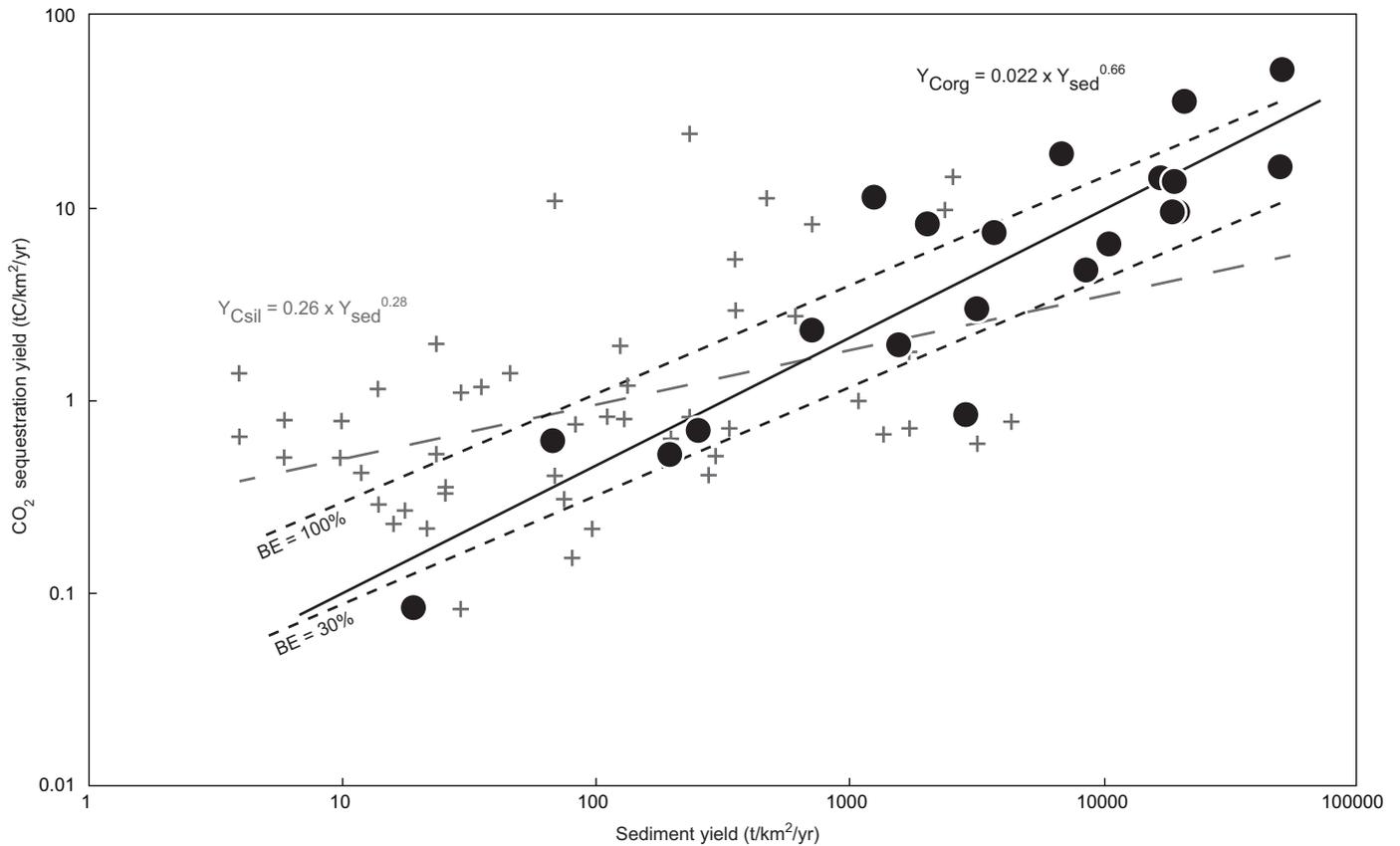
**Estimating global riverine petrogenic and biospheric POC fluxes.** Robust relationships between both petrogenic and biospheric POC yields and suspended

sediment yield enable the use of global suspended sediment yield to estimate global petrogenic and biospheric POC fluxes. Global suspended sediment fluxes have been the subject of extensive research over the past several decades. Suspended sediment fluxes for individual gauged rivers have been compiled<sup>25,26,35,36</sup> and used to derive estimates of global suspended sediment fluxes. Usually, fluxes were first extrapolated regionally (for example by grouping rivers according to the oceanic basin they drain into) to account for regional differences in average suspended sediment yield (for example very high in small ocean islands versus small in the Russian Arctic). Global fluxes were then obtained by summing the regional fluxes for all areas draining into oceanic basins (in other words, endorheic systems are excluded from the global estimate). Peucker-Ehrenbrink<sup>26</sup> and Milliman and Farnsworth<sup>25</sup> have provided the two most comprehensive recent compilations of gauged suspended sediment fluxes. They estimated the global suspended sediment flux to the ocean at 18,548 and 19,000 Mt yr<sup>-1</sup>, respectively. Recently, Larsen *et al.*<sup>27</sup> estimated the global exorheic denudation flux at 19,000 Mt yr<sup>-1</sup>, using an empirical denudation model based on the strong relationship between denudation rates and topography. Here we use a global suspended sediment flux of  $19,000 \pm 500$  Mt yr<sup>-1</sup>. Graham *et al.*<sup>37</sup> estimated the global exorheic land surface at  $110 \times 10^6$  km<sup>2</sup>, whereas Syvitski *et al.*<sup>38</sup> proposed a slightly lower estimate of  $106 \times 10^6$  km<sup>2</sup>. The difference between these two estimates probably derives from differences in corrections for endorheic drainage areas<sup>26</sup>. Here we use an average value of  $(108 \pm 2) \times 10^6$  km<sup>2</sup>, resulting in an estimated global suspended sediment yield of  $176 \pm 8$  t km<sup>-2</sup> yr<sup>-1</sup>. It should be noted that this value attempts to correct, as far as possible, for recent damming of river basins, because pre-dam sediment fluxes were used to estimate the global suspended sediment fluxes whenever possible both by Peucker-Ehrenbrink *et al.*<sup>26</sup> and by Milliman and Farnsworth<sup>25</sup>. Finally, we use the calculated value for global suspended sediment yield ( $176 \pm 8$  t km<sup>-2</sup> yr<sup>-1</sup>) and our relationships between petrogenic and biospheric POC yields and suspended sediment yield to derive estimates of the global petrogenic and biospheric POC fluxes. To estimate the uncertainties associated with calculated global fluxes we first use Monte Carlo simulations (10,000 repetitions) to account for the uncertainty associated with the determination of the relationships between petrogenic and biospheric POC yields and suspended sediment yield. Then we propagate the uncertainty ( $\pm 8$  t km<sup>-2</sup> yr<sup>-1</sup>) in the global suspended sediment yield. Last, we compare the lower bound of the calculated global petrogenic and biospheric POC fluxes with the sum of all petrogenic and biospheric POC fluxes, respectively, and use the highest of the two values as the lower bound of our final estimate.

- Voss, B. M. *Spatial and Temporal Dynamics of Biogeochemical Processes in the Fraser River, Canada: a Coupled Organic-Inorganic Perspective*. PhD thesis, Massachusetts Institute of Technology and Woods Hole Oceanographic Institution (2014).
- Wakeham, S. G. *et al.* Partitioning of organic matter in continental margin sediments among density fractions. *Mar. Chem.* **115**, 211–225 (2009).
- Alam, M. J., Nagao, S., Aramaki, T., Shibata, Y. & Yoneda, M. Transport of particulate organic matter in the Ishikari River, Japan during spring and summer. *Nuclear Instrum. Meth. Phys. Res. B* **259**, 513–517 (2007).
- Hilton, R. G. *et al.* Erosion of organic carbon in the Arctic as a geological carbon dioxide sink. *Nature* (submitted).
- Drenzek, N. J., Montlucon, D. B., Yunker, M. B., Macdonald, R. W. & Eglinton, T. I. Constraints on the origin of sedimentary organic carbon in the Beaufort Sea from coupled molecular  $^{13}\text{C}$  and  $^{14}\text{C}$  measurements. *Mar. Chem.* **103**, 146–162 (2007).
- Milliman, J. D. & Meade, R. H. World delivery of river sediment to the oceans. *J. Geol.* **1**, 1–21 (1983).
- Milliman, J. D. & Syvitski, P. M. Geomorphic/tectonic control of sediment discharge to the ocean: the importance of small mountainous rivers. *J. Geol.* **100**, 525–544 (1992).
- Graham, S. T., Famiglietti, J. S. & Maidment, D. R. Five-minute, 1/2 degrees, and 1 degree data sets of continental watersheds and river networks for use in regional and global hydrologic and climate system modeling studies. *Wat. Resour. Res.* **35**, 583–587 (1999).
- Syvitski, J. P. M., Vorosmarty, C. J., Kettner, A. J. & Green, P. Impact of humans on the flux of terrestrial sediment to the global coastal ocean. *Science* **308**, 376–380 (2005).
- Rosenheim, B. E. *et al.* River discharge influences on particulate organic carbon age structure in the Mississippi/Atchafalaya River System. *Glob. Biogeochem. Cycles* **27**, 154–166 (2013).
- Gaillardet, J., Dupré, B., Louvat, P. & Allègre, C. J. Global silicate weathering and CO<sub>2</sub> consumption rates deduced from the chemistry of large rivers. *Chem. Geol.* **159**, 3–30 (1999).

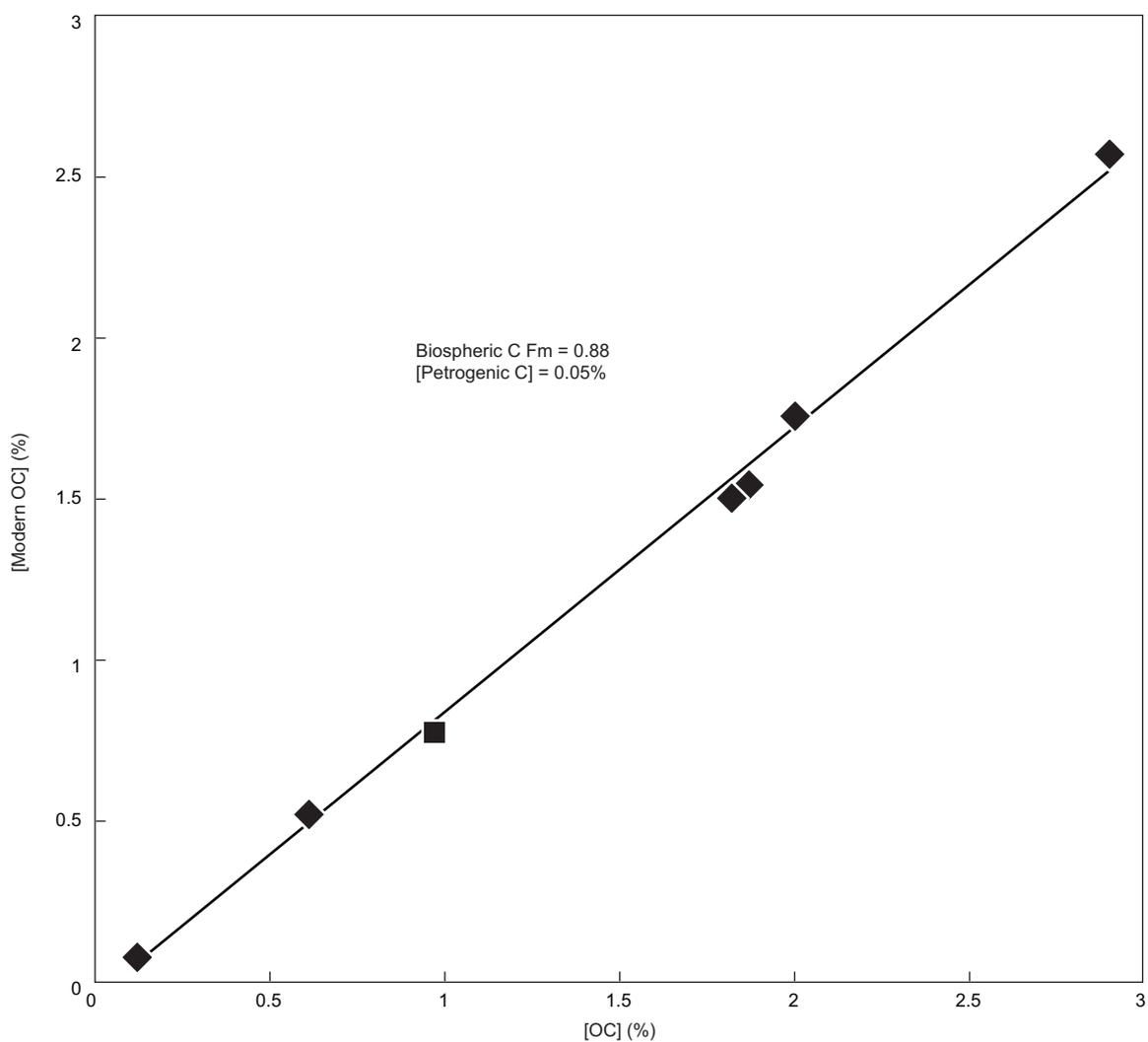


**Extended Data Figure 1 | Global relationship between biospheric POC yield and NPP.** Basin-averaged NPP estimates were derived from the MOD17 database<sup>24</sup>.



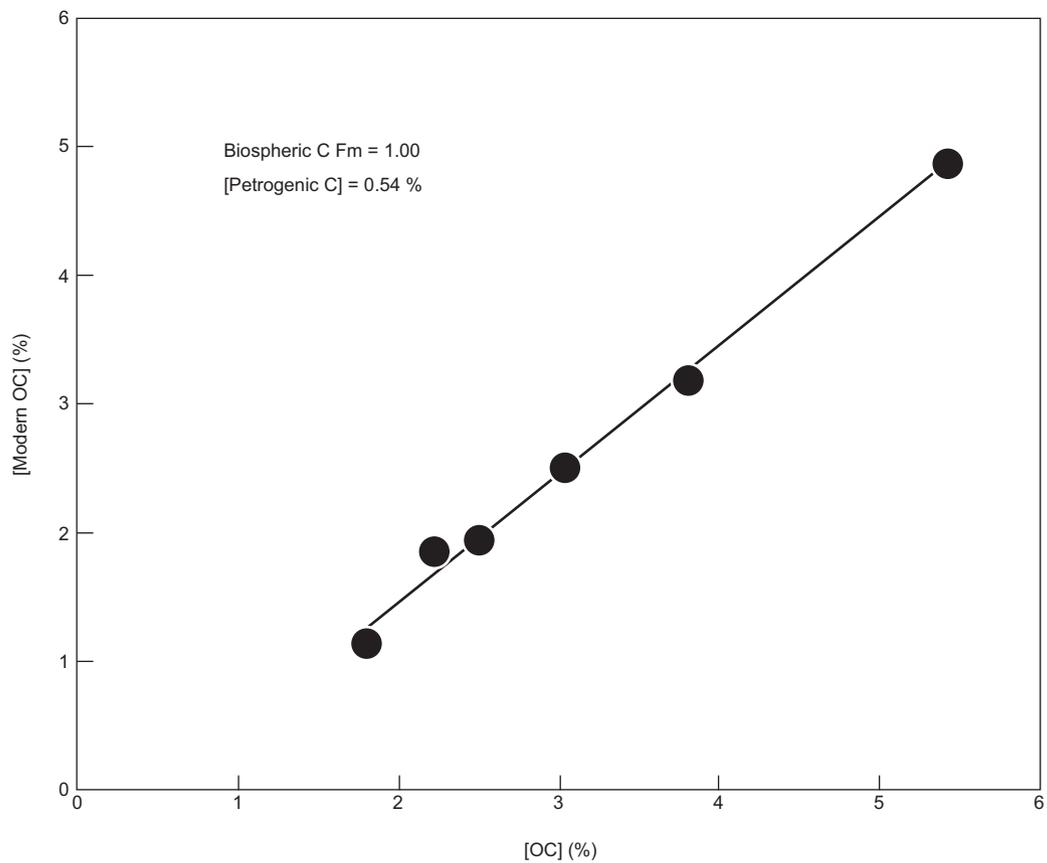
**Extended Data Figure 2 | Global relationship between long-term CO<sub>2</sub> sequestration yield and sediment yield.** CO<sub>2</sub> sequestration through terrestrial biospheric POC burial (black dots;  $Y_{Corg}$ ) is more sensitive to sediment yield than CO<sub>2</sub> sequestration through silicate weathering (grey crosses;  $Y_{Csil}$ ). At high physical erosion rates (that is, high sediment yield), the burial of terrestrial

biospheric POC becomes the dominant long-term atmospheric CO<sub>2</sub> sink. The dotted lines show CO<sub>2</sub> sequestration through terrestrial biospheric POC burial for the entire set of biospheric POC export data (Fig. 2), assuming constant burial efficiencies (BE) of 30 and 100%. CO<sub>2</sub> sequestration data through silicate weathering are from Gaillardet *et al.*<sup>40</sup>.  $P = 0.001$ ;  $r^2 = 0.80$ .



**Extended Data Figure 3 | Organic carbon and radiocarbon contents of bulk suspended sediments and grain size fractions in the Mississippi River.** Results are expressed as modern organic carbon (that is, the product of modern

fraction (Fm) and organic carbon content). The linear best fit gives the absolute petrogenic OC content (0.05%) as well as the Fm of the biospheric POC (0.88). Data from Wakeham *et al.*<sup>31</sup> and Rosenheim *et al.*<sup>39</sup>.  $P = 0.001$ ;  $r^2 = 0.99$ .



**Extended Data Figure 4 | Organic carbon and radiocarbon contents of bulk suspended sediments from the Ishikari River, collected over a wide range of flow regimes.** Results are expressed as in Extended Data Fig. 3. The linear best

fit gives the absolute petrogenic OC content (0.54%) as well as the Fm of the biospheric POC (1.00). Data from Alam *et al.*<sup>32</sup>.  $P = 0.001$ ;  $r^2 = 0.99$ .