Aquatic carbon cycling in the conterminous United States and implications for terrestrial carbon accounting

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Edited by William H. Schlesinger, Cary Institute of Ecosystem Studies, Millbrook, NY, and approved November 12, 2015 (received for review June 29, 2015)

Inland water ecosystems dynamically process, transport, and sequester carbon. However, the transport of carbon through aquatic environments has not been quantitatively integrated in the context of terrestrial ecosystems. Here, we present the first integrated assessment, to our knowledge, of freshwater carbon fluxes for the conterminous United States, where 106 (range: 71–149) teragrams of carbon per year ($TgC{\cdot}y^{-1}$) is exported downstream or emitted to the atmosphere and sedimentation stores 21 (range: 9–65) $TgC{\cdot}y^{-1}$ in lakes and reservoirs. We show that there is significant regional variation in aquatic carbon flux, but verify that emission across stream and river surfaces represents the dominant flux at 69 (range: 36–110) $TgC{\cdot}y^{-1}$ or 65% of the total aquatic carbon flux for the conterminous United States. Comparing our results with the output of a suite of terrestrial biosphere models (TBMs), we suggest that within the current modeling framework, calculations of net ecosystem production (NEP) defined as terrestrial only may be overestimated by as much as 27%. However, the internal production and mineralization of carbon in freshwaters remain to be quantified and would reduce the effect of including aquatic carbon fluxes within calculations of terrestrial NEP. Reconciliation of carbon mass–flux interactions between terrestrial and aquatic carbon sources and sinks will require significant additional research and modeling capacity.

carbon | aquatic ecosystems | terrestrial ecosystems | carbon flux | inland waters

Management of terrestrial carbon stocks and fluxes is considered a sustainable approach to reducing the anthropogenic contribution to increasing atmospheric CO_2 (1). Therefore, accurate accounting of carbon storage and flux is essential to understand the role that natural ecosystems can play in regional, national, and global carbon cycles. However, current modeling approaches for estimating net ecosystem production [NEP = GPP − R, where GPP is gross primary production and R is autotrophic and heterotrophic respiration (2, 3)] assume that carbon is converted to terrestrial storage terms only without adequate accounting of aquatic carbon processes. These aquatic carbon processes include downstream carbon transport, lake and reservoir sedimentation, and CO_2 emission. Recent studies (4–6) indicate that downstream export of carbon through rivers, carbon burial in lakes and reservoirs, and carbon emissions across water surfaces are large enough to alter the calculation of terrestrial NEP significantly. Therefore, when the processing and removal of carbon through inland waters is properly accounted for, the calculated capacity of soils and biomass to store carbon is reduced (7, 8).

Most studies that have made concurrent measurements of terrestrial and aquatic fluxes of carbon were conducted in small watersheds and have used different metrics to estimate total carbon accumulation across terrestrial landscapes. For example, recent efforts suggest that upward of 10% of the net ecosystem exchange (NEE) in Sweden is represented by the evasion of CO_2 from stream and river surfaces (9). On a larger scale, Striegl et al. (10) suggest that about 6% of net primary production (NPP) is exported downstream in the Yukon basin. When combined with river CO_2 emission (11, 12), the total aquatic carbon flux amounts to 12% of NPP for the basin. Across smaller watersheds, the magnitude of the freshwater carbon flux can reach 34% of the estimated NEP, excluding carbon burial via sedimentation (13). These studies are beneficial in identifying the diversity of carbon sources and potential influences of different carbon cycle processes, and they also document removal via lateral carbon export and CO_2 emissions.

The total mass fluxes of carbon moving out of aquatic systems are poorly constrained at the global scale, yet recent estimates suggest these fluxes may exceed 3 petagrams of carbon (PgC) per year (14, 15). Data are lacking at this scale to apportion these fluxes across the landscape. Emerging research highlights the difficulty in identifying the major sources of carbon to freshwater ecosystems, which is critical to link the terrestrial and aquatic carbon cycles properly. Efforts in the Brazilian Amazon River basin produce differing results depending upon geographic location and prevailing hydrologic conditions. Research suggests upstream terrestrial carbon previously degraded in soil and stream environments can fuel heterotrophy in the lower river reaches (16), whereas in another study, respiration and organic carbon (OC) from fringing wetlands may provide significant CO_2 to match the supersaturated chemistry of the larger central Amazon (17). These two sources are not mutually exclusive. In the conterminous United States, input of supersaturated ground water is more important in small streams, whereas internal production of CO_2 is higher in larger river systems (18).

In lake and reservoir ecosystems, CO_2 supersaturation is common, but the processes maintaining elevated CO_2 concentrations are diverse and regionally variable (19–21). Respiration

Significance

Inland waters provide habitat for aquatic organisms; are sources of human drinking water; and integrate, transport, and process carbon across continents. Estimates of the accumulation of carbon in terrestrial environments suggest that agricultural and forest ecosystems have annual net gains in carbon storage. These ecosystems are considered sinks of atmospheric carbon dioxide. None of these estimates have considered the loss of carbon to and also through aquatic environments at the national or continental scale. We show that aquatic ecosystems in the conterminous United States export over 100 teragrams of carbon (TgC) per year, highlighting the need to attribute the sources of aquatic carbon more accurately, and assert that inland waters play an important role in carbon accounting.

Author contributions: D.B., S.S., E.S., C.P.M., D.W.C., and R.G.S. designed research; D.B., S.S., E.S., C.P.M., and D.W.C. performed research; D.B., S.S., E.S., C.P.M., D.W.C., and R.G.S. analyzed data; and D.B., S.S., E.S., C.P.M., D.W.C., and R.G.S. wrote the paper.

The authors declare no conflict of interest.

This article is a PNAS Direct Submission.

Freely available online through the PNAS open access option.

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This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1512651112/-/DCSupplemental.

of allochthonous OC results in net heterotrophy in many lake systems (22). Inorganic carbon inputs can also contribute substantially to CO_2 emissions from lakes (19, 23). Furthermore, freshwater systems have strong seasonal patterns (22). Changes in precipitation alter stream and river flow and carbon concentrations (24). Lakes can vary between net heterotrophy and net autotrophy with changes in temperature and across spatial gradients (19). Man-made reservoirs can function like natural lake systems; however, they also sequester large amounts of carbon through sedimentation (25). Much of the OC delivered to sediments is potentially remineralized to CH_4 and CO_2 and returned to the atmosphere over millennia (26–28).

Here, we present the first combined estimates of the major removal fluxes, to our knowledge, including burial in lakes and wetlands, downstream transport to coastal areas, and the CO_2 emission to the atmosphere for the conterminous United States. In this effort, we aggregate vertical gas evasion, lake and reservoir sedimentation, and the lateral fluxes of carbon by modified versions of major two-digit US Geological Survey (USGS) hydrologic regions (HUCs) and compare estimates across each of these regions (Fig. S1). In its simplest form, the calculated total US aquatic carbon flux is represented by Eq. **1** (expanded methods are provided in *SI Materials and Methods*):

$$\begin{aligned} \textit{Total US Flux}\ \left(\mathrm{TgC}\cdot\mathrm{y}^{-1}\right) = \sum_{HUC} & \textit{Stream Emission} + \textit{Lateral Flux} \\ & + \textit{Lake \& Reservoir Emission} \\ & - \textit{Lake \& Reservoir Burial}, \end{aligned} \quad [1]$$

where *Stream Emission* represents the vertical evasion of CO_2 from stream and river surfaces in teragrams of carbon (TgC) per year, *Lateral Flux* represents the total dissolved inorganic plus OC fluxes draining from the landscape to coastal systems in TgC per year, *Lake & Reservoir Emission* represents the vertical evasion of CO_2 across lake and reservoir surfaces in TgC per year, and *Lake & Reservoir Burial* represents the total organic carbon (TOC) burial in lakes plus reservoirs in TgC per year. This total flux was calculated for only the conterminous US land area, because adjustments were made to reduce vertical emission and sedimentation for those areas of each contiguous basin in Fig. S1 that originate outside of the United States. Because fluxes were calculated using consistent national methodologies, we are able to make regional comparisons in aquatic flux magnitudes across systems as well as explore the importance of terrestrial drivers on aquatic flux variability.

North America has been identified as a potentially large carbon sink ranging from 0.1 to 2 $PgC\cdot y^{-1}$ (29). Although much of this sink may be attributed to increasing rates of forest regrowth (30), there remains significant variability among the current suite of terrestrial biosphere models (TBMs) to identify and account for a terrestrial sink in any one component of the biosphere (31). Here, we use aggregated results of the Multiscale Synthesis and Terrestrial Model Intercomparison Project (MsTMIP) part of the North American Carbon Program to capture both the variability that exists across models and the uncertainty associated with the calculation of terrestrial carbon stocks and flows across large spatial scales. The transport and processing of carbon in aquatic systems is not estimated in any of the predictive models. In this context, the inputs of terrestrial carbon into aquatic environments in both inorganic and organic forms, as particles or dissolved, represent either a loss of either terrestrial carbon uptake or relocation of respiration as CO_2.

Results and Discussion

Total Carbon Fluxes. The total flux of freshwater carbon from the conterminous United States is 106 (range: 71.4–148.9) $TgC\cdot y^{-1}$ (Table 1) as calculated by Eq. **1**. Aggregated to the conterminous United States, the atmospheric flux of CO_2 from streams and rivers represents 65.4% of the total positive flux of inland water carbon at 69.3 (range: 36–109.6) $TgC\cdot y^{-1}$ (Table 1). This estimate for stream efflux is 27.7% lower than a previous estimate of 97 $TgC\cdot y^{-1}$ (32). The difference between the water surface emissions presented here and in the study by Butman and Raymond (32) is predominantly due to modifications in the modeling related to estimating the total surface areas of streams and rivers and the gas transfer velocity coefficient across the air–water interface (33). In particular, the gas transfer velocity was not allowed to exceed 30 $m\cdot d^{-1}$, derived from new hydraulic geometry coefficients (Table S1), and this restriction affected the magnitude of fluxes in small-order streams in steep topography (*SI Materials and Methods*).

The total lateral flux, representing only the export of carbon to coastal systems, is 41.5 (39.4–43.5) $TgC\cdot y^{-1}$. This estimate is similar to past research, which suggests a lateral export of 41–49 $TgC\cdot y^{-1}$ developed from USGS gaging station discharge and water quality data (34). The dominant component of the lateral flux is dissolved inorganic carbon (DIC) at ∼70% of the total flux, whereas the remaining 30% of the total flux is dissolved organic carbon (DOC), which aligns with previous large-scale flux estimates (14, 34) (Table S2). In this effort, the methods to calculate both the CO_2 emission and the lateral export of DIC use the same underlying dataset on alkalinity, temperature, and pH to calculate inorganic carbon in freshwaters. CO_2 uptake in terrestrial soils by mineral weathering contributes to the total DIC in inland waters. However, degassing of inorganic carbon to the atmosphere is determined by the concentrations of H_2CO_3 and dissolved CO_2 in surface waters.

Total CO_2 efflux from lake surfaces is 16 (range: 14.3–18.7) $TgC\cdot y^{-1}$, whereas TOC burial is 20.6 (range: 9.0–65.1) $TgC\cdot y^{-1}$. Aggregated at the national scale, lakes and reservoirs accumulate carbon through sedimentation in equal proportion to the release of carbon across the air–water interface. However, the level of uncertainty associated with the burial of OC and emission of CO_2 from lakes and reservoirs highlights a gap in our understanding of the processes involved, and reinforces that these numbers are best estimates from existing data (35).

Identifying appropriate terrestrial modeling products to evaluate against the cycling of aquatic carbon remains difficult. Here, we have included a subset of the MsTMIP model intercomparison efforts to look at the potential relationships between NEE in grams of carbon (gC) per square meter per year, NEP in gC per square meter per year, NPP in gC per square meter per year, total live biomass in kilograms of carbon per square meter, total respiration in gC per square meter per year, total soil carbon in kilograms of carbon per square meter, and the fluxes of carbon in inland waters. Here, we have averaged for the period 1990–2010 only the results of the Biome-Biogeochemical Model (BGC) (36), the Canadian Land Surface Scheme and Canadian Terrestrial Ecosystem Model ($CLASS\text{-}CTEM^{N+}$) (37), the Community Land Model (CLM4) (38), the CLM4-Variable Infiltration Capacity (VIC) (39), the Dynamic Land Ecosystem Model (DLEM) (40), and the Integrated Science Assessment Model (ISAM) and TRIPLEX-GHG (41), given the data availability through Oak Ridge National Laboratory (42, 43). These data only include the scenarios that allowed for land use change to occur and for nitrogen deposition. This scenario was chosen to represent the model ensemble averages for the best estimate of carbon stocks and flows under naturally varying conditions.

Average annual NEP for the conterminous United States was 70.0 (SD = 84.0) $gC\cdot m^{-2}\cdot y^{-1}$. NEP varies across HUCs from a low of 21.2 (SD = 1.0) $gC\cdot m^{-2}\cdot y^{-1}$ in HUC 15, representing the desert southwest, to a high of 141.0 (SD = 12.6) $gC\cdot m^{-2}\cdot y^{-1}$ in HUC 03, dominated by the productive and dense forests of the southeastern United States (Fig. 1*C*). Additional results of the aggregation of the TBM outputs are presented in Fig. S2.

Regional Variability of Aquatic Carbon Fluxes. There is significant variability across each of the 19 HUCs for all of the inland water carbon fluxes. Total carbon fluxes ranged from 0.9 (range: 0.5–1.6) $TgC\cdot y^{-1}$ in HUC 15, the southern portion of the arid Colorado River Basin, to 11.8 (range: 6.9–17.7) $TgC\cdot y^{-1}$ in the eastern portion

Table 1. Total carbon fluxes by two-digit HUCs across the conterminous United States using Eq. 1

Two-digit HUC	Stream efflux, TgC·y^{-1}	Total lateral flux, TgC·y^{-1}	Lake efflux, TgC·y^{-1}	Lake burial, TgC·y^{-1}	Total aquatic carbon, TgC·y^{-1}	Carbon yield, gC·m^2·y^{-1} (Eq. **1**)	Carbon yield gC·m^2·y^{-1} (Eq. **2**)
01	1.8 (0.9–2.9)	1.1 (1–1.2)	0.7 (0.5–0.9)	0.5 (0.3–0.9)	3.1 (2–4.4)	18.1 (11.8–25.9)	24 (16.5–33)
02	3.6 (1.8–5.7)	1.9 (1.8–2)	0.6 (0.3–1.1)	0.4 (0.2–1)	5.6 (3.6–8.3)	20.3 (13–29.9)	23.2 (15.1–33.5)
03	5.5 (2.8–8.8)	3.7 (3.4–4)	2.5 (1.6–3.7)	3.1 (1.2–11.7)	8.6 (4.8–13.3)	12.3 (6.9–19.1)	21.2 (15.5–28.2)
04	1.5 (0.8–2.3)	6.6 (6.5–6.6)	1.1 (0.8–1.5)	0.6 (0.3–1.5)	8.5 (7.6–9.7)	28 (25–31.8)	32 (28.2–36.4)
05	5 (2.5–8)	5.7 (5.6–5.7)	0.4 (0.3–0.6)	0.5 (0.3–0.9)	10.6 (8–13.7)	25.1 (19–32.5)	27.5 (20.9–35.3)
06	2.6 (1.3–4.2)	0.8 (0.7–0.7)	0.2 (0–0.6)	0.4 (0.2–0.6)	3.2 (1.7–5)	30.2 (16–47.2)	37.8 (21.7–56.6)
07	2.3 (1.2–3.7)	6.3 (6.2–6.5)	0.7 (0.4–1.1)	0.9 (0.5–2.2)	8.4 (7–10.3)	17.2 (14.2–20.9)	20.8 (17.5–25)
08	3.1 (1.6–5.1)	1.9 (1.9–1.9)	2.3 (1.5–3.4)	5.4 (1.3–25.4)	1.9 (−0.1 to 4.7)	7.2 (−0.4 to 17.9)	48.3 (38.4–61.3)
09	0.3 (0.1–0.5)	0.4 (0.4–0.5)	1.1 (0.6–2.1)	0.4 (0.1–1)	1.5 (1–2.4)	9.3 (6.4–15.3)	14.4 (7.6–24.2)
10	5.3 (2.8–8.5)	3.8 (3.8–3.9)	1.2 (1–1.5)	1.8 (1–3.6)	8.5 (6–11.9)	6.4 (4.5–9)	9.1 (7–12)
11	4 (2.1–6.4)	2.5 (2.4–2.5)	1.8 (1.4–2.3)	1.2 (0.7–2.9)	7.1 (4.9–9.8)	11.1 (7.6–15.3)	14.8 (10.7–19.6)
12	1.5 (0.8–2.4)	1.4 (1.1–1.7)	2 (1.3–3.3)	1.9 (0.8–7.1)	3 (1.5–5.3)	6.5 (3.2–11.4)	14.7 (10.6–20.5)
13	1.4 (0.7–2.2)	0 (0–0)	0.1 (0–0.3)	0.2 (0.1–0.3)	1.3 (0.5–2.3)	3.8 (1.4–6.5)	4.9 (2.5–7.6)
14	4 (2–6.5)	0.7 (0.7–0.7)	0.1 (0–0.2)	0.3 (0.2–0.5)	4.5 (2.4–7.1)	15.3 (8.2–24.2)	17.3 (10.2–26.2)
15	1.1 (0.6–1.8)	0.1 (0–0)	0.1 (0.1–0.2)	0.3 (0.2–0.5)	0.9 (0.5–1.6)	2.5 (1.4–4.3)	4.2 (2.4–6.5)
16	2.6 (1.3–4.3)	0.8 (0.8–0.8)	0.5 (0.2–0.8)	0.6 (0.3–1.3)	3.3 (1.8–5.2)	8.9 (4.9–14.2)	12.2 (7.6–18)
17 Coast	6.3 (3.4–9.6)	1 (0.9–1)	0.02 (0–0.04)	0.2 (0.1–0.4)	7.1 (4.1–10.4)	41.6 (24-60.9)	44 (26.4–63.3)
17 Dry	10.5 (5.7–16.2)	2.2 (2–2.4)	0.1 (0–0.3)	1 (0.7–1.5)	11.8 (6.9–17.7)	20.8 (12.1–31.1)	24.3 (14.9–35.3)
18 Dry	6.8 (3.6–10.5)	1.6 (1.2–2)	0.4 (0.1–0.9)	0.9 (0.5–1.8)	8 (4.1–12.4)	20.1 (10.4–31.3)	24.7 (14.4–36.4)
Total	69.3 (36–109.6)	41.5 (39.4–43.5)*	16 (14.3–18.7)	20.6 (9–65.1)	106.2 (71.4–148.9)	13.5 (9.1–19)	18.8 (13.8–24.8)

Values in parentheses represent error terms; fifth and 95th percentiles for both lake and stream efflux; SE associated with DIC, DOC, and total lateral fluxes; and the SD for the burial of OC. Lateral fluxes represent the net fluxes at the most downstream point within each contiguous HUC. Total aquatic carbon flux errors represent the ± sum of all presented error terms. Carbon yield is expressed on a per unit area of the two-digit HUC.
*Total lateral flux represents only those HUCS that drain off of the land mass into coastal oceans and the Great Lakes, and exclude HUC 16, the Great Basin.

of HUC 17 inclusive of the Columbia River (Fig. 1*A* and Table 1). Previous work has suggested stream and river efflux is the dominant flux of carbon from freshwater ecosystems at the national scale (14, 32), but that finding is not uniform across all regions presented here (Table 1). HUCs 04, 05, 07, 08, 09, and 12 were dominated by either the lateral export of carbon (4, 5, 7) or the efflux of carbon from lake surfaces (9, 12). Estimates for the burial of carbon in lake sediments for HUC 08 are 5.4 TgC·y^{-1}, and represent the dominant removal and storage term for aquatic carbon.

The gaseous evasion of CO_2 from streams and rivers ranged from 0.3 (range: 0.1–0.5) TgC·y^{-1} in HUC 09 to 10.5 (range: 5.7–16.2) TgC·y^{-1} in HUC 17 Dry (Table 1). Stream and river efflux dominated those regions where there are high slopes (the western United States inclusive), and where there existed a combination of moderate alkalinity, lower pH, and high estimated water velocity. Water velocity is an important variable for the modeling of gas transfer coefficients (*SI Materials and Methods*), and is a function of both high slopes (HUCs 13, 14, 15, 16, and 17 Dry) or high precipitation (HUCs 02, 03, and 17 Coast).

Lateral fluxes of inorganic carbon and OC varied significantly across regions as well. Total DIC fluxes were largest from HUC 04 [4.9 (range: 4.9–4.9) TgC·y^{-1}], HUC 05 [4.4 (range: 4.3–4.4) TgC·y^{-1}], and HUC 07 [5.1 (range: 5.0–5.2) TgC·y^{-1}] (Table S2). This large inorganic carbon flux has been attributed to agricultural practices in the upper midwestern United States contributing bicarbonate, as well as weathering interactions within soils derived from underlying carbonate bedrock (34, 44). In our flux estimates, DIC was calculated from alkalinity and pH, and it was dominated by HCO_3^- and CO_3^{-2}. These ions are formed through weathering reactions in the soil, with as much as 36% originating from geological sources through carbonate weathering (45). These weathering reactions do not represent the rapid transit of atmospheric CO_2 into inland waters and require further investigation to account correctly for the temporal imbalance involved with the use of total DIC from terrestrial sources when compared with the cycling of carbon in terrestrial systems over decades and not millennia. In general, DIC exports were three- to fivefold greater than OC in the upper Midwest. OC dominated the lateral carbon fluxes from HUCs 01, 03, and 08 only. These high organic carbon fluxes are predominantly the result of significantly higher average concentrations of TOC. However, the proportion of wetlands within these HUCs was relatively high at 8%, 18%, and 20%, respectively, providing a large DOC source. There is strong correlation between weighted TOC yields and the proportion of wetlands within a region ($r^2 = 0.5$, $P < 0.01$).

Because of human activities, including agriculture and dam construction, sediment transport and burial in reservoirs and lakes has been assumed in the past to comprise a large component of the aquatic carbon budget (46). At the national scale, CO_2 emission from the water surfaces and burial in lakes and

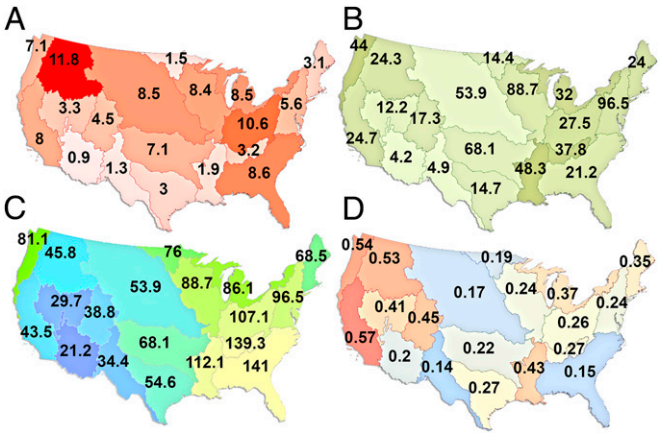

Fig. 1. (*A*) Total aquatic carbon flux measured in TgC per year by modified two-digit HUCs. (*B*) Normalized aquatic carbon yields in gC per square meter per year per area of each HUC using Eq. **2**. (*C*) Average NEP gC per square meter per year for the years 1990–2010 derived from the MsTMIP model outputs. (*D*) Maximum potential fraction of NEP accounted for by aquatic carbon if all aquatic carbon is derived from terrestrial sources [does not include (f_a) from Eq. **2**] represented as in *B* and *C*. Uncertainty ranges are presented in Table S5.

reservoirs is correlated ($r^2 = 0.73$, $P < 0.001$; Fig. S3 and Tables S3 and S4), but the residuals of the relationship reveal some distinct geographic trends. Overall, burial exceeds lake emissions by 28% and is nonlinear, where proportionally more burial takes place as lake emissions increase. However, this estimate is driven by the imbalance presented in HUC 08, where burial is more than double CO_2 emission. Carbon burial in lakes, ponds, and reservoirs correlates with the presence of wetlands. This drives the high carbon burial rates within HUC 08 (35). In arid regions of the United States dominated by large river systems, including the Columbia River and Colorado River (HUCS 14, 15, 17 Dry, and 18 Dry), carbon burial exceeds CO_2 emission by up to 900%, possibly reflecting the combined effects of greater soil loss through erosion due to sparse vegetation cover and agricultural nutrient enrichment of water bodies resulting in higher levels of autochthonous primary productivity (primary productivity in aquatic environments). Conversely, lake and reservoir CO_2 emission is greater than burial in HUCs 01, 02, 09, and 11. We do not currently have a unified hypothesis as to why emissions are larger than burial across such distinctly different regions. HUC 09 is dominated by lake systems shown to be net heterotrophic in the past (23), whereas carbon burial remained a small component over short time periods.

Carbon yields, defined as the carbon flux on a per unit watershed basis within a HUC, are a useful way to examine aquatic carbon export because they help to illustrate the connectivity between the aquatic fluxes and contributing terrestrial drainage area (Fig. 1*B*, Table 1, and Tables S5 and S6). HUC 17 Coast, representing the western Cascade and Coastal mountain ranges in the US Pacific Northwest, has an estimated yield of 41.6 (range: 24–60.9) $gC{\cdot}m^{-2}{\cdot}y^{-1}$. This high flux is dominated by high emission rates from stream and river surfaces. HUC 15 represents the lowest carbon yields at 2.5 (range: 1.4–4.3) $gC{\cdot}m^{-2}{\cdot}y^{-1}$, where total inland water carbon fluxes are smallest across all components of the aquatic ecosystem.

Terrestrial and Aquatic Carbon Accounting. There are two ways to evaluate the carbon fluxes through aquatic systems in the context of terrestrial carbon accounting depending on the goals of the analysis. As presented in Eq. **1**, the end accounting of carbon represents the net balance of carbon for the conterminous United States, where carbon burial in lakes, ponds, and reservoirs is considered stored within the national boundaries. This estimate is most relevant for the total accounting of carbon at the national level when considering the various natural and anthropogenic sources and sinks over time. However, to begin developing a broad framework to account for carbon in aquatic systems that may have originated from terrestrial vs. aquatic sources, summing all of the components of Eq. **1** reveals the total carbon moving into and out of aquatic systems aggregated to these modified two-digit HUCS (Fig. 1 *B–D* and Table 1). In addition, some fraction of each carbon flux must be accounted for as originating from autochthonous production and respiration:

$$\begin{aligned} HUC\ Flux\ (\mathrm{TgC}\cdot\mathrm{y}^{-1}) = \sum_{HUC} & [Stream\ Emission - (f_a)] \\ & + [Lateral\ Flux - (f_a)] \\ & + [Lake\ \&\ Reservoir\ Emission - (f_a)] \\ & + [Lake\ \&\ Reservoir\ burial - (f_a)]. \end{aligned} \quad [2]$$

The (f_a) term in each of the aquatic carbon fluxes represents a fraction of the related flux derived from autochthonous sources. In this analysis, we are unable to quantify the internal production and consumption of carbon within aquatic systems at such a large scale, but the effect of this process will reduce the overall mass of terrestrial carbon needed to support these flux estimates. Research suggests that the internal production of DOC and mineralization to CO_2 may contribute up to 28% of the total CO_2 efflux in rivers (18). However, the origin of that DOC is not quantified, suggesting that this DOC could remain a source from terrestrial carbon. Supersaturation in lakes across the conterminous United States has been shown to be supported by riverine DIC inputs; however, research is still unable to attribute the source of this inorganic carbon spatially (19). To our knowledge, there do not exist large-scale attempts to identify (f_a) that would be relevant for the accounting of carbon within the conterminous United States, which represents a significant knowledge gap requiring future research (an expanded discussion on autochthonous inputs and model spatial scales is provided in *SI Materials and Methods*). The net effect of autochthonous production for the attribution of total carbon fluxes will reduce the importance of terrestrial inputs by an unknown quantity.

A strong correlation exists between the total aquatic carbon yields calculated using Eq. **2** and average annual precipitation ($r^2 = 0.68$, $P < 0.001$; Fig. 2*A*). This correlation suggests that areas with high rates of water throughput, and available water storage in soils, have higher rates of carbon export into aquatic environments. Precipitation is emerging as a driver of many components that comprise the carbon chemistry in freshwaters (14, 32, 47). In contrast to previous work, we show that this relationship is maintained across regions where the lateral export of carbon or the surface evasion of carbon is dominant. These findings show that broad climatic patterns influence the movement of carbon across the terrestrial–aquatic interface and can provide insight into how aquatic carbon cycling can be integrated into a modeling framework.

Estimated freshwater carbon yields also correlate strongly with terrestrial NEP across the United States ($r^2 = 0.4$, $P < 0.01$; Fig. 2*B*) as well as terrestrial NPP ($r^2 = 0.35$, $P < 0.01$), total aboveground biomass ($r^2 = 0.49$, $P < 0.01$), and total respiration ($r^2 = 0.45$ $P < 0.01$) (data presented in Table S6). There was not a significant correlation between total soil carbon and total aquatic carbon yields. This last point is surprising because we hypothesize that landscapes with higher soil carbon stores will offer an opportunity for additional inorganic carbon and OC to be mobilized into aquatic environments. Given the strong correlation between aquatic carbon yields and precipitation, the relationship between terrestrial NEP and aquatic carbon yield is to be expected because precipitation is a significant variable that determines the turnover and processing of carbon within each of the models. Precipitation is a primary driver that determines the presence and form of above-ground biomass over time, as well as many of the surface energy balance terms (42). These relationships also support the intuitive notion that systems having high rates of carbon processing and larger estimates of NEP potentially have more carbon available for mobilization into the aquatic system (48). The finding that total soil carbon did not correlate with aquatic carbon yield may indicate that the rates of processing of carbon are more influential on

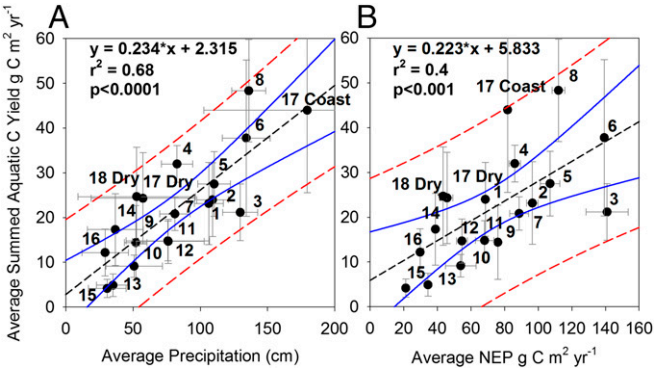

Fig. 2. Correlation between the summed aquatic carbon yield (Eq. **2**) and average annual precipitation (PRISM Climate Group, Oregon State University; prism.oregonstate.edu) (*A*) and NEP (MsTMIP model ensemble mean) (*B*) by HUC. Points are labeled by HUC. Red lines indicate the 95% prediction interval, and the blue lines represent the 95% confidence interval. Error bars represent the SD about the mean.

aquatic ecosystems, and not on the long-term standing stock of carbon for the conterminous United States.

By comparing summed regional aquatic carbon fluxes with the MsTMIP model output, where each model produces only an estimate for terrestrial carbon sources and sinks, we can assess the maximum potential proportion of NEP that is offset by the removal of carbon through streams, rivers, lakes, and reservoirs. For this analysis, we consider aquatic carbon fluxes and burial to be a removal of terrestrial carbon only across each of the two-digit HUCs. The inability to assess (f_a) from Eq. **2** biases these estimates high. At the national scale, a maximum of 26.8% of the estimated net uptake of carbon in terrestrial ecosystems may be offset by the inclusion of freshwater carbon cycling (Tables S5 and S6), calculated as the ratio of the average carbon yield from Eq. **2** (Fig. 1*B*) to the average terrestrial NEP from the MsTMIP output (Fig. 1*C*). There are significant regional differences in the relative importance of freshwater systems to terrestrial carbon accounting. Our analysis indicates that in the Pacific Northwest, between 53% and 56.8% of the total NEP in region 17 could potentially be redirected to freshwater ecosystems, and returned to the atmosphere through remineralization and respiration (reduced NEP) or deposited in coastal and ocean environments (net sink) (Fig. 1*D* and Table S5). These estimates are potentially biased high due to poorly constrained estimates of the surface gas transfer velocities in streams within steep topography (49). HUC 03 represents the smallest differential between the positive aquatic carbon flux and the terrestrial sink of carbon (15%). This small ratio is driven by the high rate of NEP in the southeastern United States, 141 (SD = 125.8) $gC{\cdot}m^{-2}{\cdot}y^{-1}$ (Fig. 1*C*). These potential reductions in the net terrestrial sink of carbon would reduce regional estimates of the efficiency of managed forested and agricultural ecosystems as a means to reduce greenhouse gas emissions. However, further work is needed to partition the contribution of internal production of inorganic carbon and OC (f_a) within inland waters spatially to constrain the linkages across terrestrial and aquatic ecosystems better (18).

Reconciling Carbon Budgets Across Ecosystems: Future Needs. For larger scale studies, the extent to which aquatic carbon cycling is included in carbon budgets depends upon the accounting methods used. Atmospheric inversion modeling includes the degassing of CO_2 from water surfaces within the model domain, and eddy covariance measurements capture emissions from water surfaces within the tower footprint; however, lateral fluxes are missed. Inventory-based methods of calculating NEP, designed to measure the accumulation of carbon in soils and biomass physically at the scale relevant for national carbon accounting, exclude any loss of inorganic carbon and OC to aquatic systems, and hence any relocation of carbon in soils.

The terrestrial carbon fluxes presented here are derived from dynamic TBMs. The ability to use spatially explicit aquatic carbon models to match terrestrial ecosystems models has not yet been developed but is critical to identify fully where and how freshwater ecosystems may affect the calculated uptake of carbon in both land and water and to provide resource managers integrated tools to strategize policy decisions for carbon sequestration at differing landscape scales (50).

At 106 $TgC{\cdot}y^{-1}$, the removal of carbon by inland waters within the conterminous United States is substantial at the national scale. However, this analysis represents a snapshot of a dynamic environment that is continuously processing both allochthonous and autochthonous carbon (22, 51). Developing a model framework that couples the biogeochemical processing of inorganic carbon and OC in soils to the movement of water through soils to inland waters is needed to quantify (*i*) terrestrial carbon sources to inland waters, (*ii*) seasonal inputs of carbon to freshwater systems under a changing climate, and (*iii*) anthropogenic influences on freshwater carbon processing (47, 52, 53). There are few direct measurements of the suite of carbon-based greenhouse gases in freshwater ecosystems, particularly methane (11). Methane is emerging as a persistent significant greenhouse gas from freshwater systems (54). Similarly, large uncertainty remains for calculations of lake and reservoir sedimentation and the impacts of physical processes that affect the distribution of sediments along lake and reservoir beds. Continued monitoring and the integration of new measurement techniques for dissolved carbon and carbon gases are important to begin developing both the modeling and databases needed to understand fully the role of inland waters in the carbon cycle.

Materials and Methods

An expanded description of all methods used in this analysis is presented in *SI Materials and Methods*. Total flux estimates for carbon emissions with each two-digit HUC from water surfaces are described by Stackpoole et al. (33) using the general equation:

$$[fCO_2C](\mathrm{Tg}) = \sum_{HUC}\left[\sum_{SO}\left[\left[CO_{2-water} - CO_{2-air}\right] * kCO_2 * SA\right]\right]. \quad [3]$$

The term fCO_2C is the total net emission of CO_2 from riverine, lake, and reservoir systems of the conterminous United States. $CO_{2\text{-}water}$ is the aquatic CO_2 concentration (moles per liter) derived from calculated estimates of dissolved CO_2 using measured alkalinity, temperature, and pH available at the National Water Information System (NWIS; waterdata.usgs.gov/nwis/) for streams and rivers, and from the Environmental Protection Agency National Lakes Assessment (www2.epa.gov/national-aquatic-resource-surveys/national-lakes-assessment) for lakes and reservoirs. $CO_{2\text{-}air}$ is the CO_2 equilibrium concentration of atmospheric CO_2 in water (moles per liter). kCO_2 is the gas transfer velocity of CO_2 across the air–water interface (meters per day). *SA* is aquatic surface area (square meters). Lateral carbon flux is represented by multiple rivers for coastal regions of the United States, including the Atlantic Coast, Pacific Coast, Gulf Coast, and Great Lakes (HUCs 01, 02, 03, 04, 12, 17 Coast, and 18). Carbon concentration and flow data to estimate longitudinal carbon fluxes were obtained from the NWIS (waterdata.usgs.gov/nwis/). Carbon fluxes (kilograms per day) were estimated from water quality and daily stream flow data, using the USGS Load Estimator Model (LOADEST) (55). Methods to estimate OC burial in lakes and reservoirs are fully described by Clow et al. (35) and outlined in *SI Materials and Methods*. OC burial was estimated for each water body in the conterminous United States (CONUS) using the following equation:

$$\mathrm{OC_{burial}}\left(\mathrm{TgC}\cdot\mathrm{y}^{-1}\right) = \left[\mathrm{WB_{area}}\ \left(\mathrm{m}^2\right) \times \mathrm{SedRt}\left(\mathrm{g}\cdot\mathrm{m}^{-2}\cdot\mathrm{y}^{-1}\right) \times \mathrm{OC_{conc}}(\%)/100 \times \mathrm{BE}(\%)/100\right] \times 10^{-12}, \quad [4]$$

where OC_{burial} is the OC burial rate, WB_{area} is water body area, SedRt is sedimentation rate, OC_{conc} is OC concentration in sediments (percentage by dry weight), BE is burial efficiency, and 10^{-12} is a conversion factor from grams to teragrams (35). Water bodies are derived from similar sources as those sources used for efflux calculations. All TBM data processing is available as NetCDF files from the MsTMIP model (nacp.ornl.gov/mstmipdata/).

ACKNOWLEDGMENTS. Aggregation of the data presented here was performed by Xiaodong Chen (University of Washington Department of Civil & Environmental Engineering). This work was fully supported by the US Geological Survey (USGS) Land Carbon Program, through the broader efforts of the USGS Biological Carbon Sequestration Program. Data management support for preparing, documenting, and distributing model driver and output data was performed by the Modeling and Synthesis Thematic Data Center at Oak Ridge National Laboratory (nacp.ornl.gov), with funding through National Aeronautics and Space Administration Grant NNH10AN681.

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