## Author responses and changes made in revision

In response to the thorough and thoughtful reviews provided by an anonymous reviewer and Dr. Pizzuto, we have substantially revised the main text of our manuscript and have added a new panel to Figure 1. We feel that these revisions appropriately address all of the comments provided by each of the reviewers and have yielded an overall stronger manuscript. Below, we provide a detailed response to each individual comment from both reviewers along with an explanation of the changes made to the manuscript.

## Reviewer #1

I. 9-10: I find this statement ("sediment transport [. . .] terrestrial realm") a bit weird. Storage in upland soils is clearly also a big player a setting the the maximum time OC persists on land. Although I understand what the authors mean, I do not see why rivers "define" this maximum time, more than upland soils do, for example. I suggest rephrasing.

This is a good point. In addition to during fluvial transport and storage, sediments "age" as they are uplifted by tectonic forces and subsequently transported down hillslopes. We have modified our sentence to reflect these additional controls on sediment ages. Line 37 now reads, *"Thus, rivers influence the amount of time POC can persist within the terrestrial realm and integrate over areas that are large compared to the spatial scales of variability in biogeochemical processes."* A similar change has been made to the abstract.

I. 16 and 77: Doesn't "ameliorates" imply an improvement? If yes, and as I do not see why such "judgmental" word would be needed here, I would rather suggest "modulates", or more simply "affects [too]".

We have replaced "ameliorate" with "limit" in these sentences.

I. 113: Add (if correct) after "transit time distribution": "of this collection of grains".

## We added this suggested phrase.

I. 113-114: I think the physical reason why such transit time distribution is mathematically the result of "n" convolutions of the storage distribution deserves to be more explicitly stated, to keep most readers on board.

For clarity, we modified this sentence on line 114 to read, "Mathematically, the transit time distribution of this collection of grains ( $p_{Tr}(t)$ ), which is the distribution function for the sum of n random values drawn from  $p_s(t)$ , is given by the convolution of the storage distribution by itself n times."

Equation 2: First, this equation does not make much sense mathematically speaking. If  $^{1}4C = f(t)$  (l. 116-117), in no way can f<sup>-</sup>-1(t) be written (f<sup>-</sup>-1 [provided that it can be defined, which requires the f(t) to be monotonic, by the way, which is not necessarily the case for all C = f(t) functions - depending on POC dynamics - although it is the case in the present paper] is a function of  $^{1}4C$  or of something that has the same dimension, at least). In addition, p\_F and p\_Tr, as probability distributions, are dimensionless, unlike dt/d<sup>1</sup>4C: eq. (2) therefore has a unit issue. Second, for the sake of clarity, I think the reason why the derivative term appears on the

right-hand side should be better explicited, again to allow all the readers to understand what the authors are doing.

Very helpful! We thank the reviewer for catching our notation error. The inverse of the function  ${}^{14}C = f(t)$  is  $t = f^{-1}({}^{14}C)$ . Substituting in this correct version of the inverse equation also fixes the issue with the units by canceling  $dt/d^{14}C$ .

I. 147-149: The readers who are not familiar with this model will be interested in knowing how "local and upstream-weighted curvature" influence the local rates of relative channel migration. This is important because later in the paper, it was unclear to me which parameters were directly specified by the authors, and which ones were the result of the model.

We completely agree that the details of the numerical meandering model are relevant to the present study, which is why we dedicated multiple paragraphs in Section 2.2 to describing our application of the Howard and Knutson (1984) model. However, it is beyond the scope of our manuscript to re-state all of the model equations. Instead, readers are referred to the original publication and Limaye and Lamb (2013) for additional model details.

To make our model description clearer and address the reviewer's comment, we added the sentence: "Smoothly curving meander bends initially develop from small perturbations in the channel centerline trace, and migrate at rates that vary in both space and time." to the model description starting on line 155. This, along with addition of other model details requested by both reviewers, should help readers better understand our application of the Howard and Knutson (1984) model.

I. 154-155: I think the sentence "In the model topology [. . .] bed elevation change" should be moved to I. 147 (after "discrete nodes") as it refers to a very general feature of the model that should be given upfront.

We moved this sentence to the beginning of the paragraph as suggested (now starting on line 152).

I. 158-175: These explanations would benefit from an example of how a model result looks like, at a given time step or at then of a simulations (this could be added to Fig. 1, the explanatory interest of which is limited). For example, I still wonder whether these are 2D (maps) structures of the river channel and alluvial deposits?

## Good point. We added a new panel to Figure 1 (panel B) that shows example model output.

I. 158-188: Reading this, I also wondered how the initial conditions (river channel pathway, initial sediment deposit age distribution. . .) of the alluvial plain were defined in these simulations. This comment relates to some missing information I. 185-188 regarding "replicate simulations": how did these replicate simulations differ exactly (e.g. were the initial conditions randomly set for each simulations, and if yes for which subset of parameters?)? And why a set of replicate simulations for storage duration distribution and another set for deposit age distribution? Why 5 in the first set and 50 in the second? These replicate numbers come up again I. 425 and I. 428, but not anywhere else.

Replicate simulations were generated using using different initial channel centerlines generated by adding random perturbations to a straight line. We revised the text to make this clear for the reader. Line 196 now reads, *"Different initial channel centerlines for the replicate model runs were generated by adding random perturbations on the order of 0.01 channel widths to otherwise straight channels. These perturbations also initiate meandering in the model runs".* 

To track the age distribution of sediments being eroded at each time step (storage duration), we needed to modify the code used in Limaye and Lamb (2013) and reprocess the original model results. Therefore, we reprocessed a subset (5) of the original model runs (50) to track storage durations. To make this clearer, the manuscript now states (Line 195), "We used a subset of five replicate simulations to determine storage duration distributions."

I. 196: The T\_cut value of 350 years was obtained from the explicit tracking of meander cutoffs in model runs: is this number actually stable across different replicate simulations (whatever the difference between these replicate simulations, see comment above)? Exactly equal to 350 years? It seems that T\_cut can vary depending on simulation parameters (I. 199-207); so I would guess that this number of 350 years pertains to particular simulation conditions, but this is unclear when reading the article.

This is a valuable point. The  $T_{cut}$  of 350 years pertains to the simulations used to generate the age distributions. We did run additional simulations to determine the relationship between maximum channel migration rates and  $T_{cut}$ , but did not use or report age distributions generated from these additional model runs.

In a single model run, the timescale for meander bend growth and cutoff varies. Based on an ensemble of 3105 cutoffs from an individual model run, we determined that the peak in the probability distribution of cutoff times was equal to 350 years and represented the characteristic  $T_{cut}$  for the given set of model parameters. By measuring  $T_{cut}$  from a large population of individual cutoff events, it accounts for variations in channel trajectories between replicate model simulations (i.e., model runs with identical model parameters but different initial channel centerlines). In the main text, we have added information pertaining to the calculation and selection of  $T_{cut}$  (Line 206), which reads, "While the cutoff time varied in the model runs, the peak of the probability distribution function occurred for Tcut = 350 years".

## I. 216: Add "of" before "two timescales".

## This additional word has been added.

I. 217: "E\_L" has not be defined at this stage (unlike "E\_L,max") - it is defined only I. 238. In addition, the way is retrieved is not clear. I sort of understand that E\_L,max is specified by the operator, but E\_L is measured and is a mean of all values obtained from the model nodes?

We have added a definition of  $E_{L}$  in the text (Line 217). The reviewer is correct in assuming that it is a mean of all values along a river channel.

I. 218: To me, the fact that "relative T\_cut values can be determined [. . .] through comparison of  $w^3/Q_s$ " is implied by the correlation of Fig. 2b, not by the two correlations of Figs. 2a-b.

We cite both figure panels as we feel that correlation between  $T_{cut}$  and lateral migration rates

(panel A) is relevant since only migration rates are directly measured in the field.

I. 242: "n\_x" is not defined anywhere. I guess it is equal to L / x\_tran? If yes, such equation should be added (which requires defining L, defined for now not before I. 256).

Yes, the reviewer is correct in assuming  $n_x$  is calculated from  $L/x_{tran}$ . We have added a definition of L and  $n_x$  in the text (Line 252).

I. 287: Avoid using "lambda" (rate constant for radioactive decay of 14C) here to avoid confusion with "lambda" (wavelength of meander bends) I. 213.

Agreed that it is unfortunate that both radioactive decay constants and wavelengths share the lambda symbol, however because both are so ingrained we are reluctant to use an alternative symbol for the <sup>14</sup>C decay constant as lambda is the standard symbol. However, for clarity, we now use  $\lambda$  to represent the <sup>14</sup>C decay constant and  $\lambda_{bend}$  to represent the meander bend wavelength.

I. 309-310: The concentration of POC does not necessarily have to "increase" with time in a sediment deposit (it will decrease if the initial POC concentration is higher than the steady-state concentration).

This is a great point. We have modified the text accordingly. It now reads, "This equation predicts that the concentration of POC within a sediment deposit evolves towards a steady-state concentration equal to P/k. The e -folding time of this evolution is equal to 1/k." (Line 323).

I. 316: I think the steady-state value of "F\_m" is actually (k\*R)/(lambda+k)/(14C/12C)\_modern (note the division by "(14C/12C)\_modern" compared to what is stated I. 316). (k\*R)/(lambda+k) is rather the steady-state value of "(14C/12C)\_sample\*" (following eq. 6).

Yes. We have modified the text accordingly. It now reads, "When combined with Equation 8, Equation 9 predicts that the Fm of biospheric POC decreases with time to a steady-state value equal to  $k/(\lambda+k)$ " (Line 330).

I. 317: Wouldn't "a given" be more appropriate than "constant" (as "steady state" implies "constant" by definition)?

Yes! We have modified the text accordingly.

I. 325: Note that eq. 2 refers only to 14C dynamics - POC and Fm also need an equation for 12C. For this sentence to be correct, eq. 2 would need to be generalized (using e.g.  $nC = f_n(t)$ ).

Corrections made. The sentence on line 336 now reads, "These integrated equations can then be used to transform any age distribution into a distribution of POC concentrations and Fm (analogously to Equation 2)."

I. 345-347: The relative steady-state values of the fast and slow-cycling POC pools also depend on the production rate P (I. 309 and 316).

Specifying both the steady-state value and the consumption rate constant fixes the production rate.

I. 351: Add a dot after "0.01 yr<sup>-1</sup>".

## Added.

I. 352: "affect" -> "effect".

## Changed.

I. 397: I'm not arguing against the use of a distribution law with finite moments, but in a way many river systems will not recycle all the sediments they store (e.g. in case of floodplain subsidence, as acknowledged by the authors I. 603-616), or at least only over time scales which make the present model pretty irrelevant. Maybe this statement should be altered to reflect this fact.

This point is well taken. We have modified our statement on line 418, which now states, "Since our model simulations show evidence for an upper bound (Figure 3a) and natural river systems have a finite size and, in the absence of external forcing, are expected to eventually recycle more or less all the sediment they store, we employed a tempered Pareto distribution (Cartea and Del-Castillo-Negrete, 2007; Rosiński, 2007) to describe our model results".

I. 421-422: Isn't this ratio of input to output fluxes equal to 1 at "steady-state" (I. 415)? Or does eq. 15 outside of steady state (unlike what is suggested by the way this whole paragraph is written)? And what is this "reservoir" in the "total reservoir size"?

Yes, at steady-state, input and output fluxes are equal. So, either can be compared to the total sediment reservoir size to calculate  $\tau$ . We have modified the text to make this clearer. Line 444 now reads, "... where  $\tau$  is a constant of proportionality that is equal to the ratio of the total sediment reservoir size to either the input or output fluxes."

I. 519-522: I understand that this should be possible "in principle", but in practice this requires that the parameters relevant to the POC dynamics ( $k_S$ , %S...) remain the same across the river course. This is an important requirement, which might not be fulfilled in many, relatively large river systems.

In our sentence, we stated that sampling sediments along a downstream profile could provide samples with variable mean transit times. This statement is independent of POC dynamics. However, we agree that, when interpreting the radiocarbon content of riverine POC, one has to consider spatial variations in OC cycling rates as well as changes in sediment ages. As our sentence, as written, only pertains to sediment transit times, we have opted not to modify it further. However, we now acknowledge the potential complications of spatial variations in OC cycling on line 548 with the sentence, *"However, spatial variations in OC cycling rates may complicate the interpretation of such analyses".* 

I. 603-616: This paragraph points out several limitations of the model, which has to be credited to the authors. However, reading this, I thought that it could be slightly extended to reflect the

fact that the model used focuses on only one type of river morphology, namely meandering rivers. Although I agree that this river type is widespread, other types of river morphology exist (and are especially represented in the dataset shown in Table 2: braided rivers in the Ganges-Brahmaputra, straight or only slightly bended rivers with stabilized banks by persistent vegetation in the Amazin. . .). While the authors acknowledge the limitations of their model in terms of processes that might take place even in meandering contexts (such as overbook deposition), this reference to other river morphologies (in which these other processes might be even more important) is lacking. Additionally, one could also emphasize that the sediment grain dynamics addressed by their model referring to banks subjected to erosion / deposition along the channel, the corresponding results are most likely most relevant for coarse grain sizes. However, the "bulk" POC characteristics measured in a river sediment - especially if this sediment is transported as suspension - might be more reflective of fine grains (often OC-richer) that are affected by other processes such as overbook deposition. This could result in patterns for POC characteristics partly decoupled from meandering dynamics.

This is a good point because our efforts here are focused on rivers where sedimentation is tied to lateral migration of the channel. While this doesn't capture all alluvial river systems that one might like to examine, lateral channel migration is common to many types of river. So, in that way, our results are generalizable. Similarly, while we neglect overbank deposition, field studies show that it is focused near the channel (Pizzuto 1987, Marriot 1992, Aalto et al. 2008). As a result, the dynamics of lateral channel migration should also affect the storage times of sediments deposited via overbank deposition. These points are both stated in the main text (Starting on lines 138 and 630). On line 630, we modified the text to read, "*Presently, our model only accounts for lateral movements of a meandering river with time. In natural river systems, channel pattern (Eaton et al. 2010) and elevation changes driven by overbank deposition, subsidence, and aggradation may also influence the age structure of riverine POC."* 

I. 627-669: Compared to the rest of the article, I find this section a bit weak. First, these "findings" are not related to POC dynamics (especially to processes affecting POC along the lowland river course) at all but rather to grain dynamics, right? And as such they also apply to any tracer deemed as conservative, the signal of which is set in upland rivers and not modified in alluvial plains, right? So why focus the message on biomarkers (which is organic carbon, making the whole point a bit misleading. . .)? And anyway, aren't these "findings" (Fig. 6) a bit textbook? I mean that most readers will probably know that convoluting a periodic signal with some filter that has a reasonable frequency distribution, the original signal will be dampened and offset in phase, with strongest modulations obtained for high frequency (compared to some representative metric of the frequency of the filter)? Therefore, I suggest removing this section and Fig. 6.

We completely understand this point. It is well known that filtering a periodic signal modifies its phase and amplitude. In this section, we sought to illustrate how the mechanics outlined in our study could be used to help identify the shape of the filter most relevant to terrestrial biomarker studies. In previous work on catchment biomarker storage (e.g., Douglas et al. 2014), researchers selected arbitrary filter shapes. While our study is unlikely to provide the exact shape, we suspect that the heavy-tailed dynamics of river systems are imprinted in the age distribution of terrestrial biomarkers.

In the revised version of our manuscript, we have opted to keep Figure 6, but have rewritten the associated text to better emphasize the points mentioned above and the fact that

amplitude dampening / phase lag is a generic outcome of signal filtering. For example, on Line 668, we state, *"While the extent of amplitude damping and phase lag at different frequencies is a generic outcome of signal filtering (Figure 6), our model framework links these effects to physical properties of river systems (e.g., lateral migration rates, sediment fluxes, and channel length)"*. In part, this decision was made because the other reviewer, Dr. James Pizzuto, had a much more positive view of Figure 6 and the associated analysis.

Fig. 3: In panel a, why are the numerical survivor function data (and the corresponding fit) in red on the graph and in black in the legend? Also, It would be informative to represent other functional forms for the possible fit to the numerical data (e.g. exponential. . .). Finally, The red curve should continue as a flat line below t/T\_cut \_ 1.2, at a value of 1. This is not visible on this figure.

We apologize for the labeling confusion and have modified the color scheme appropriately. Additionally, we have added an exponential distribution to panel A of the figure for comparison.

## Fig. 4 caption: I think %S is defined for steady state. Maybe this should be written explicitly.

Good point. This description of the variable has been added to the figure caption.

## Reviewer #2 (Dr. Pizzuto)

1. Line 102 - Please indicate here that you are establishing the usage of an important term. "Transit time" usually refers to the time spent in a "reservoir", which here would represent time spent "waiting" in an alluvial deposit. Here, however, the term is used to describe how long it takes for a particle to traverse a specified distance along a river corridor. I prefer the term "delivery time" for this concept, but whatever term is used should be defined clearly when introduced to avoid confusion.

We agree that the distinction between different age-related variables is crucial and often varies between research groups. Following work in catchment hydrochemistry, we use transit time to refer to the age distribution of material exiting the system, which, for sediments, will be the cumulative time spent "waiting" in alluvial deposits. By defining a characteristic length scale over which sediments are transported before being re-deposited, we link the transport distance and the sediment transit time. However, we do not define the transit time as the time required to travel a specific distance. To make this as clear as possible for the reader in the main text, we have revised the prose to make these definition and distinctions clearer. For example, on line 109 we state, *"Thus, for some number of storage events n, the cumulative age, or transit time, of the sediment can be calculated as the sum of n draws from the probability distribution of storage durations".* 

2. Line 119, equation (2). Please explain where the dt/dC14 term comes from.

This term is necessary to convert a distribution of ages into a distribution of radiocarbon concentrations. After correcting a notation error pointed out by reviewer #1, Equation 2 should now be easier to follow.

3. Line 245. What does this mean? Transit time usually refers to the time spent in a reservoir. So why do they increase downstream? Or does this refer to the number of storage events as

## one moves downstream,? Either way, additional explanation would be desirable.

We expect that, with increased transport distance, sediments will enter and exit alluvial storage more times leading to cumulative aging. As a result, the age distribution of material in the channel, which we refer to as the transit time distribution, will increase downstream. We have modified the text to make this point more clearly. Line 256 now reads, *"By relating the number of transport events to a characteristic length scale (Equation 4), our model predicts that sediment transit times will increase with increasing transport distance due to repeated storage in fluvial deposits."* 

4. Line 252. The Pizzuto et al. reference was published in 2017, not 2016.

We have corrected this typographical error.

5. Line253. The authors might note that Lauer and Parker quote a much larger range in the number of storage times. Also, Pizzuto et al. (2017) note that x\_tran increases with transport distance (scale). Might be worth noting here.

Good point. We revised the prose to note that x\_tran may increase with increasing transport distance on line 266 by stating, *"However, we note that Pizzuto et al. (2017) also predict that the transport length scale increases with increasing river catchment area"*.

6. Lines 320-330. I didn't really understand the description of the mathematics here. More, and clearer, explanation is needed if readers are expected to really understand what the authors are doing here.

This is a really helpful point. In this section, we intended to emphasize the logic behind our approach. Directly transforming the transit time distribution using the OC cycling function (as outlined in Equation 2) would imply that, between storage events, a sediment deposit "knows" to start with the same POC concentration and *Fm* it ended with in the previous storage event. In reality, we expect that, between transport events, POC and sediments sourced from multiple variably aged deposits are homogenized in the active river channel. To account for this mixing, we set the initial POC concentration and *Fm* of a sediment deposit to the average values produced from the preceding storage event. Since this only requires changing the initial values for Equations 8 and 9, we elected not to walk the reader through the arithmetic. However, we have edited this entire section for clarity.

The section, which starts on line 335, now reads, "Equations 8 and 9 can be integrated in order generate equations that predict the concentration and Fm of POC as a function of time. These integrated equations can then be used to transform any age distribution into a distribution of POC concentrations and Fm (analogously to Equation 2). Simply transforming the transit time distribution would imply that, in between transport events, POC sourced from the erosion of variably-aged sediment deposits is not mixed together in the river channel, which is unlikely to be true. Instead, to simulate in-channel mixing, the initial concentration and Fm of POC deposited can be made to vary downstream based on the extent of aging during each transport event.

For the first transport event, the storage duration distribution is transformed with the integrated forms of Equations 8 and 9 where the initial POC concentrations are equal to zero. This

accounts for the POC produced and consumed during the first storage event as sediment transits through a river system. When these upstream sediment deposits are eroded, they will transport a mixture of variably-aged POC downstream until it is re-deposited. We assume that the initial concentration and Fm of POC incorporated into these downstream deposits is equal to the means of the concentration and Fm (weighted by concentration) of eroded deposits from the preceding storage event. These initial values are used to generate new functions from Equations 8 and 9 to transform the storage duration distribution for the next storage event".

# 7. Line 355.Values selected for these parameters seem pretty arbitrary and perhaps not too well justified, but. . .ok.

We readily acknowledge that to an extent these values appear arbitrary because there are few strong empirical constraints. Because of this, we chose values that match the data compilation of Middleburg (1989).

8. Line 415. Please discuss the assumption of a steady state in the methods section. It is common in reservoir theory modeling but a rather extreme prediction for natural fluvial systems. In the rivers intended for this paper to represent, what is the characteristic time scale for a steady state to be achieved? Is this a reasonable assumption? Likely not. Perhaps this merits some discussion. . ..in the discussion section of the manuscript, as well as in the methods section.

This is an important point. While "steady-state" assumptions are often powerful for modeling studies, it's never certain how much value they hold for natural systems. Because of the way that we constructed our model calculations, we do not expect that the simulations do a good job of capturing the processes and timescales associated with dynamic changes in things like sediment fluxes or migration rates.

However, it is also not known by how much the model results would differ between the steadystate and transient case. Consequently, we have not labored to try to extract that type of meaning from our calculations. The point of our approach here is to provide some new context for the interpretation of field measurements. In that sense, our steady-state model is useful in that it identifies measurable parameters that are expected to influence the radiocarbon content of riverine POC. In our compilation of field data, we observed trends consistent with our model predictions, which highlights the utility of a steady-state model despite the expectation that natural systems may not all be at steady-state.

We have now added an additional mention of our assumption of steady state in the methods section on line 178. Specifically, we state, *"Thus, we consider only the time-averaged behavior of the model under steady conditions without any additional forcing."* 

9. Line 563. How is Qs assessed? From stream gaging station records? Are these estimated given in a table somewhere in the manuscript? They should be. More discussion of these data is warranted, also. Generally, useful estimates of Qs are not available.

In Table 2, we report the Qs values that we use in the second column, which is labeled sediment flux. Largely, these values are from sediment gauging records. While there are possible biases with sediment gauging measurements, we compared catchments with sediment fluxes that vary by over five orders of magnitude. We expect that these large, first-

order differences between sites are accurate, which is what we focus on in Figure 5b. On line 486, we now state, "We note that field estimates of sediment fluxes are often imperfect (e.g., Kirchner et al. 2001). However, we expect that large differences in sediment fluxes between field sites, such as those in our data compilation, will provide meaningful insights into differences in sediment storage using Equation 16".

10. Line 573. "though geometric constraints temper or limit the distribution." This is not DEMONSTRATED in the manuscript, it is really simply assumed. The text should be modified to reflect this – it is not a RESULT obtained either from data analysis or computations, but an assumption of the author's approach.

This is a fair point, but in addition to basic expectations for real limits on size, our results support this inference. Our implementation of the numerical meandering model does show an inflection point in the probability distribution of storage durations consistent with exponential tempering (Figure 3a). So, in that sense, tempering is not purely an artifact of our decision to fit the model data with a particular probability distribution. This is now emphasized on line 418 where we state, "*Since our model simulations show evidence for an upper bound (Figure 3a) and natural river systems have a finite size and, in the absence of external forcing, are expected to eventually recycle more or less all the sediment they store, we employed a tempered Pareto distribution (Cartea and Del-Castillo-Negrete, 2007; Rosiński, 2007) to describe our model results".* 

However, we do assume that natural systems also show some degree of tempering, which we do not test in this manuscript. We have modified the manuscript to better reflect this assumption. On line 692, we now state, *"Using simplified models that capture the physical processes associated with sediment storage for meandering rivers, we found that sediment transit times distributions have power-law behavior, though geometric constraints temper or limit the distribution in our model simulations."* 

## 11. Line 919. Pizzuto's name is misspelled here.

We apologize for this typographical error! All corrections have been made to the bibliographic entries.

## 12. Line 927. Correct citation year is 2017, not 2016.

## Fixed.

13. Figure 1. Isn't the length of the valley reach an important variable to consider? How about the geometry of the meandering river domain simulated, perhaps in units of river widths or something? Please explain and clarify. It is also possibly worth noting that the storage time distribution as defined here cannot be measured using observations, unless suspended particles in transport could be "tracked" and dated in some way. It is more elegant to determine the ages of particles as they leave a storage reservoir by dating eroding bank deposits, for example. This definition of storage time can actually be defined by field measurements.

Yes, channel length is an important variable to consider. While we did not explicitly highlight it in Figure 1a, we did evaluate it in the main text. As this figure is highly schematized, it is

difficult to depict the size of the "real" model domain. However, as stated in Limaye and Lamb (2013), "The extent of the model domain parallel to the valley axis scales with the average meander wavelength and is long enough that the channel curvature integration never spans the entire channel centerline". To better highlight the importance of channel length, we included a schematic of  $x_{tran}$  in Figure 1a and, on line 153, state, "The model domain extends for 125 channel widths in the mean downstream direction, and is unconstrained in the mean cross-stream direction".

As we've defined it, storage durations can be measured using time-series of satellite images. Over longer timescales, it should be possible to use fallout radionuclides (e.g., Black et al. 2010) provided that the data can be corrected for inheritance. We've attempted to make this clearer by changing in language on line 147. It now states, *"In order to capture the full range of relevant time and space scales, which are unavailable in existing field observations of lateral migration (Black et al. 2010, Constantine et al. 2014), we used an established numerical model of river meandering (Howard and Knutson 1984, Limaye and Lamb 2013) to derive a processbased probability distribution of storage durations."* 

# 14. Figure 2, panel 2. The range of x\_tran quoted by Pizzuto et al. 2017 is much larger than the data illustrated here. This should be noted in the manuscript.

We appreciate this point, though the differences aren't all that notable. Our definition of transport length scales is distinct from the approach used in Pizzuto et al. 2017, but leads to pretty comparable numbers. For example, the mean  $x_{tran}$  we use is 100 km, which means that sediments are deposited and eroded on average 10 times as they transit down a 1000 km long channel. This compares well with what Pizzuto et al. estimate for 10 storage/erosion events for 1000 km of transport distance. However, like Pizzuto et al. (2017), we expect different river systems to have different transport length scales and we use a mean value only for illustrative purposes. As indicated in a previous response, we added a statement about variable transport length scales in Pizzuto et al. (2017) on line 266.

15. Figure 3. It is odd to show the storage duration data in red, but then present the legend associated with these data in black. Please keep the color scheme consistent.

Helpful point! This color-coding issue have been addressed.

## 16. Figure 6. Great figure!

We thank the reviewer for this support and note that reviewer #1 had the opposite opinion of this figure. Based on both reviewer comments as well as the opinions of all co-authors, we have opted to keep Figure 6 and modify the associated section 4.2 to address reviewer #1's concerns.

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## Model predictions of long-lived storage of organic carbon in river deposits

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### Abstract.

The mass of carbon stored as organic matter in terrestrial systems is sufficiently large to play an important role in the global biogeochemical cycling of  $CO_2$  and  $O_2$ . Field measurements of radiocarbon-depleted particulate organic carbon (POC) in rivers suggest that terrestrial organic mat-

- 5 ter persists in surface environments over millennial (or greater) timescales, but the exact mechanisms behind these long storage times remain poorly understood. To address this knowledge gap, we developed a numerical model for the radiocarbon content of riverine POC that accounts for both the duration of sediment storage in river deposits as well as the effects of POC cycling. We specifically target rivers because sediment transport defines influences the maximum amount of time organic
- 10 matter can persist in the terrestrial realm and river catchment areas are large relative to the spatial scale of variability in biogeochemical processes.

Our results show that rivers preferentially erode young deposits, which, at steady-state, requires that the oldest river deposits are stored for longer than expected for a well-mixed sedimentary reservoir. This geometric relationship can be described by an exponentially-tempered power-law distri-

- 15 bution of sediment storage durations, which allows for significant aging of biospheric POC. While OC cycling partially ameliorates limits the effects of sediment storage, the consistency between our model predictions and a compilation of field data highlights the important role of storage in setting the radiocarbon content of riverine POC. The results of this study imply that the controls on the terrestrial OC cycle are not limited to the factors that affect rates of primary productivity and
- 20 respiration, but also include the dynamics of terrestrial sedimentary systems.

#### 1 Introduction

45

Terrestrial organic matter present in the biosphere, soils, and other shallow sedimentary deposits represents an enormous reservoir of carbon (4 to  $5 \times 10^{12}$  tonnes C; Fischlin et al., 2007) whose dynamics influence atmospheric O<sub>2</sub> and CO<sub>2</sub> concentrations over annual (Keeling, 1960; Keeling

- and Shertz, 1992; Stallard, 1998) to geologic timescales (Bird et al., 1994; France-Lanord and Derry, 1997). Understanding the links between terrestrial organic carbon (OC) cycling and atmospheric O<sub>2</sub> and CO<sub>2</sub> concentrations requires knowledge of the timescales over which OC persists in the environment before being oxidized (Berner, 1989) as well as the underlying processes that set these timescales. Identifying these processes and timescales is challenging because existing measurements
- 30 of OC lifetimes show scale-dependence (i.e., the measured rate of organic carbon oxidation decreases with increasing measurement timescale; Middelburg 1989; Katsev and Crowe 2015). Consequently, measurements of terrestrial OC cycling need to made at large spatial and temporal scales to be quantitatively linked to global biogeochemical cycles.

Large rivers integrate the dynamics of OC cycling at spatial and temporal scales large enough to relate to global biogeochemical cycles. Rivers transport particulate OC (POC) eroded from across their catchment areas to the ocean, where POC is either oxidized or buried in marine sediments (Blair and Aller, 2012). Thus, rivers set-influence the amount of time POC can persist within the terrestrial realm and integrate over areas that are large compared to the spatial scales of variability in biogeochemical processes. Annually, rivers transport large masses of POC to the ocean (1 to 2 ×

40  $10^8$  tonnes C yr<sup>-1</sup>; Galy et al., 2015), making fluvial transport a relevant carbon cycle flux over a range of timescales.

At the scale of river catchments, radiocarbon (<sup>14</sup>C) provides a natural tracer of the lifetime of OC in surface environments. Previous studies have measured large variations in the <sup>14</sup>C/<sup>12</sup>C of riverine POC (Masiello and Druffel, 2001; Martin et al., 2013; Clark et al., 2013; Tao et al., 2015), including significant variations in <sup>14</sup>C/<sup>12</sup>C with depth in river channels (Galy et al., 2008; Bouchez

- et al., 2014). Direct interpretation of measured  ${}^{14}C/{}^{12}C$  in terms of ages would imply that most riverine POC is old (thousands to tens of thousands of years). However, much of the variation in  ${}^{14}C/{}^{12}C$  has been interpreted in the context of variable admixtures of two carbon components: radiocarbon-bearing organic compounds synthesized within the river catchment ("biospheric" POC)
- 50 and radiocarbon-dead organics derived from the erosion of ancient sedimentary rocks ("petrogenic" POC; Masiello and Druffel 2001; Galy et al. 2008).

The erosion of petrogenic POC need not affect the atmospheric budgets of  $CO_2$  and  $O_2$  unless it is oxidized during transit (Bouchez et al., 2010), which releases  $CO_2$  to and consumes  $O_2$  from the atmosphere. Conversely, biospheric POC is a sink for  $CO_2$  and source of  $O_2$  over the course of

55 its lifetime, which is thought to vary widely between different compound classes and environments (Schmidt et al., 2011). While the lifetime of biospheric POC should be reflected in its <sup>14</sup>C/<sup>12</sup>C, determining this ratio from bulk radiocarbon measurements of riverine sediments may be complicated by the mixing of biospheric POC ( ${}^{14}C/{}^{12}C \le atmospheric ratio$ ) with petrogenic POC ( ${}^{14}C/{}^{12}C = 0$ ).

- 60 Compound-specific radiocarbon analyses of terrestrial biomarkers, which can be interpreted as dominantly reflecting the biospheric component, suggest that biospheric POC has a substantially lower <sup>14</sup>C/<sup>12</sup>C than the atmosphere in some large river systems (Galy and Eglinton, 2011; Feng et al., 2013; Tao et al., 2015; Schefuß et al., 2016). These observations require that some biospheric POC is stored in terrestrial reservoirs over (at least) millennial timescales before being transported to
- 65 ultimate depocenters in marine basins. It is reasonable to ask if these observations of aged biospheric POC in modern rivers result, at least in part, from transient storage in river sediment deposits (Galy and Eglinton, 2011; Feng et al., 2013; Tao et al., 2015; Schefuß et al., 2016).

To investigate how storage in river deposits influences the <sup>14</sup>C content of riverine POC, we developed a numerical model that explicitly accounts for the effects of both sediment storage and OC

- 70 cycling. The sediment storage component of our model is focused on how the stochastic nature of fluvial processes leads to the preferential recycling of young sediment deposits (Nakamura and Kikuchi, 1996; Bradley and Tucker, 2013), which, at steady-state, requires that the oldest floodplain-deposits are stored for longer than expected for a well-mixed sedimentary reservoir (Bolin and Rodhe, 1973; Bradley and Tucker, 2013). This geometric relationship engenders a heavy-tailed (power-law) dis-
- 75 tribution of sediment storage durations that may allow for the significant aging of biospheric POC during storage in river deposits and impart a characteristic shape to the distribution of POC ages measured in rivers. However, the production and consumption of POC during floodplain storage can modify POC ages and may serve to ameliorate limit the effects of sediment storage on the POC age distribution. By including both fluvial and biogeochemical processes, our approach provides new
- 80 insights into the interpretation of the terrestrial OC cycle and the radiocarbon content of riverine POC.

Starting from a generic theory for predicting the duration of sediment storage in river systems (Section 2.1), we developed a sediment storage model based on the dynamics of meandering rivers (Sections 2.2 and 2.3). We coupled this sediment storage model to a biogeochemical cycling model

- 85 (Section 2.4) in order to build a full model for predicting the radiocarbon content of riverine POC under different sedimentary and biogeochemical scenarios. Predictions generated using this coupled model were compared with a compilation of field data from diverse global sites, demonstrating that the dynamics of sediment storage in shallow deposits have significant predictive skill in explaining–at least in part–the radiocarbon content of organic matter observed within many rivers
- 90 (Section 3.3.1).

#### 2 Model development

125

#### 2.1 Generic theory for organic carbon and sediment storage

After being eroded from an upland source, fluvial sediments are routed through a transport network that includes multiple temporary storage reservoirs (e.g., channel and floodplain deposits; Malmon et al., 2003; Lauer and Parker, 2003; Lauer and 2003; Lauer and 2003; Lauer and 2004; Lauer and

95 These sediment reservoirs can also store POC, potentially leading to a decrease in its radiocarbon content due to radioactive decay. The magnitude of this effect will depend on: 1) the duration of sediment storage and 2) the rate at which the <sup>14</sup>C content of POC changes with time. Developing simple models for these two factors forms the basis of our approach.

- Sediment grains will spend some portion of their time in transient storage within sediment deposits and the remainder of their time in active transport associated with the river channel. We posit that the time spent in storage is much greater than the time spent in active transport (e.g., Sadler, 1981; Ganti et al., 2011) and, as a result, that the total transit time of sediments from source to sink can be approximated as the total time spent in storage. Since sediment grains likely enter and exit temporary storage reservoirs multiple times during transit, the total storage time can be separated into two
- 105 components: 1) the number of times that grains enter and exit storage reservoirs and 2) and the duration of each storage event.

Fluvial processes are expected to cause natural variations in the duration of each storage event. This variability can be simulated by representing storage duration of a single storage event as a random variable with some probability distribution  $p_S(t)$ . Thus, for some number of storage events

- 110 *n*, the transit time cumulative age, or transit time, of the sediment can be calculated as the sum of *n* draws from the probability distribution of storage durations. Since the sediment load transported by rivers is composed of a collection of sediment grains with different storage histories, it will be characterized by some distribution of transit times that reflects both  $p_S(t)$  as well as the total number of storage events. Mathematically, the transit time distribution of this collection of grains  $(p_{T_T}(t))$  is
- 115 , which is the distribution function for the sum of n random values drawn from  $p_S(t)$ , is given by the convolution of the storage distribution by itself n times

$$p_{Tr}(t) = p_S(t)^{*n} \tag{1}$$

To predict the change in the radiocarbon content of POC that results from sediment storage, the inverse of the functional relationship between the radiocarbon content of POC and time,  $f(t)^{14}C = f(t)$ ,

120 can be used to transform the transit time distribution into a distribution of radiocarbon contents  $(p_F(^{14}C))$  where:

$$p_F(^{14}C) = p_{Tr}(f^{-1}(\underline{t}^{14}C))\frac{dt}{d^{14}C}$$
(2)

We note that, while f(t) must be monotonic, it need not be the radioactive decay equation if POC is continuously produced and consumed during fluvial transit, which is considered in greater detail in Section 2.4.2.

#### 2.2 Application of a meandering model to determine storage duration

Identifying appropriate mathematical expressions for the probability distribution of storage durations is critical to developing realistic models for several geochemical tracers in river sediments, including the radiocarbon content of POC, that have nonlinear changes in concentration with time. Most pre-

- 130 vious models of sediment storage in meandering river systems have assumed that storage durations are exponentially distributed (Malmon et al., 2003; Lauer and Parker, 2008a; Lauer and Willenbring, 2010). This assumption requires that deposits of all ages are equally likely to be eroded at any given time, which is inconsistent with some field data (Nakamura and Kikuchi, 1996; Lancaster and Casebeer, 2007) as well as the common observation that the position of the river channel is persistent
- 135 in time (Bradley and Tucker, 2013). To develop a more realistic storage duration distribution, it is necessary to consider the physical processes that govern sediment exchange in natural river systems. In lieu of following a static path, many rivers migrate laterally across their floodplains with time. Single-thread, migrating (i.e., meandering) channels are one of the most common channel types and show morphodynamic processes common to all alluvial rivers (Dunne and Aalto, 2013). Over time,
- lateral migration of single-thread rivers leads to the development of arcuate meander bends. Meander bends can grow until a cutoff event occurs where the river bypasses an existing portion of its reach in favor of a straighter path (e.g., Hooke, 1995). Together, lateral migration and cutoff allow rivers to traverse back and forth across their floodplains over time and continuously exchange sediments in active transport with those in passive storage. Thus, the spatial and temporal patterns of lateral migration and cutoff directly impact the duration of sediment storage in fluvial systems (Bradley and
- Tucker, 2013).

In order to capture the full range of relevant time and space scales, which are inaccessible in unavailable in existing field observations of lateral migration (Black et al., 2010; Constantine et al., 2014), we used an existing established numerical model of river meandering (Howard and Knut-

- 150 son, 1984; Limaye and Lamb, 2013) to derive a process-based probability distribution of storage durations. The meandering model assumes a fixed channel width and represents the channel position using a series of discrete nodes. The channel is inset in a planar floodplain surface and migrates laterally with no bed elevation change. The model domain extends for 125 channel widths in the mean downstream direction, and is unconstrained in the mean cross-stream direction. At each
- 155 node, local rates of relative channel migration depend upon are calculated based on the local and upstream-weighted upstream- weighted channel curvature. Local rates of lateral migration are then Smoothly curving meander bends initially develop from small perturbations in the channel centerline trace, and migrate at rates that vary in both space and time. Absolute migration rates are computed from the relative channel migration rate, sinuosity, and a user-defined bank erodibility coefficient,
- 160 which sets average lateral migration rate for each simulation. Neck cutoffs occur whenever the channel intersects itself; chute cutoffs are not modeled. We fixed the maximum channel lateral erosion rate at 0.05 channel widths per year, which is typical for actively migrating meandering rivers (e.g.,

Hickin and Nanson, 1975; Constantine et al., 2014), and ran each model simulation for a total of  $10^5$  years. In the model topology, the channel is inset in a planar floodplain surface and migrates

165 laterally with no bed elevation change. We neglect over bank deposition and the loss of sediment due to subsidence, but discuss these in Section 4.1. A schematic of the model is shown in Figure 1.

The numerical model runs yield a time-series of channel positions. For all timesteps, we define areas within the active channel as having a sediment age of zero. As time proceeds and the channel migrates, it abandons sediments along the inner bank (i.e., point bar deposits), which begin to age.

- 170 When the active channel overlaps existing deposits, the time elapsed since the emplacement of those deposits is recorded as the storage duration. Since areas bounded by the channel are defined as having a sediment age of zero, the model does not allow for cumulative aging as a result of multiple deposition and transport events (i.e., once material is eroded, its age is reset to zero). As a consequence, the model does not measure the total amount of time sediments spend in storage (transit
- 175 time), but instead tracks the amount of time spent in storage for a single deposition/erosion event (storage duration).

Using the time-series of channel positions, we calculated storage durations as the ages of river deposits that are eroded by lateral migration at each time step. The storage duration for each eroded deposit is weighted by its areal extent along the active channel, and observations from each timestep

- 180 were combined to yield a full distribution of storage durations for the model. Thus, we consider only the time-averaged behavior of the model under steady conditions without any additional forcing. We note that excluding data from the beginning (e.g., the first 50%) of the model runs does not significantly affect the full distribution since short storage durations are more probable overall. We also examined the age distribution of deposits remaining on the floodplain at the end of the model 185 run which we termed the deposit age distribution
- 185 run, which we termed the deposit age distribution.

Our approach of using a numerical model of river meandering is similar to that of Bradley and Tucker (2013), who developed a quasi-static storage duration distribution from a single meandering model simulation. The river meandering model used in this study (Howard and Knutson, 1984) differs from the Lancaster and Bras (2002) model used by Bradley and Tucker (2013). A salient differ-

- 190 ence between the models is that only the Lancaster and Bras (2002) model has been shown to develop compound bends prior to cutoff. However, similar bend geometries rapidly develop in the Howard and Knutson (1984) model due to meander cutoff. Also, compared to Bradley and Tucker (2013), our analysis uses a suite of model runs with <u>periodic</u> boundary conditions that allow freer channel motion(e.g., the model uses a fixed upstream boundary condition, which limits drift of the mean channel axis; Limaye and Lamb, 201
- 195 including drift of the mean channel axis (Limaye and Lamb, 2013). To systematically account for the sensitivity of the meandering model to the initial conditionschannel planform geometry, which can impact modeled river trajectories (e.g., Frascati and Lanzoni, 2009), we used fifty replicate model runs to determine deposit age distributions. We used a subset of five replicate simulations to determine storage duration distributionsand fifty replicate simulations to determine deposit age

200 distributions... Different initial channel centerlines for the replicate model runs were generated by adding random perturbations on the order of 0.01 channel widths to otherwise straight channels. These perturbations also initiate meandering in the model runs.

#### 2.2.1 Scaling model results to natural systems

- Our meandering model simulations should capture the appropriate shape of the storage duration 205 distribution. However, absolute values should vary as a function of the lateral migration rate. Specifically, rivers that migrate quickly should have, on average, shorter storage durations relative to rivers that migrate slowly. In order to account for the effects of variable migration rates, we normalized all storage durations by the time required for the channel to migrate laterally to the point of meander bend cutoff ( $T_{cut}$ ). By explicitly tracking In our model simulations, we explicitly tracked
- 210 meander bends from growth to cutoff (Schwenk et al., 2015), we find that  $T_{cut}$  is equal to following the approach of Schwenk et al. (2015). While the cutoff time varied in the model runs, the peak of the probability distribution function occurred for  $T_{cut}$  = 350 years in our meandering model simulations. Normalizing the meandering model results by this  $T_{cut}$  value yields a dimensionless storage duration distribution that can be re-scaled to produce variable sediment storage times by varying  $T_{cut}$ .
- To aid in model-data comparisons, it is useful to develop a prediction for how  $T_{cut}$  may vary between river systems. Based on a separate set of meandering model runs with variable maximum lateral migration rates (0.0005 to 0.05 channel widths per year), we found that  $T_{cut}$  is proportional to the inverse of the maximum lateral migration rate such that

$$T_{cut} = c_1 \times \left(\frac{w}{E_{L,max}}\right) \tag{3}$$

- 220 where w is the channel width (meters),  $E_{L,max}$  is the maximum lateral migration rate (meters year<sup>-1</sup>) and  $c_1$  is an empirical constant equal to 13.6 ± 3.3 (Figure 2a). Maximum lateral migration rates ( $E_{L,max}$ ) are used for this comparison because they are specified in the model runs; average rates mean rates ( $E_L$ ) are typically a factor of about-3 lower.
- Field compilations show that mean lateral migration rates are positively correlated with sediment fluxes ( $Q_s$ ; Figure 2; Aalto et al., 2008; Grenfell et al., 2012; Constantine et al., 2014). This correlation may reflect the fact that in order for the river channel to migrate by one channel width, sufficient sediment must be supplied so that a deposit of equivalent volume is created. Along a meander bend, the volume of the sediment deposit produced after a river migrates one channel width should be proportional to the product of the bend wavelength ( $\lambda \lambda_{bend}$ ), channel depth (h), and channel width (w).
- 230 In field data (e.g., Williams, 1986), both  $\lambda \underline{\lambda_{bend}}$  and h are correlated with w such that the deposit volume can be approximated as being proportional to  $w^3$ . In Figure 2, the correlation between lateral migration rates and sediment fluxes (Constantine et al., 2014) is recast in terms of two timescales: the time required to migrate one channel width  $(w/E_L)$  and the time required to supply a proportional volume of sediment  $(w^3/Q_s)$ . Taken all together, the correlations depicted in Figure 2a-b imply that

235 relative  $T_{cut}$  values can be determined for field systems through comparison of  $w^3/Q_s$ . That is, natural rivers with higher  $w^3/Q_s$  appear to migrate more slowly and therefore store sediments for longer.

#### 2.3 Model for the number of storage events

- Following the generic theory presented in Section 2.1, the sediment transit time is equal to the sum of independent draws from the storage duration distribution with the number of independent draws being equal to the total number of storage events (Equation 1). Thus, calculating transit time distributions (TTDs) from the storage duration distribution requires a model for the total number of storage events sediment undergoes during riverine transit.
- Following previous approaches (e.g, Malmon et al. 2003; Lauer and Parker 2008b; Pizzuto et al. 2014), we defined a characteristic length scale over which eroded sediment particles are transported before being re-deposited. While particles are transported variable distances (depending for example on particle size and current velocity), we made the simplifying assumption that the dispersion spread of the distribution of transport lengths is small relative to the mean transport length. We defined the characteristic transport length ( $x_{tran}$ ) by balancing the flux of sediment carried downstream with the
- 250 lateral flux of sediment that results from channel migration. In this way, x<sub>tran</sub> represents the length of channel required to exchange the entire sediment flux with river deposits via lateral migration. This definition of x<sub>tran</sub> is comparable to previous studies (Malmon et al., 2003; Lauer and Parker, 2008b; Pizzuto et al., 2014) and was calculated with the equation:

$$x_{\rm tran} = \frac{Q_s}{E_L \times h} \tag{4}$$

where  $Q_s$  is the volumetric sediment flux (m<sup>3</sup> yr<sup>-1</sup>),  $E_L$  is the mean lateral migration rate (m yr<sup>-1</sup>), and h is the channel depth (m). The appropriate number of transport events  $(n_x)$  can be determined by taking the nearest integer of the ratio of the total channel length (L) to  $x_{tran}$  ( $n_x = \lfloor L/x_{tran} \rfloor$ ). The sediment transit time distribution can then be determined by convolving the storage duration distribution with itself  $n_x$  times (Equation 1).

#### 260 2.3.1 Prediction of downstream changes

By relating the number of transport events to a characteristic length scale (Equation 4), our model predicts an increase in transit times as sediments are transported downstreamthat sediment transit times will increase with increasing transport distance due to repeated storage in fluvial deposits. This relationship between channel length and transit time forms the basis of our comparison be-

tween model results and field data as it allows datasets without direct measurements of sediment transit times to be used. Correlations apparent in field data (Figure 2b) imply that variations in  $x_{tran}$ should be minimal between river systems such that variations in channel length are the dominant control on  $n_x$  (see Equation 4). Here, we set  $x_{tran}$  equal to 100 km, which is close to the mean  $x_{tran}$  (109±68 km) calculated from the data compilation shown in Figure 2c and agrees with a

- complementary, but independent, analysis by *Pizzuto et al.* (2017), which suggests that particles 270 enter and exit storage reservoirs 10 times as they transit along a 1000 km long channel. However, we note that Pizzuto et al. (2017) also predict that the transport length scale increases with increasing river catchment area.
- The predicted changes in sediment transit times with increasing transport length can be cast in terms of catchment area by taking advantage of the power-law relationship between channel length 275 (L; km) and catchment area  $(A; \text{ km}^2; \text{ Hack, 1957})$ . This relationship is useful because catchment areas have been reported for all field data within our compilation (see Section 2.5). We transformed modeled channel lengths using the equation:

 $L = 1.4 \times A^{0.5}$ (5)

280 The value of the exponent selected (0.5) reflects dimensional consistency and a compromise between small ( $< 2 \times 10^5$  km<sup>2</sup>) and large ( $> 2 \times 10^4$  km<sup>2</sup>) river catchments, which are characterized by larger (0.6) and smaller (0.47) exponents, respectively (Mueller, 1973). To model catchments with channel lengths shorter than  $x_{tran}$ , we assumed they have a mixture of POC with a zero age and POC aged by one transport event. The relative proportion of aged material is equal to the ratio of the channel 285

length to  $x_{tran}$ .

#### 2.4 Linking sediment transit times to POC ages

#### 2.4.1 End-member case with zero cycling

To link our prediction of sediment transit time distributions (TTDs) to POC ages, we started by assuming that the age distribution of POC is equal to the sediment TTD. This can be conceptualized as a system where sediments from a source area contain an initial amount of POC with a zero age. 290 The POC and sediments from the source area then transit through a floodplain river system with no subsequent POC oxidation or production. This simplified approach serves as a useful end-member case where POC increases in age as much as allowed by sediment storage. To contrast with this, section 2.4.2 describes an approach that explicitly incorporates the effects of POC cycling into the prediction of POC age distributions.

295

The radiocarbon content of riverine POC is often used as a tracer of the timescale of POC cycling. Typically, radiocarbon measurements are reported as the fraction modern (Fm), which is defined as:

$$Fm = \left(\frac{{}^{14}C}{{}^{12}C}_{sample*}\right) / \left(\frac{{}^{14}C}{{}^{12}C}_{modern}\right)$$
(6)

where the subscript sample\* refers to the ratio of  $\frac{^{14}C}{^{12}C}$  to  $\frac{^{12}C}{^{12}C}$  in a sample normalized to 300 a fixed  $\frac{^{13}\text{C to} ^{12}\text{C ratio} (\delta^{13}\text{C} ^{-13}\text{C to} ^{12}\text{C ratio} (\delta^{13}\text{C} = -25 \%)$  and the subscript modern refers to the  $\frac{^{14}C \text{ to } ^{12}C^{-14}C}{^{12}C}$  to  $^{12}C$  ratio of a standard. Unlike calendar ages, Fm mixes linearly, making it appropriate for use in systems where POC is composed of a mixture of components with different ages.

305 Assuming conservative behavior of POC, the appropriate function to transform the sediment transit time distribution into a distribution of Fm values (following Equation 2) is the radioactive decay equation:

$$Fm = e^{-\lambda t} \tag{7}$$

where  $\lambda$  is the  $\frac{14}{C} \frac{14}{C} \frac{$ 

### 2.4.2 Modeling POC cycling in floodplains

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If POC is produced and/or consumed during floodplain storage, its age will not be exactly equal to the age of the sediment deposit in which it occurs. If, for example, new POC is produced as older POC is consumed, then the radiocarbon content of bulk POC will increase and be shifted to younger ages relative to the sediment deposit. Production and consumption of POC during sediment storage is consistent with existing radiocarbon measurements from soil chronosequences. Importantly, soil chronosequence studies from environments where the parent material contains little to no petrogenic POC show a general decrease in the Fm of biospheric POC with deposit age (Torn et al.,

1997; Lawrence et al., 2015). This relationship implies that, even with active POC cycling, sediment storage will affect the age distribution of riverine POC.

Including the effects of POC cycling in our modeling framework requires a description of the kinetics of POC production and consumption. We adopted a simple approach in order to demonstrate the general effects of POC cycling (based on Jenny et al., 1949). This model assumes that the time

325 rate of change in the POC content of a sediment deposit depends on the balance between POC production and consumption. For simplicity, POC consumption is assumed to be first-order with respect to POC concentrations, which yields the equation

$$\frac{dC}{dt} = P - k(C) \tag{8}$$

where C is the POC concentrations (g cm<sup>-3</sup>), P is the production rate (g cm<sup>-3</sup> yr<sup>-1</sup>), and k is 330 the consumption rate constant (yr<sup>-1</sup>). This equation predicts that the concentration of POC within a sediment deposit increases with time until it reaches evolves towards a steady-state concentration equal to P/k. The *e*-folding time of this increase evolution is equal to 1/k.

To incorporate radiocarbon (as Fm) into this model, it is necessary to write separate versions of Equation 8 for <sup>12</sup>C and <sup>14</sup>C. For <sup>14</sup>C, an additional term is required to account for radioactive decay, which yields the equation:

$$\frac{d^{14}C}{dt} = {}^{14}P - k({}^{14}C) - \lambda({}^{14}C)$$
(9)

When combined with Equation 8, Equation 9 predicts that the Fm of biospheric POC decreases with time to a steady-state value equal to  $(k \times R)/(\lambda + k)$  where R is the ratio of the production rate of <sup>14</sup>C to the production rate of <sup>12</sup>C. For constant  $k/(\lambda + k)$ , For a given steady-state concentrations

340 concentration of POC, the time required for the system to reach steady-state with respect to Fm scales negatively with the POC consumption rate constant (i.e., systems with more slowly cycling carbon require more time to reach a steady-state).

Equations 8 and 9 can be integrated from the minimum to maximum sediment transit time in order to simulate the evolution of POC concentrations (Equation 8) in order generate equations that predict

- 345 the concentration and Fm (ratio of Equations 8 and 9) with time for different POC production and consumption rates of POC as a function of time. These integrated equations can then be used to transform the storage duration any age distribution into a distribution of POC concentrations and Fm for one transport event ((analogously to Equation 2). To model additional Simply transforming the transit time distribution would imply that, in between transport events, the POC sourced from
- 350 the erosion of variably-aged sediment deposits is not mixed together in the river channel, which is unlikely to be true. Instead, to simulate in-channel mixing, the initial concentration and Fm of POC deposited can be made to vary downstream based on the extent of aging during each transport event.

For the first transport event, the storage duration distribution is transformed with the integrated forms of Equations 8 and 9 where the initial POC concentrations are equal to zero. This accounts

- for the POC produced and consumed during the first storage event as sediment transits through a river system. When these upstream sediment deposits are eroded, they will transport a mixture of variably-aged POC downstream until it is re-deposited. We assume that the initial concentration and Fm of POC incorporated into these downstream deposits is equal to the means of the concentration
- 360 and Fm (weighted by concentration) distributions of eroded deposits from the preceding transport eventwere set as the initial values for the integrated forms of storage event. These initial values are used to generate new functions from Equations 8 and 9 used to transform the storage duration distribution. We used this method to account for in-channel mixing, which will tend to homogenize POC in between transport events for the next storage event. Note that the Fm values modeled in this
- manner are not strictly "ages" in any meaningful way, but instead represent the Fm that results from a dynamic balance between POC production, consumption, and radioactive decay.

#### 2.4.3 Accounting for the chemical heterogeneity of POC

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Applying Equations 8 and 9 requires specifying values for the POC production rate and consumption rate constant. Natural OC POC is a compositionally heterogeneous material, and cannot be described

370 by single values for these parameters due to differing rates of biological production and/or resistance to (bio)degradation. To account for this expected heterogeneity, Equations 8 and 9 can sum across multiple POC "pools" with differing production rates (P) and consumption (k) rate constants (Jørgensen, 1978; Berner, 1980; Boudreau and Ruddick, 1991). For POC concentrations, this can be written as:

375 
$$\frac{dC}{dt} = \sum_{i=1}^{j} P_i - k_i(C_i)$$
(10)

where *i* represents an individual POC pool and *j* is the total number of POC pools. An analogous equation can be written for <sup>14</sup>C by adding the term for radioactive decay for each POC pool.

Applying Equation 10 requires specifying the number of POC pools as well as their individual steady-state concentrations and consumption rate constants. A simple version of such a model involves two POC pools (j = 2): "fast" cycling POC and "slow" cycling POC. The fast cycling POC pool has a higher steady-state concentration and a higher consumption rate constant relative to the slow cycling pool. As a result, POC concentrations are dominated by the fast pool while Fm values are more sensitive to the slow cycling pool.

- Using the sediment TTDs we developed, we calculated the bulk radiocarbon content of river-385 ine POC using different parameter values of the 2-pool POC cycling model. In all models, the consumption rate constant for the fast cycling POC pool had a fixed consumption rate constant of was set equal to 0.01 yr<sup>-1</sup>. Similarly, the production rates of fast POC were fixed to produce a steady-state concentration of fast POC equal to 0.1 g cm<sup>-3</sup>. As long as the rate constant for fast cycling POC is greater than 0.001 yr<sup>-1</sup>, its value has little affect effect on the model results as this pool
- 390 cycles rapidly enough to maintain a  $Fm \approx 1$ . To produce POC with a low Fm value, we set the rate constant describing the slow cycling POC pool to either  $2 \times 10^{-6}$  or  $2 \times 10^{-5}$  yr<sup>-1</sup>. Production rates of fast POC were fixed to produce a steady-state concentration of fast POC equal to 0.1 g cm<sup>-3</sup>. Production rates of slow POC were varied such that the steady-state concentration of slow POC was between 5 and 80% of the total (fast + slow) steady-state POC concentration. While consistent
- with some available field data (e.g., Middelburg, 1989), these parameter values were largely selected in order to produce the range of biospheric Fm values observed in natural rivers (see Section 2.5 below).

#### 2.5 Field data compilation and analysis

To benchmark our model results, we compiled field data on the radiocarbon content of riverine 400 POC. As previously mentioned, the bulk radiocarbon content of riverine POC is strongly affected by the mixing of biospheric POC ( $Fm \le 1$ ) with petrogenic POC (Fm=0). Since our model predicts only the change in the radiocarbon content of biospheric POC, it is necessary that we correct for the proportion of petrogenic POC in field data. This is accomplished using a modified version of the Galy et al. (2008) two-component mixing model, which requires datasets with more than 2 measurements

405 of Fm per site.

We identified 50 river systems with more than 2 measurements of the concentration and radiocarbon content of riverine POC (full reference list in Table 1). Following Galy et al. (2008), the bulk Fm measurement can be related to the proportions of petrogenic POC (POC<sub>p</sub>) and biospheric POC (POC<sub>b</sub>) by the equation:

410 
$$Fm_{bulk} \times [POC]_{bulk} = (Fm_b \times [POC]_b) + (Fm_p \times [POC]_p)$$
 (11)

where [POC] is the concentration of POC in units of  $g C g^{-1}$  grams of C per gram of sediment. If it is assumed that petrogenic POC is present at a fixed concentration in sediments, then the relationship:

$$[POC]_p = [POC]_{bulk} - [POC]_b \tag{12}$$

can be substituted into Equation 11. Since the Fm of petrogenic C is equal to zero, the assumptions 415 stated above yield the hyperbolic equation:

$$Fm_{bulk} = \frac{Fm_b \times ([POC]_{bulk} - [POC]_p)}{[POC]_{bulk}}$$
(13)

All the data from each individual site in our compilation were fit with the non-linear form of the mixing equation (Equation 13) using the Trust-Region algorithm available in the MATLAB 2015a Curve Fitting Toolbox. Since Equation 13 predicts a hyperbolic relationship between POC concen-

trations and Fm that is concave down, we screened all of the regression results in order to identify instances where a two-component mixing model was inconsistent with the data. Of the 50 river systems in the data compilation, 21 were consistent with the two-component mixing model (Equation 13) and yielded an estimate of the Fm of bulk biospheric POC (Results shown in Table 2). Of these 21 river systems, we excluded one (The Rhône River; biospheric  $Fm = 1.41 \pm 0.15$ ) from further analysis due to its high Fm, which we attribute to anthropogenic contamination.

#### **3** Results

#### 3.1 Meandering model predictions of storage durations

Averaged across our model simulations, the probability distributions distribution of storage durations show shows a power-law decay in probability as the storage duration increases (Figure 3a). We considered this to be a key feature of the meandering model results, and aimed to capture it in our statistical representation of the modeled age distributions. Without an upper bound, power law distributions can have infinite moments and thus have limited value in describing the full range of behavior of natural systems. Since our model simulations show evidence for an upper bound (Figure 3a) and natural river systems have a finite size and, in the absence of external forcing, are

435 expected to eventually recycle more or less all the sediment they store, we employed a tempered Pareto distribution (Cartea and Del-Castillo-Negrete, 2007; Rosiński, 2007) to describe our model results. The tempered Pareto distribution displays is characterized a power-law decay until some upper limit where it becomes exponentially tempered scale and it is exponentially tempered beyond this scale. Relative to the truncated Pareto distribution (Mantegna and Stanley, 1994), which simply 440 has a fixed eutoff at some upper limitupper bound, the tempered Pareto distribution is less restrictive as it allows the upper bound to also be a an exponentially-distributed stochastic quantity.

Using the approach outlined in Meerschaert et al. (2010), we fit the dimensionless storage duration distribution determined from the meandering model simulations to a tempered Pareto distribution (p(t); Figure 3a), which has a probability density function (pdf) given by:

445 
$$p(t) = \gamma t^{-\alpha - 1} \times e^{-t/\beta} \times (\alpha + t/\beta)$$
(14)

where t is the storage duration,  $\gamma$  is a scale parameter,  $\alpha$  is a tail-index, and  $\beta$  is a tempering parameter. The  $\gamma$  parameter relates to the lower bound of the probability distribution and thus sets the minimum storage time. The  $\alpha$  parameter describes the power-law decay in the relationship between probability and storage duration. The  $\beta$  parameter describes the storage duration at which power-law behavior ceases and the storage duration distribution begins to follow an exponential function. Our

- 450 behavior ceases and the storage duration distribution begins to follow an exponential function. Our best-fit values of  $\gamma$ ,  $\alpha$ , and  $\beta$  are 1.2, 0.8, and 120. We used equation 14 with these values as the storage duration distribution in order to calculate the sediment TTDs used in all subsequent results (Equation 1).
- At steady-state, the storage duration distribution can be uniquely related to the ages of sediment deposits remaining in storage after one transport event (Bolin and Rodhe, 1973; Bradley and Tucker, 2013). Specifically, Bradley and Tucker (2013) demonstrated that the pdf of deposit ages ( $p_A(t)$ ) is proportional to the survivor function (or complementary cumulative distribution function) of storage durations ( $S_S(t)$ )

$$p_A(t) = \tau \times S_S(t) \tag{15}$$

- 460 where  $\tau$  is a constant of proportionality that is equal to the ratio of the input/output fluxes to the total reservoir size total sediment reservoir size to either the input or output fluxes. Given the relationship between density and survivor functions, Equations 14 and 15 make a prediction for the relationship between the tail-indices of the of the deposit age ( $\alpha_d$ ) and storage duration ( $\alpha_s$ ) distributions. Specifically,  $\alpha_d = 1 - \alpha_s$  at steady-state. Individually fitting the results of each of our five replicate
- 465 simulations yields  $\alpha_s$  values that range from 0.8 to 1. This range of  $\alpha_s$  values, which we assume represents the uncertainty of our estimate, predicts a range of  $\alpha_d$  values that overlaps with our bestfit estimate of  $\alpha_d$  from fitting Equation 14 to the deposit age distribution derived from fifty replicate model simulations ( $\alpha_d = 0.1$ ; Figure 3a). This consistency between the storage duration and deposit age distributions suggests that our model simulations were at steady-state with respect to sediment
- 470 storage statistics.

By using the tempered-Pareto distribution (Equation 14) to represent the distribution of storage durations, we excluded storage durations less than the lower bound, which is related to the  $\gamma$  parameter and approximately equal to one cutoff time. In our model simulations, the proportion of the age distribution with storage durations less than the lower bound is small (Figure 3a), which implies

that the lower bound imposed by the tempered-Pareto distribution may not significantly effect our modeled transit time distributions. Similarly, for modeling POC cycling, a fixed lower bound may be a reasonable approximation since soil chronosequence studies imply that the onset of significant organic carbon accumulation is lagged relative to the time of sediment deposition (Torn et al., 1997; Masiello et al., 2004; Lawrence et al., 2015). Consequently, the small proportion of young sediments not included in our model is not expected to participate significantly in organic carbon cycling.

#### 3.1.1 Non-dimensional model behavior

To show the general model behavior, we started by comparing the dimensionless sediment TTDs generated using Equations 1 and 14 for 1 to 20 transport events (Figure 3b). As expected, our calculations show that sediment ages increase with the number of transport events. They also show that 485 the shape of the transit time distribution changes with increasing number of transport events (Figure 3c). This change in shape is due to the central limit theorem, which states that sum of independent random variables tends towards a normal distribution even when drawn from a distribution that is not normal. The central limit theorem applies in this case because the tempered-Pareto distribution has finite moments due to the exponential tempering of the longest storage durations. However, as a semi-heavy tailed distribution, sums of tempered-Pareto variables take longer to converge to a normal distribution relative to exponentially-distributed variables. As a result, sediment TTDs skewed

- towards older ages are expected despite the mixing effects of multiple transport events (Figure 3b,c). The dimensionless sediment TTDs show a roughly linear increase in the mean transit time (MTT)
- with increasing number of transport events (Figure 3b). Assuming that  $x_{tran}$  is relatively constant, 495 which is consistent with field data (Figure 2c), the number of transport events should increase with the ratio of the square root of catchment area to  $x_{tran}$  (Equation 5; Hack, 1957). This metric for the relative number of transport events ( $\sqrt{A}/x_{tran}$ ) can be combined with our metric for relative  $T_{cut}$ values ( $w^3/Q_s$ ; Section 2.2.1) to produce a metric for the relative transit time of sediments where:

Relative Transit Time 
$$\underline{=} \underbrace{\propto n_x T_{cut}}_{x_{tran}} \underbrace{\frac{\sqrt{A}}{x_{tran}}}_{Q_s} \underbrace{\frac{w^3}{Q_s}}$$
 (16)

- 500 This simplified metric captures the expected effects of sediment supply (Constantine et al., 2014) and channel length (Malmon et al., 2003; Lauer and Parker, 2008b; Pizzuto et al., 2014) on the duration of sediment storage in river deposits and is useful for comparing systems where more direct measurements of sediment ages are unavailable. We note that field estimates of sediment fluxes are often imperfect (e.g., Kirchner et al., 2001). However, we expect that large differences in sediment
- 505 fluxes between field sites, such as those in our data compilation (Table 2), will provide meaningful insights into differences in sediment storage using Equation 16.

#### 3.2 Coupling sediment storage to POC cycling

#### 3.2.1 Radiocarbon as a POC storage tracer

The radiocarbon content of biospheric POC is a tracer of the lifetime of POC in surface environ-510 ments, which reflects both the rate of POC cycling and the duration of time over which these reactions occur. For a sediment deposit with a single age, the bulk radiocarbon content (as Fm) is set by the POC production rate, the POC consumption rate constant, as well as the deposit age (Equation 9). For river sediments, which are composed of a mixture of variably-aged deposits (Figure 3), it is expected that the relationship between the bulk Fm, POC cycling parameters, and storage time

- 515 will depart significantly from the behavior expected for a single-age deposit (Equation 9). The magnitude of the difference between the heterogeneous (sediment TTD) and homogeneous (single-age deposit) cases depends on the transit time distribution as well as the values of the POC consumption rate constants. The direction and magnitude of the offset is important to constrain as it underlies the quantitative interpretation of field data.
- In analyzing the model predictions, we start by comparing the predicted relationships between the mean transit time (MTT) and bulk Fm for heterogeneous systems with a distribution of sediment transit times versus homogeneous systems with a single transit time. For this analysis, we re-scaled the dimensionless sediment TTDs shown in Figure 3b using  $T_{cut}$  values selected such that the MTT for each distribution shape varied between 10<sup>3</sup> and 10<sup>6</sup> years. This approach is consistent with field estimates of MTTs, which range from 10<sup>3</sup> to  $5 \times 10^5$  years when measured using sediment budgeting
- (Blöthe and Korup, 2013) or radionuclide approaches (Dosseto et al., 2006; Granet et al., 2010; Wittmann et al., 2015, 2016; Li et al., 2016).

The exact difference in Fm between the heterogeneous and homogeneous cases depends on the MTT (Figure 4a). When the MTT is long relative to the time it takes OC cycling to reach steady-state,

- 530 there is no difference between the Fm predicted by the heterogeneous and homogeneous models. However, when a significant portion of the sediment TTD consists of transit times less than the time required to reach steady-state with respect to OC cycling, there are significant differences between the Fm predicted by the heterogeneous and homogeneous models. For the cases considered here, these differences can approach 0.1 Fm units (Figure 4a), which is two orders of magnitude greater
- 535 than typical analytical uncertainties. Consequently, applying models based on homogeneous systems (e.g., Equations 8, 9, and 10) to radiocarbon measurements of riverine POC may yield parameter values that are off by large factors relative to their true values. In particular, applying homogeneous models to riverine POC is likely to yield apparent OC cycling rates that are fast relative to the "true" rates due to the significant proportion of sediments with short transit times.
- 540 While heterogeneous sediment TTDs lead to quantitatively distinct relationships between sediment MTTs and POC Fm values, the large differences in parameter values we selected allow the OC cycling models to be distinguished from one another despite the differences induced by variable

TTD shapes (Figure 4a). When there is only a very small fraction (i.e., 5%) of slow OC, bulk Fm values remain close to 1 at all MTTs (Figure 4a). Larger portions of slow OC result in more variable

545 Fm values. For systems with identical values of POC consumption rate constants, increasing the portion of "slow" OC decreases the Fm observed at a given MTT (Figure 4a).

In general, OC cycling models with lower POC consumption rate constants (i.e., slower OC cycling) have lower steady-state Fm values, but take longer to reach steady-state. As a result, for intermediate MTTs, the bulk Fm of riverine POC can have a higher value for systems with slower POC

- 550 cycling rates relative to those with faster cycling rates (Figure 4a). This counter intuitive result stems from mass balance constraints within the model, which require more slowly cycling compounds to be produced at slower rates if their concentrations are to remain a fraction of the total POC concentration. In other words, for the same MTT, more slowly cycling POC produced at a slower rate can yield a similar bulk Fm to more rapidly cycling POC produced at a faster rate (Figure 4).
- Results show that a single observation of the bulk Fm of riverine POC can yield a non-unique interpretation of the underlying POC cycling dynamics even if the sediment TTD is known independently (Figure 4a). While this is also true when analyzing soil POC, the chronosequence approach (i.e., analyzing soils of variable, but independently known ages from the same site) can be used to better distinguish between different models. Our analysis highlights an analogous approach that can
- theoretically be applied to river systems where bulk Fm values can be compared within a given river system or between similar river systems with differing sediment transit times. In our results, OC cycling rates are distinguished by the shape of the relationship between MTT and bulk Fm (Figure 4a). In principle, samples with variable MTTs can be collected from the same river system by analyzing a downstream profile of riverine POC over a length greater than the characteristic transport length
- 565 scale (Equation 4). However, spatial variations in OC cycling rates may complicate the interpretation of such analyses.

#### 3.3 The downstream profile of POC radiocarbon

For fixed values of POC cycling rates, the bulk Fm of riverine biospheric POC depends on the MTT (Figure 4a), which increases as the number of transport events increases (Figure 3b). Thus, our model

570 predicts that the bulk Fm of biospheric POC should decrease downstream. The exact shape of this decrease depends on the POC cycling rates as well as the relationships between channel length and sediment transit times set by the transport length scale  $(x_{tran})$ , the number of transport events  $(n_x)$ , and the cutoff time  $(T_{cut})$ .

For short channel lengths, all Fm-channel length relationships show Fm values near 1 (i.e., mod-575 ern POC) due to limited storage (Figure 4b). As channel length increases, each relationship tends

towards the steady-state Fm that is set by the POC consumption rate constants (Figure 4b). For each curve shown in Figure 4b, the relative increase in channel length required to reach the steady-state Fm is set by  $T_{cut}$  and  $x_{tran}$ . In other words, low Fm values require slow POC cycling rates, but

these values can only be expressed with sufficient sediment storage, which increases with increasing

580 channel length. This result illustrates that the downstream profile of the radiocarbon content of biospheric POC contains information regarding the interplay between sediment storage and OC cycling (Figure 4b).

#### 3.3.1 Benchmark with natural systems

The model results shown in Figures 4a illustrate that increasing the proportion of slow cycling POC results in a decrease in Fm, whereas decreasing the consumption rate constant can increase Fm. With this in mind, we compared our field data to POC cycling models with fixed POC consumption rate constants (fast and slow rates of 0.01 and  $2 \times 10^{-5}$  yr<sup>-1</sup>, respectively), but variable proportions of slow POC (10%, 33%, and 80%). These OC cycling models are contrasted with an "inert" model (Section 2.4), where POC is assumed to have the same age distribution as the sediments. To produce 590 a range of sediment MTTs, we consider simulations with  $T_{cut}$  values of 350 and 1000 years.

All field data within our compilation fall within the region defined by our model predictions after taking into account the uncertainties in our estimates of biospheric Fm from field measurements (Figure 5a). Rivers with small catchment areas tend to have high Fm values while rivers with larger catchments show a wider range of Fm (Figure 5a). This behavior is predicted by our model wherein

sufficient sediment storage is required to express differences in Fm values that result from variable POC cycling rates.

Our model predicts that variations in Fm for rivers with large catchment areas should depend on the rates of OC cycling, which we represent as the portion of slow POC. While it is difficult to independently quantify POC cycling rates at each of these sites, the observed meridional dependence

600 of POC cycling implies that latitude can be used as a proxy for POC cycling rates (Carvalhais et al., 2014). Consistent with this, the larger catchments with high Fm values are all from low latitudes while the larger catchments with low Fm values are from high latitudes (Figure 5a).

Our model predictions of the relationship between catchment area and Fm are sensitive to both the portion of slow POC as well as  $T_{cut}$  (Figure 5a). In an attempt to control for the dependence of  $T_{cut}$  on the relationships shown in Figure 5a, we compare Fm values with the relative transit time metric developed in Section 3.1.1 (Equation 16), which accounts for predicted differences in  $T_{cut}$ using variations in  $w^3/Q_s$ . Comparing the relative transit time of each river with the biospheric Fmyields a relationship consistent with our model predictions (Figure 5b). As the relative transit time increases, biospheric Fm decreases as a result of radioactive decay during storage (Figure 5b). As

610 expected, the exact relationship between relative transit time and biospheric Fm is variable between sites due to the effects of OC cycling (Figure 5b). By analyzing relative transit times (Figure 5b), we showed that the observed variations in the Fm of riverine biospheric POC are not solely due to variations in OC cycling, but also arise from variations in sediment storage.

## 4 Discussion

#### 615 4.1 Behavior of biogeochemical cycles

The storage of aged POC predicted by our model has implications for global biogeochemical cycles. Observations (Torn et al., 1997; Lawrence et al., 2015) and basic OC cycling models (e.g., Jenny et al., 1949) link the age of a soil or sediment deposit to its radiocarbon content and OC inventory. At the scale of river catchments, the amount of time available for OC accumulation and radioactive

decay is set by the patterns of channel migration with time. These patterns lead to the preferential erosion of young deposits (Figure 3a). Consequently, old deposits, which have accumulated more OC, are preferentially retained in floodplains (Figure 3a). Thus, the dynamics of lateral channel migration imply that river deposits are a more sophisticated reservoir of OC than would be assumed by the *null* hypothesis of a well-mixed system due to their age structure. Similarly, river dynamics
also influence the expression of these storage processes in geochemical tracers (e.g., *Fm*) by setting the relative proportions of variably-aged river deposits sampled by fluvial erosion (Figure 3).

Typically, the low radiocarbon content of riverine POC is assumed to result from the erosion of petrogenic OC from sedimentary rocks (Masiello and Druffel, 2001; Bouchez et al., 2010; Galy et al., 2008; Tao et al., 2015). Our results suggest that biospheric POC stored in river deposits can be

- 630 another source of riverine POC that is depleted in radiocarbon. While our model predicts the bulk Fm of this stored OC reservoir, this value is associated with the time-averaged behavior of the river system and represents the average of all POC eroded by channel migration. On short timescales, the Fm of biospheric POC sampled by fluvial erosion may deviate from the time-averaged value predicted by our model depending on the ages of fluvial deposits being eroded. Potentially, such
- 635 short term variations in the Fm of the biospheric end-member may account for the fact many of the rivers in our compilation do not conform to two-component mixing between petrogenic and biospheric POC (Section 2.5).

Many previous studies of the terrestrial OC cycle have focused on the transport of terrestrial OC to marine depo-centers, where it can be buried and stored over geologic timescales (Galy et al., 2015;

- 640 Hilton, 2016). While this is undoubtedly an important process, our calculations suggest that sediment storage times in river deposits may approach geologic timescales (e.g., up to 10<sup>6</sup> years; see also ?)(e.g., up to 10<sup>6</sup> years; see also Pizz and thus may play an important role in buffering changes in atmospheric CO<sub>2</sub> and O<sub>2</sub> concentrations. Moreover, the fluvial processes that dictate sediment storage timescales will also influence how terrestrial OC is transfered to marine basins. As a result, linkages between tectonic/climatic
- 645 forcings and OC burial fluxes should include the mechanics of the reservoirs associated with terrestrial sediment-routing systems.

Presently, our model only accounts for lateral movements of the river channel a meandering river with time. In natural river systems, vertical channel pattern (e.g., Eaton et al., 2010) and elevation changes driven by overbank deposition, subsidence, and aggradation may also influence the age

- 650 structure of riverine POC. Since most overbank deposition is focused near the active channel (?Marriott, 1992; Aalto et al., 2008)(Piz the spatio-temporal pattern of lateral channel migration likely approximates the storage durations associated with these deposits. Overbank deposition distal to the channel coupled with subsidence can lead to the burial of sediment deposits below the scour depth of the migrating channel. By selectively removing the oldest deposits, this additional sediment (and POC) sink can truncate the distribution
- of sediment ages. Assuming a constant floodplain elevation, the time to bury a deposit beneath the scour depth can be approximated as the ratio of the channel depth to the subsidence rate. As long as this burial timescale is long relative to the upper bound of the storage duration distribution ( $\beta$ ; Equation 14), then subsidence will play a subordinate role in setting the age distributions of riverine sediments and POC. This approach of comparing the timescales associated with lateral migration
- and sediment burial can be used to evaluate whether the model presented here is appropriate for a particular field system.

All together, the results of this study imply that the controls on the terrestrial OC cycle are not limited to the factors that affect rates of primary productivity and respiration, but also include the dynamics of terrestrial sedimentary systems (see also: Stallard, 1998; Galy et al., 2015; Hilton, 2016).

- 665 Sediment transport processes dictate the time and space scales over which OC cycling occurs, warranting their explicit consideration in models of global biogeochemical cycles. Since the terrestrial biosphere, in turn, influences the behavior of fluvial systems (Tal and Paola, 2007; Gibling and Davies, 2012), there is scope for feedbacks between biogeochemistry, sediment transport processes, and the architecture of fluvial processes to have changed over Earth's history (Algeo and Scheckler, 1008; Cibling and Davies, 2012).
- 670 1998; Gibling and Davies, 2012).

#### 4.2 Interpretation of time-varying environmental signals

In addition to affecting the behavior of biogeochemical cycles, our predicted age distributions also have implications for how we interpret environmental changes preserved in sedimentary archives. For example, compound-specific isotopic analyses of terrestrial biomarkers are widely employed as

- 675 proxies for environmental conditions (Eglinton and Eglinton, 2008). In systems where these compounds are delivered to a basin via an alluvial river system, they are likely to inherit some age structure as a result of sediment storage. This mixing of variably-aged biomarkers will act as a signal filter and modify the sedimentary expression of environmental changes preserved in the isotopic composition of these compounds (Douglas et al., 2014). While such "shredding" of environmental signals has
- 680 been extensively considered for sediments (?Ganti et al., 2014; ?)(Jerolmack and Paola, 2010; Ganti et al., 2014; Pizzuto et al., 2017) here, we extended this type of analysis to organic biomarkers.

To explore the implications of our model predictions for the time-series analysis of terrestrial biomarkers, we convolved each simulated In Figure 6, we show the effects filtering periodic signals of varying frequency with our sediment transit time distribution with a known periodic signal to

685 investigate distributions. Applied to biomarkers, these results show how an environmental signal

recorded in the upstream portion of a catchment will be modified as a result of transient storage during transport from source to sink. While the extent of amplitude damping and phase lag at different frequencies . We specifically looked at the sediment transit time distribution (Figure 3), and not the POC age distribution, to determine the maximum effect of sedimentary averaging on the

- 690 modulation of environmental signals. This might also be more appropriate for biomarker studies, which often target recalcitrant compounds that resist degradation (Eglinton and Eglinton, 2008). By treating the problem in this manner, we made the limiting assumption that biomarkers are not continually produced during sediment transit (c.f. Ponton et al., 2014). As such, the results can be viewed as reflecting how the expression of a signal recorded in biomarkers sourced from an upland
- 695 region is modulated by episodic downstream transport.-

Our results show that low frequency (i.e., forcing period >> mean transit time ) environmental signals are likely to be robustly recorded in the isotopic composition of terrestrial biomarkers (Figure 6). However, higher frequency variability shows complex phasing and significant amplitude damping is a generic outcome of signal filtering (Figure 6), our model framework links these effects to

- 700 physical properties of river systems (e.g., lateral migration rates, sediment fluxes, and channel length). For high frequency signals, the relatively long transit time of biomarkers produced in the upstream portion of a river system creates a large phase lag between the original environmental signal and its sedimentary record (Figure 6). In part, the large phase lags are due to our limiting assumption that biomarkers are not continually produced during sediment transit. Consequently, the time for
- 705 a signal generated upstream to advect downstream increases with the number of transport events. As an end-member case, we can consider a scenario where the Assuming these upland-sourced biomarkers are mixed with an *in situ* a more distal floodplain source that records environmental signals approximately in phase. In this case, the large phase lag between the two biomarker pools will cause destructive interference in the sedimentary expression of the environmental signal.
- As we considered only a single end-member case here, we acknowledge that broad Based on our simplified analysis, we expect that knowledge of both the sediment transit time and POC age distributions may aid in the interpretation of proxy systems affected by fluvial averaging. We acknowledge that the application of our model framework will likely require the explicit modeling of the production and consumption of organic biomarkers during fluvial transit (Galy et al., 2011; Ponton et al.,
- 715 2014) and site specific information about sediment storage timescales. However, by identifying some of the potential controls on sediment transit times distributions, our study may be useful in terms of site selection for paleoclimate studies. Nevertheless, our main point is that knowledge of both the sediment transit time and POC age distributionsmay aid in the interpretation of proxy systems affected by fluvial averaging. Conversely, if the magnitude of the expected environmental signal was
- 720 known independently, then its expression within a biomarker record could be used to infer properties of POC age distribution, as is done for water transit through catchment systems (e.g., McGuire and McDonnell, 2006; Kirchner, 2015). While this general approach was attempted by Douglas

et al. (2014) using time-series of terrestrial biomarkers in lake sediments, they assumed an arbitrary shape for the distribution of POC ages (a bimodal Gaussian distribution). Consequently, our mod-

eling approach can be used to improve such efforts by providing a framework for generating more

725

mechanistic POC age distributions.

### 5 Conclusions

Using simplified models that capture the physical processes associated with sediment storage for meandering rivers, we found that sediment transit times distributions have exhibit a power-law
behaviordecay, though geometric constraints temper or limit the distribution in our model simulations. Coupling our model for sediment transit time distributions to a simple model of OC cycling yields a full model for the radiocarbon content of riverine POC that can help interpret field observations. Thus we have to consider sediment storage We find that riverine sediment storage is a major aspect of biogeochemical cycling that introduces a time continuum that runs from annual to potentially
million-year timescales. A basic inference from the results of this study is that biomarkers will exhibit a delay and/or mixed signal that convolves both the forcing and the storage. Though complicating the interpretation of sedimentary records, which will incorporate a mixture of variably-aged POC, this river-floodplain exchange behavior presents an opportunity to understand the timescales over which CO<sub>2</sub> is stored as organic matter in surface deposits, which has important implications for

740 understanding global environmental change.

#### 6 Data Availability

The raw data utilized in the study were compiled from previously published works and are available in the cited manuscripts. In Table 2, we provide results derived from the data compilation. A working example of our sediment and carbon storage model is included as supplementary MATLAB script.

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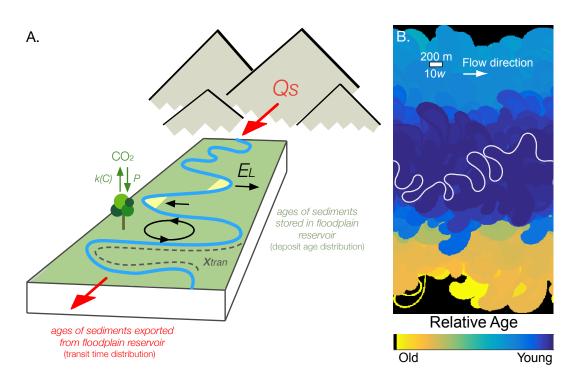
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## 1050 List of Figures

1 Model schematic schematics highlighting key geometric relationships. (A) The sediment flux  $(Q_s)$  from an upstream area is routed though an alluvial valley. As a result of lateral channel migration  $(E_L)$ , sediment deposits are created and eroded leading to transient sediment storage. During sediment storage, the fixation of atmospheric 1055  $CO_2$  by biota leads to the production (P) of particulate organic carbon (C), which is degraded back to  $CO_2$  at a rate (k) proportional to its concentration in sediments. At any timeAs sediments (and POC) move downstream, they can be re-deposited and stored. Here, we define a characteristic length scale  $(x_{tran})$  that reflects the distance sediments travel before being re-deposited. The ages of sediments and or-1060 ganic carbon can be described by two distributions. The storage duration transit time distribution describes the ages of material being actively removed from the system by erosion and thus have reached their maximum age. The deposit age distribution describes the ages of material that remains stored in river deposits and will continue to age until subsequent erosion. (B) A map view of example model output where the white line shows the final position of the river channel, which flows from left 1065 to right across the model domain. Sediment ages range from relatively old (yellow and orange) at the lower edge of the domain to relatively young (blue) nearer the channel. Areas of the model domain not visited by the channel are shaded black. . . 33 2 Sediment transport parameters in the model and field data. (aA) The results of the 1070 numerical meandering model simulations that show that the meander bend growth timescale  $(T_{cut})$  is correlated with the time required for the channel to migrate one channel width at the maximum lateral migration rate  $(E_{L,max})$ . (bB) Field evidence for a correlation between the mean time required to migrate one channel width  $(w/E_L)$  and the sediment supply timescale  $(w^3/Q_s)$ . (eC) Jitter plot of calculated 1075  $x_{tran}$  values (Equation 4) for the field data shown in (bB). Together, these panels show that  $T_{cut}$  scales with the lateral migration rate, which, in field data, is correlated with suspended sediment fluxes (Constantine et al., 2014). The correlation between lateral migration rates and sediment fluxes results in minimal variation in the transport length scale (c) and provides a proxy for  $T_{cut}$  (i.e.,  $w^3/Q_s$ ) for rivers 1080 3 Dimensionless age distributions. (aA) The survival function (complementary cumulative distribution function) of sediment storage durations (red points) derived from the meandering model. The sediment storage durations are normalized by a cutoff time  $(T_{cut})$  of 350 years based on explicit tracking of meander bend growth (Schwenk et al., 2015) following the approach of Schwenk et al. (2015). The model 1085 results were fit to a best fitting tempered Pareto distribution and the best-fitting model is exponential distributions are shown as a solid and dashed red lines, respectively. Also shown is a tempered Pareto fit to the distribution of deposit ages at the end of the model simulation (black line). (b-eB-C) The change in the shape of the sediment transit time distribution (normalized by  $T_{cut}$ ; Equation 3) with in-1090 creasing number of transport events. (bB) The mean, median, and 90% confidence interval of the sediment transit time distributions for 1 to 20 transport events. (eC) The probability density function of sediment transit time distributions for 1 and 20 transport events. Consistent with the central limit theorem, the shape of the sediment 1095 35

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**Figure 1.** Model schematic schematics highlighting key geometric relationships. (A) The sediment flux  $(Q_s)$  from an upstream area is routed though an alluvial valley. As a result of lateral channel migration  $(E_L)$ , sediment deposits are created and eroded leading to transient sediment storage. During sediment storage, the fixation of atmospheric CO<sub>2</sub> by biota leads to the production (P) of particulate organic carbon (C), which is degraded back to CO<sub>2</sub> at a rate (k) proportional to its concentration in sediments. At any time As sediments (and POC) move downstream, they can be re-deposited and stored. Here, we define a characteristic length scale  $(x_{tran})$  that reflects the distance sediments travel before being re-deposited. The ages of sediments and organic carbon can be described by two distributions. The storage duration transit time distribution describes the ages of material being actively removed from the system by erosion and thus have reached their maximum age. The deposit age distribution describes the ages of material that remains stored in river deposits and will continue to age until subsequent erosion. (B) A map view of example model output where the white line shows the final position of the river channel, which flows from left to right across the model domain. Sediment ages range from relatively old (yellow and orange) at the lower edge of the domain to relatively young (blue) nearer the channel. Areas of the model domain not visited by the channel are shaded black.

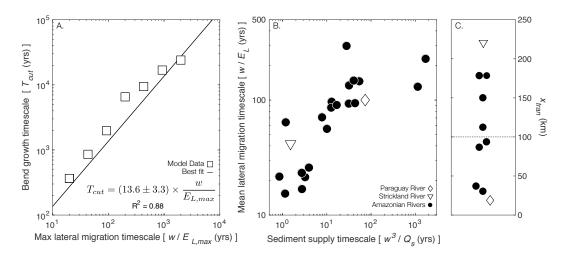
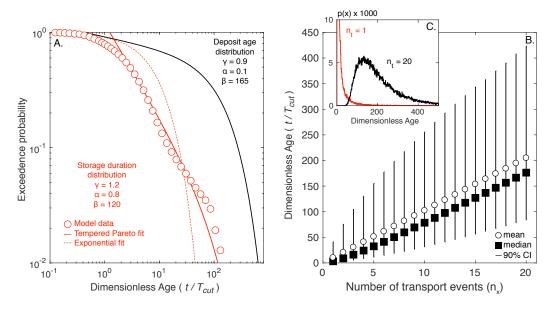
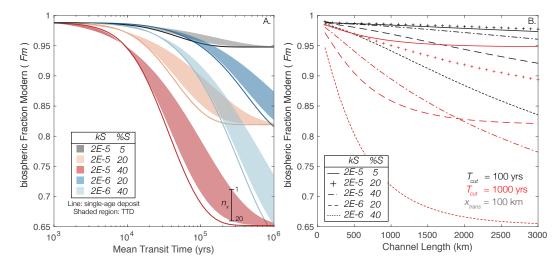


Figure 2. Sediment transport parameters in the model and field data. (aA) The results of the numerical meandering model simulations that show that the meander bend growth timescale  $(T_{cut})$  is correlated with the time required for the channel to migrate one channel width at the maximum lateral migration rate  $(E_{L,max})$ . (bB) Field evidence for a correlation between the mean time required to migrate one channel width  $(w/E_L)$  and the sediment supply timescale  $(w^3/Q_s)$ . (eC) Jitter plot of calculated  $x_{tran}$  values (Equation 4) for the field data shown in (bB). Together, these panels show that  $\frac{T_{cut}}{T_{cut}}$  scales with the lateral migration rate, which, in field data, is correlated with suspended sediment fluxes (Constantine et al., 2014). The correlation between lateral migration rates and sediment fluxes results in minimal variation in the transport length scale (e) and provides a proxy for  $T_{cut}$  (i.e.,  $w^3/Q_s$ ) for rivers where lateral migration rates are unknown.



**Figure 3.** Dimensionless age distributions. (aA) The survival function (complementary cumulative distribution function) of sediment storage durations (red points) derived from the meandering model. The sediment storage durations are normalized by a cutoff time ( $T_{cut}$ ) of 350 years based on explicit tracking of meander bend growth (Schwenk et al., 2015) following the approach of Schwenk et al. (2015). The model results were fit to a best fitting tempered Pareto distribution and the best-fitting model is exponential distributions are shown as a solid and dashed red linelines, respectively. Also shown is a tempered Pareto fit to the distribution of deposit ages at the end of the model simulation (black line). (b-eB-C) The change in the shape of the sediment transit time distributions for 1 to 20 transport events. (eC) The probability density function of sediment transit time distributions for 1 and 20 transport events. Consistent with the central limit theorem, the shape of the sediment transit distribution varies with the number of transport events.



**Figure 4.** Model predictions of the radiocarbon content of POC. (aA) Predicted relationships between sediment mean transit times (MTTs) and the Fm of biospheric POC for POC cycling models with variable proportions of slow POC (%S) and variable slow POC consumption rate constants (kS;  $yr^{-1}$ ). Note that %S is equal to the proportion of slow POC at steady-state. The Fm values shown as shaded areas were calculated using sediment transit time distributions (TTDs) with shapes set by between 1 to 20 transport events (Figure 3) and MTTs between  $10^3$  and  $10^6$  years. The model results show that plausible sediment TTDs yield significantly different relationships between the MTT and Fm relative to predictions for systems with a single transit time (solid bold lines). While TTD shape is an important factor reflected in the vertical range of each shaded area, the model results suggest that large differences of OC cycling (different colors; see figure legend) are distinguishable in the relationships between the MTT and Fm. (bB) Predicted downstream profiles of Fm for a fixed  $x_{tran}$  (100 km), but variable  $T_{cut}$  (100 and 1000 years) and POC cycling parameters (different line styles; see figure legend). The model results suggest that observed downstream profiles are sensitive to both sediment storage timescales, which are controlled by  $T_{cut}$ , as well as OC cycling parameters.

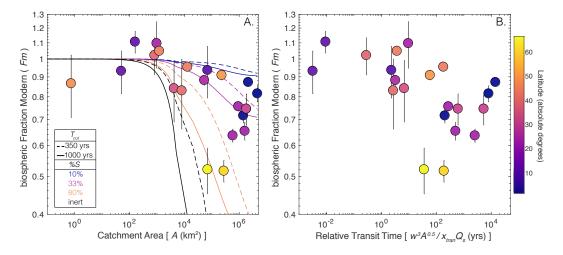


Figure 5. Field data compilation and comparison with model predictions. Field data are color coded based on their latitude as a proxy for POC cycling rates. (aA) Model curves and field measurements of the relationship between catchment area and bulk biospheric Fm. Model curves are drawn for fixed POC consumption rate constants, but variable steady-state proportions of "slow" cycling POC and  $T_{cut}$  values (see legend). (bB) Measured biospheric Fm values compared to our relative transit time metric (Equation 16), which is based on Hack's Law (Hack, 1957) and the scaling relationships shown in Figures 2 and 3. The field data are consistent with the model predictions of aged POC in rivers with larger catchment areas and slower migration rates (inferred from variations in  $w^3/Q_s$ ) and thus longer sediment storage times. We note that in our compilation of rivers with measurements of biospheric Fm, catchment area and  $w^3/Q_s$  are correlated.

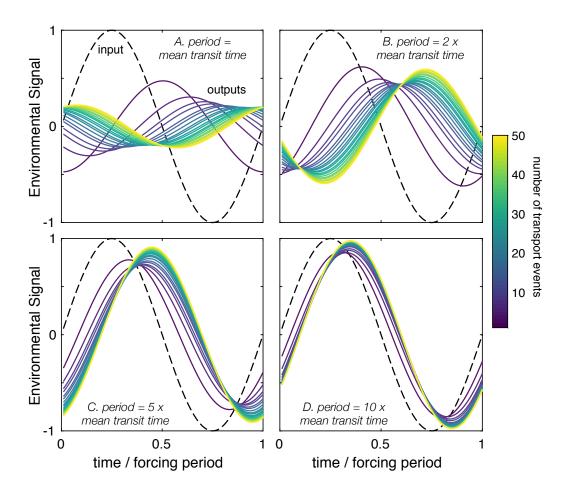


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Physical characteristics and inferred biospheric Fm values for a subset of rivers	
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Table 1. List of all rivers and their associated citations included in the radiocarbon compilation

River	Citation		
Alsea	Hatten et al. (2012)		
Amazon	Bouchez et al. (2010, 2014)		
Arctic Red	Hilton et al. (2015)		
Atchafalaya	Gordon and Goni (2003); Rosenheim et al. (2013)		
Avon	Adams et al. (2015)		
Beni	Bouchez et al. (2010)		
Brahmaputra	Galy et al. (2007)		
Calder River	Adams et al. (2015)		
Changjiang (Yangtze)	Wang et al. (2012); Li et al. (2015)		
Chontabamba	Townsend-Small et al. (2007)		
Chorobamba	Townsend-Small et al. (2007)		
Conwy	Adams et al. (2015)		
Dee	Adams et al. (2015)		
Eel	Leithold et al. (2006)		
Erlenbach	Smith et al. (2013); Turowski et al. (2016)		
Esperanza	Townsend-Small et al. (2007)		
Fly	Alin et al. (2008)		
Fraser	Voss (2014)		
Ganges	Galy et al. (2007)		
Garin	Adams et al. (2015)		
Hodder	Adams et al. (2015)		
Huancabamba	Townsend-Small et al. (2007)		
Kosi	Galy et al. (2007)		
Kosñipata	Clark et al. (2013)		
Lanyan Hsi	Kao and Liu (1996)		
Liard	Hilton et al. (2015)		
Liwu	Hilton et al. (2010)		
Llamaquiz	Townsend-Small et al. (2007)		
Madiera	Bouchez et al. (2010, 2014)		
Meghna	Galy et al. (2007)		
Mekong	Martin et al. (2013)		
U			
Mississippi	Rosenheim et al. (2013) Galv et al. (2007)		
Narayani Navarro	Galy et al. (2007) Laithold et al. (2006)		
	Leithold et al. (2006)		
Noyo	Leithold et al. (2006) Hilton et al. (2015)		
Peel			
Pozuzo	Townsend-Small et al. (2007)		
Rhône	Cathalot et al. (2013)		
Ribble	Adams et al. (2015) Marialla and Derffel (2001): Kannada et al. (2004)		
Santa Clara	Masiello and Druffel (2001); Komada et al. (2004)		
Siuslaw	Leithold et al. (2006)		
Solimoes	Bouchez et al. (2010, 2014)		
Strickland	Alin et al. (2008)		
Tokachi	Nagao et al. (2005)		
Umpqua	Goñi et al. (2013)		
Waiapu	Leithold et al. (2006)		
Waipaoa	Leithold et al. (2006)		
Yellow (Huanghe)	Wang et al. (2012); Tao et al. (2015); Hu et al. (2015		
Zengjiang	Gao et al. (2007)		

**Table 2.** Physical characteristics and inferred biospheric Fm values for a subset of rivers in the radiocarbon compilation. The values of sediment fluxes are taken from the original studies and are based on either suspended sediment gauging or cosmogenic nuclide concentrations. We report the uncertainty on our estimate of biospheric Fm using equation 13 as one standard deviation ( $\sigma$ ).

River	Sediment Flux (Mt/yr)	Channel Width (m)	Catchment Area (km <sup>2</sup> )	Biospheric Fm <sub>bio</sub>	$\sigma$	Additional Data S
Alsea	0.07	65	1220	1.05	0.01	USGS
Amazon	785	4767	4618750	0.82	0.04	HYBAM
Beni	212	410	69980	0.94	0.14	Wittmann et al. (2
Brahmaputra	540	4052	583000	0.64	0.03	
Changjiang	472	2000	1810000	0.75	0.07	Google Earth
Eel	14.00	250	8063	0.83	0.17	USGS
Erlenbach	0.001	5	0.74	0.87	0.16	Nitsche et al. (20
Fraser	20	450	230000	0.91	0.03	Google Earth
Ganges	660	1910	935000	0.76	0.02	
Kosi	135	412	53610	0.88	0.03	Google Earth
Kosñipata A	0.06	10	50	0.93	0.12	
Kosñipata B	0.18	17	160	1.11	0.07	
Lanyan Hsi	2.90	320	980	1.10	0.15	Google Earth
Liard	46	850	275000	0.52	0.03	Google Earth
Madiera	433	1401	1420000	0.72	0.03	Google Earth
Meghna	0.24	2600	1600000	0.65	0.03	Google Earth
Navarro	0.56	59	816	1.02	0.11	USGS
Peel	21	470	70600	0.52	0.07	Google Earth
Rhône	31	460	99000	1.41	0.15	Google Earth
Santa Clara	6.84	298	4200	0.84	0.15	USGS
Solimoes	569	5893	2147740	0.87	0.02	HYBAM
Umpqua	1.4	435	13000	0.95	0.02	USGS