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Global Carbon Dioxide Emissions from Inland Waters

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47 Carbon dioxide transfer from inland waters to the atmosphere is a significant
48 component of the global carbon cycle. Global estimates of CO₂ transfer have been hampered,
49 however, by a lack of a framework for estimating the inland water surface area and gas transfer
50 velocity and the absence of a global CO₂ database. Here we report regional variations in global
51 inland water surface area, dissolved CO₂ and gas transfer velocity. We obtain global CO₂ evasion
52 rates of 1.8 Pg C yr⁻¹ (1.5-2.1 5th and 95th confidence intervals) from streams and rivers and
53 0.32 Pg C yr⁻¹ (0.060-0.84 5th and 95th confidence intervals) from lakes and reservoirs. The
54 resulting global evasion rate of 2.1 Pg C yr⁻¹ is higher than previous estimates due to a larger
55 stream and river evasion rate. Our analysis predicts global hot spots in stream and river evasion
56 with about 70 percent of the flux occurring over just 20 per cent of the land surface. The source
57 of inland water CO₂ is still not known with certainty and new studies are needed to research the
58 mechanisms controlling CO₂ evasion globally.

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Inland Waters and the Global Carbon Budget

Quantifying the earth's global carbon cycle is essential for a sustainable future due to the active role CO₂ plays in the earth's energy budget. Natural ecosystems are important to this accounting because they exchange large amounts of CO₂ with the atmosphere and currently offset ~4 Pg C yr⁻¹ of anthropogenic emissions¹. To date, estimates of the global exchange of CO₂ between inland waters and the atmosphere have not been made using comprehensive, spatially resolved efforts. It was shown definitively 30 years ago that CO₂ in inland waters calculated from alkalinity and pH were substantially higher than atmospheric values². Early direct measurements, of large rivers and arctic inland waters also demonstrated super-saturation³⁻⁶. The first regional estimate of inland water degassing, which was for the Amazon, did not appear in the literature until 2002⁷. This study estimated the release of ~0.5 Pg C yr⁻¹ from streams, rivers and wetlands of this region alone, and was revised upward to account for a large degree of CO₂ super-saturation in small headwater streams⁸. Recently the total CO₂ emitted from the contiguous United States streams and rivers was estimated at ~0.1 Pg C yr⁻¹, extrapolated to 0.5 Pg C yr⁻¹ for temperate rivers between 25° and 50° north⁹.

There are few global estimates of inland waters CO₂ evasion¹⁰⁻¹³. These studies still place the efflux at only ~1 Pg C yr⁻¹¹⁰⁻¹³, despite the high fluxes estimated for temperate rivers and the Amazon. To date, global exchange calculations are simple in nature and prone to uncertainties in all three factors which determine inland water CO₂ evasion: the amount of CO₂ in water, the global surface area of streams, rivers, lakes and reservoirs, and the gas transfer velocity (*k*, a parameter which relates to the physics that determines the rate of gas exchange). Recently, studies have revisited the scaling of lake and reservoir surface area, using new

geospatial data sets¹⁴⁻¹⁶ which we adapted to produce the first spatially explicit global maps of lake and reservoir surface area divided by size classes. Other studies have also probed the controls and quantities of lake dissolved CO₂ at the large catchment scale¹⁷⁻²⁰ and improved our knowledge on the controls of the gas transfer velocity in lake systems and lakes and reservoirs^{21,22}, which we synthesized here for our global estimate.

Studies in rivers and streams have also progressed. Regional studies have attempted a more systematic estimation of stream and river evasion for Sweden, the United States and the Yukon River Basin^{9,19,23}. This approach entails utilizing stream scaling laws and high resolution remote sensing information that exists for these regions. Although similar high resolution maps are not available globally for streams and rivers, we provide a new spatially resolved global stream surface area and gas transfer velocity utilizing coarser global datasets that have recently been developed²⁴, combined with river scaling laws^{25,26}, discharge estimates for global drainage basins²⁷ and new knowledge on the controls of the gas transfer velocity for streams and rivers^{28,29}.

We have combined these new approaches for estimating the global inland water surface area and gas transfer velocity with a new global data set of calculated pCO₂ (based on the GLORICH database³⁰) in order to provide spatial maps of inland water CO₂ evasion along with uncertainty intervals using this approach. We perform our scaling using the COSCAT (Coastal Segmentation and related CATchment) drainage network segmentation framework³¹ which lends itself to drainage basin analysis and allows for the spatial representation of this exchange.

Inland Water Surface Area

We find a strong positive correlation between stream/ river surface area and precipitation and a weaker negative relationship between surface area and temperature (Supplementary Information Figure SI4). The robust relationship between stream area and precipitation is driven mostly by a strong positive correlation between stream order width and precipitation and therefore efforts that use a global average stream width for all streams and rivers will not capture higher surface area of streams and rivers in wetter regions of the globe. Globally we predict a 0.07% increase in the fraction of stream area for a 10 cm increase in precipitation and a 0.02% decrease with a 1 degree increase in temperature (Supplementary Information). These correlations, which have also been demonstrated with satellite measurements³², are important to global change studies because they reveal a potential link between water cycle changes and inland water surface area.

We first calculate a global stream and river surface area of 624,000 km² (487,000-761,000km²), or 0.47% of the earth's surface (Antarctica is excluded from this analysis). The estimate of 624,000 km² is corrected for ephemeral and intermittent stream fraction periods (Supplementary Information), which removed ~84,000km² of stream surface area from contributing to gas exchange. This is towards the upper limit of a recent estimate of 485,000-662,000 km²³³. However the latter study may not have captured first order streams, which are included here (Supplementary Information). Previous studies also did not account for spatial variability in width and therefore possibly underestimated the contribution of surface area from wet regions of the globe. Our analysis predicts a significant contribution to total stream and river surface area from small streams (Table S1) accounting for ~15% of global stream area. We also corrected for the amount of frozen streams with little gas exchange (the effective surface

area, see Supplementary Information), further reducing our estimate down to 536,000km² (Supplementary Information). Using this effective surface area weakens the strength of the negative correlation between temperature and stream surface area. High surface area is estimated in areas of the tropics and temperate regions of the globe (Figure 1).

We estimate a global lake and reservoir surface area of 3,000,000km² or 2.2% of the earth's surface, of which 91.3% is lakes and 8.7% is reservoirs. Our estimate was arrived at using a combination of empirical data for large lakes with statistical models based on regional inventories of smaller lakes (Table S4 in Supplementary Information). These estimates of surface area are lower than a recent estimate³⁴ but proximate to others³⁵. Our lake surface area is lower than some recent estimates because we estimate a smaller contribution from small lakes (Table S4) due to recent work which demonstrates that the size distribution of small lakes is independent of that of large lakes¹⁶. Combining lakes and reservoirs with streams and rivers provides a total surface area of inland waters of 3,620,000km². High coverage of lakes can be found in previously glaciated landscapes of temperate and arctic regions, and mountain regions, where glacial movements and tectonic activity have created a multitude of depressions (Figure 2). It should be noted that the estimate of surface area does not include wetlands. We believe wetlands are functionally different than inland waters due to a canopy of vegetation that can alter the direction of atmospheric CO₂ exchange.

Inland Water Carbon Dioxide

CO₂ in inland waters are generally supersaturated with respect to water in equilibrium with the atmosphere. Of the 6708 stations for streams and rivers, 95% had a median CO₂

concentration above atmospheric values (Supplementary Information). The average of these median values was $\sim 2300 \mu\text{atm}$, however in our Monte Carlo, we report an average pCO_2 of $\sim 3100 \mu\text{atm}$ when discounting for potential biases in the calculation and normalizing interpolated pCO_2 from each region to stream area (Supplementary Information). It is important to note that we were not able to assign CO_2 by stream order for this study. An average of $3100 \mu\text{atm}$ is within the range of $\sim 1300\text{--}4300 \mu\text{atm}$ for previous regional or global studies^{7,10,28,36}. The concentration of CO_2 in water was not found to be strongly related to climatic or landscape variables (Supplementary Information), which is consistent with a recent study for North America³⁰ that showed strong correlations between climatic and landscape variables and alkalinity and pH, but only weak correlations with CO_2 .

We assemble 20,632 pCO_2 observations from 7939 lakes and reservoirs which were also generally supersaturated. Three groups of lakes could be distinguished based on their pCO_2 : non-tropical freshwater lakes, tropical lakes and saline lakes; reservoirs were treated as similar to natural lakes because their pCO_2 has been shown to be elevated only during the initial ~ 15 years after impoundment^{37,38}. Non-tropical freshwater lakes had a median pCO_2 of $1120 \mu\text{atm}$ and a mean of $1410 \mu\text{atm}$ (Supplementary Information). Tropical and saline lakes were higher and lower in pCO_2 , respectively (Supplementary Information), although these lakes had very small representation in the data set (1.5 and 0.8% respectively). Also, the median values were significantly different than the mean, with the mean values being 4390 and $1190 \mu\text{atm}$, for tropical and saline lakes, respectively and 1910 and $270 \mu\text{atm}$ for the median. We therefore utilized the median values to upscale to lakes in tropical and endorheic regions due to the potential for over-estimates when calculating CO_2 from alkalinity and pH, and to avoid any bias

from a few very high $p\text{CO}_2$ values (Supplementary Information). In non-tropical freshwater lakes, CO_2 was positively correlated with the concentration of TOC and negatively correlated with lake size (Supplementary Information), and these correlations were used to extrapolate lake CO_2 for non-tropical exorheic COSCAT regions of the globe. Globally dissolved CO_2 normalized to lake area was $\sim 800 \mu\text{atm}$. Lake $p\text{CO}_2$ is highest in the humid tropics and also in some boreal regions owing to high TOC concentrations (Figure 2).

Inland Water Gas Transfer Velocity

The global average gas transfer velocity of 5.7 m d^{-1} for streams and rivers (range of 5.0-6.3) is close to recent regional studies^{28,29} but significantly higher than a number used in a recent global calculation¹⁰ and for the Amazon⁷ which was not estimated systematically in the case of the former or done before many measurements were available in the case of the latter. We also predict a decreasing gas transfer velocity with increasing stream order (Table S1 Supplementary Information), which is consistent with recent field measurements³⁹. In a new meta-data analysis of whole stream tracer releases in streams and small rivers the average value was 4.7 m d^{-1} ²⁹. These experiments, however, were limited to low discharge and because turbulence is positively correlated with discharge the value reported for small streams and rivers here are reasonable for average flow conditions. For large rivers we predict a gas transfer velocity of $\sim 3\text{-}4 \text{ m d}^{-1}$ (Table S1), which is also close to a recent synthesis for lowland rivers²⁸ which reported an average of 4.3 m d^{-1} and argued that many studies to date have probably underestimated k , which is generally higher in wet mountainous regions (Figure 1).

We used two methods to estimate the gas transfer velocity for lakes and reservoirs. The first utilized globally gridded wind speed and an empirical relationship between k_{600} and wind²¹ (Supplementary Information). The second utilized new estimates of the gas transfer velocity for lakes of different sizes²², which assumes a primary role of fetch on regulating k in these systems. The wind speed and lake size models provided global average estimates of 0.74 and 1.33 m d^{-1} , respectively. Thus a global average k for lakes and reservoirs is approximately 1.0 m d^{-1} , which is much lower than the global average for streams and rivers (Figure 2), but consistent with a recent regional study⁴⁰.

Global CO₂ Evasion from Inland Waters

Our estimated fluxes are lower than the most recent estimates for lakes and reservoirs but higher for streams and rivers. For streams and rivers we estimate a flux of 1.8 Pg C yr^{-1} . This is higher than previous studies that have reported a stream and river evasion rate of $\sim 0.5\text{--}1 \text{Pg C yr}^{-1}$ ¹⁰⁻¹², yet defensible considering stream and river evasion rates of 0.5 Pg C yr^{-1} from temperate regions⁹ and $\sim 0.6 \text{Pg C yr}^{-1}$ from the Amazon^{7,8} alone. For lakes and reservoirs our estimate of $\sim 0.3 \text{Pg C yr}^{-1}$ is lower than the most recent estimates of $\sim 0.5\text{--}0.6$ ^{10,41}, but proximate to some of the older estimates^{12,42} (Figure S7). This new estimate is lower than more recent estimates due to a smaller lake and reservoir area ($3 \times 10^6 \text{ km}^2$ compared to $4.2 \times 10^6 \text{ km}^2$), and because we used median instead of the mean as a representative value for the skewed distributions of pCO_2 , particularly in saline lakes. Lastly, we account for generally lower pCO_2 in large lakes and reservoirs, which are important to the total area (Figure 2).

There is a large amount of uncertainty associated with these estimates. We performed a Monte Carlo analysis to estimate variance of our methodology by providing a distribution for

the gas transfer velocity, surface area, and dissolved CO₂ concentration for each COSCAT region and then randomly sampled within these distributions for 1000 iterations (Supplementary Information). The simulation predicted a flux of 1.8 Pg C yr⁻¹ for streams and rivers (5th and 95th percentiles of 1.5 and 2.1 Pg C yr⁻¹) and 0.31 Pg yr⁻¹ for lakes and reservoirs (5th and 95th percentiles of 0.06-0.84 Pg C yr⁻¹). For streams and rivers the uncertainty within COSCAT regions was positively correlated to the mean value of the flux, with regions with a high flux normalized to land area having the highest standard deviation (Figure S5). For lakes and reservoirs the large range in the confidence interval is due to the non-linear relationship between lake abundance and area and uncertainty in the number/area of small lakes which currently cannot be counted at the regional scale. In addition to the uncertainty estimated by the Monte Carlo analysis, there is considerable uncertainty in inland water science that may impact these estimates. Although we attempted to account for it in our analysis by using medians and adjusting the high range for the stream/river Monte Carlo analysis (Supplementary Information), there is still the potential that this method is overestimating stream and river CO₂ due to potential biases and errors with calculating CO₂ from pH and alkalinity and the presence of organic acids (see Supplementary Information). The overestimation of CO₂ is potentially impacting areas with few calculated CO₂ values and high fluxes such as Southeast Asia (Supplementary Information). Representative pCO₂ measurements are needed globally. In addition to improved CO₂ estimates, future research is needed on the distribution of lakes to refine estimates of lake area. Another large research gap is a lack of measurements of stream k during average to high flows and in watersheds with a high slope. High resolution global maps of stream length are still missing for the high latitudes. Further research on hydraulic

relationships is needed particularly in the tropics and high latitudes. For lakes, representative winter CO₂ measurements are missing and are often several fold higher than during other seasons⁴³. A further discussion on data limitations is provided in the Supplementary Information.

A flux of 1.8 Pg C yr⁻¹ for streams and rivers is large considering their small surface area, reinforcing the concept that streams and river are hotspots for exchange. Approximately 70% of the stream CO₂ evasion originates from waters located on only ~20% of the earth's surface. Regions supporting this evasion include Southeast Asia, Amazonia, Central America, Europe, regions of South America west of the Andes, Southeast Alaska, small portions of western Africa, and the eastern edge of East Asia (Figure 1). Missing from this list is most of the northern latitude regions. The COSCAT drainages that include the Yenisei, Lena, Kolyma and Yana, for instance, make up ~6% of the earth's surface area, but are responsible for only ~2% of global evasion. It is important to note that the surface area of northern latitudes are mainly extrapolated from relationships at low latitudes (Supplementary Information) and these regions may have unique scaling laws and biogeochemistry that are currently not adequately understood. Thus the evasion of CO₂ from northern latitudes needs further research. Africa, which is under-sampled for CO₂, also has a predicted low contribution, making up ~22% of the terrestrial surface area but supporting only ~6% of annual CO₂ evasion.

This study further stresses the disproportionately high contribution of lower order streams. We report a decreasing percentage in stream surface area and gas transfer velocity with increasing stream order (Table S1 Supplementary Information). It is worth noting that the lower order systems are under-sampled for CO₂, are not consistently gauged, and their surface

area is difficult to directly measure by remote sensing. This study was not able to assign CO₂ by stream order, but previous studies argue for higher concentration of CO₂ in small streams and rivers^{9,19}. Further study on the surface area and CO₂ of small stream is needed.

For lakes and reservoirs, regions of high fluxes were estimated from the high latitudes and tropical regions (Figure 2). We also conclude that ~50% of the emissions are from the world's largest lakes due to their large surface area and gas transfer velocity (Supplementary Information). However, large lakes are currently inadequately surveyed for both concentration and k. We also conclude that tropical lakes contribute disproportionately (Figure 2), constituting only 2.4% of the global lake area, but accounting for 34% to the global lake CO₂ emission, owing to high pCO₂ and high gas exchange rates. This could be due to the higher frequency of flooding of tropical lakes which enhances terrestrial transfers. Lake CO₂ emissions per land area were highest in the humid tropics, but also in lake-rich boreal and arctic regions (Figure 2). Saline lakes, in contrast, are less important than previously reported⁴⁴, contributing ~18% to the global lake CO₂ evasion rather than ~50%. Much of this evasion is due to the Caspian Sea, the largest freshwater body on earth which has some calculated estimates of CO₂ but no proper survey.

The importance of the entire drainage network to CO₂ evasion provides information on the origins of inland water CO₂. The high evasion rate in small order streams is consistent with a large terrestrial soil CO₂ supply⁸, which could also be important to lake effluxes^{40,45}. The evasion of this CO₂ is, however, rapid⁴⁶ and cannot explain all of the evasion from higher order systems and lakes and reservoirs. Although additional terrestrial soil CO₂ can still be added to these systems via groundwater, contributions from organic matter decomposition and

inorganic and organic carbon subsidies from fringing wetlands⁴⁷ are still needed to sustain global CO₂ evasion rate of 2.1 Pg yr⁻¹. The role of wetlands could be particularly important in hotspots such as Amazonia and SE Asia. Systematic campaigns are needed to further evaluate the relative importance of different sources.

Understanding the relative importance of these sources is crucial to the global carbon budget. The evasion of terrestrial soil CO₂ in inland waters is part of terrestrial respiration. Although a 2.1Pg C yr⁻¹ displacement of global terrestrial net primary production (NPP) to inland waters represents only ~4% of NPP, the difference between terrestrial heterotrophic respiration and fires (R_{h+f}) and NPP is on the order of ~1.5 Pg C yr⁻¹⁴⁸. Terrestrial approaches that attempt to determine the difference between R_{h+f} and NPP differ in their ability to account for inland water evasion of CO₂. A recent study demonstrated that ~1.2-2.2% of terrestrial NPP is evaded from lakes in catchments of England⁴⁵, thus ignoring inland water CO₂ evasion could cause significant errors in regional-scale CO₂ budgets from methods that rely on ecosystem-level CO₂ flux measurements. The percentage of evasion supported by terrestrial OM decomposition added to the amount of terrestrial OM exported by rivers to the coastal ocean also determines the total flux of terrestrial OM from the landscape, a flux not currently well constrained globally. Finally, if only a percentage of this flux has an anthropogenic component it is important to the attribution of anthropogenic carbon in the global carbon budget^{49,50}.

Methods Summary

For inland waters we relied almost exclusively on calculated CO₂. CO₂ was calculated from pH, alkalinity and temperature using PhreeqC v2. Water chemistry data was culled from

the literature and various governmental data sets and incorporated into the GLORICH database. Data were collected and digitized over a period of ten years. For this analysis, 6708 sampling locations were identified for streams and rivers and 25,699 single observations for lakes and reservoirs (Supplementary Information).

The surface area of inland waters was estimated using various geospatial products and scaling. For streams and rivers we utilized HydroSHEDS²⁴ and NHDplus to estimate length and hydraulic equations from the literature and USGS along with global gridded runoff data²⁸ to estimate width. This could only be done for regions <60°N and for regions above this we utilized statistical relationships from regions <60°N. For lakes and reservoirs we utilized the GLWD data set for lakes >3.16km² and utilized size distribution relationships from the literature^{16,33} to extrapolate to smaller lake and reservoirs.

For streams and rivers we estimated the gas transfer velocity (k) using a recently published equation³⁰ that estimates k based on slope and velocity. Velocity was estimated using hydraulic equations from the literature and USGS along with global gridded runoff data²⁸. Slope was determined using stream lines from HydroSHEDS and elevation data from multiple sources (see Supplementary Information). For lakes and reservoirs we used two approaches for estimating the gas transfer velocity. The first utilized the relationship between k and wind speed given by Cole & Caraco (1998) while the second used the recently published relationship between lake area and k ²².

We calculated fluxes and tested the uncertainty of this efflux calculation using a Monte Carlo simulation (Supplementary Information).

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Author Contribution

Peter Raymond conceived and performed this analysis and was responsible for the majority of writing. Sebastian Sobek performed the lake and reservoir CO₂ and k analyses, and Cory McDonald modeled lake and reservoir area data and provided material for these calculations for the SI. Pirkko Kortelainen provided pCO₂ data and supported lake analyses. Ronny Lauerwald and Jens Hartmann produced the global CO₂ data set. Mark Hoover provided the GIS technical input. David Butman assisted in GIS technical input and overall analysis and helped produce the figures. Rob Striegl provided input on the use of USGS data and overall analysis. Emilio Mayorga provided global discharge and DOC data by COSCAT. Hans Durr provided COSCAT information and input on GIS analysis. Pirkko Kortelainen, Christoph Humborg and Michel Meybeck provided data for the lake CO₂ global data set. Philippe Ciais provided assistance with sensitivity analysis and writing the final paragraph. Peter Guth provided data necessary to determine average watershed area for COSCAT regions. All authors read and commented on drafts of this paper.

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476 Figure 1. Maps of stream/river gas exchange parameters. Included, from top to bottom, are the
477 $p\text{CO}_2$ of streams and rivers (a; μatm), the effective surface area (b; %), stream gas transfer
478 velocity (c; m d^{-1}), and CO_2 efflux (d; g m^{-2} of land surface yr^{-1}).

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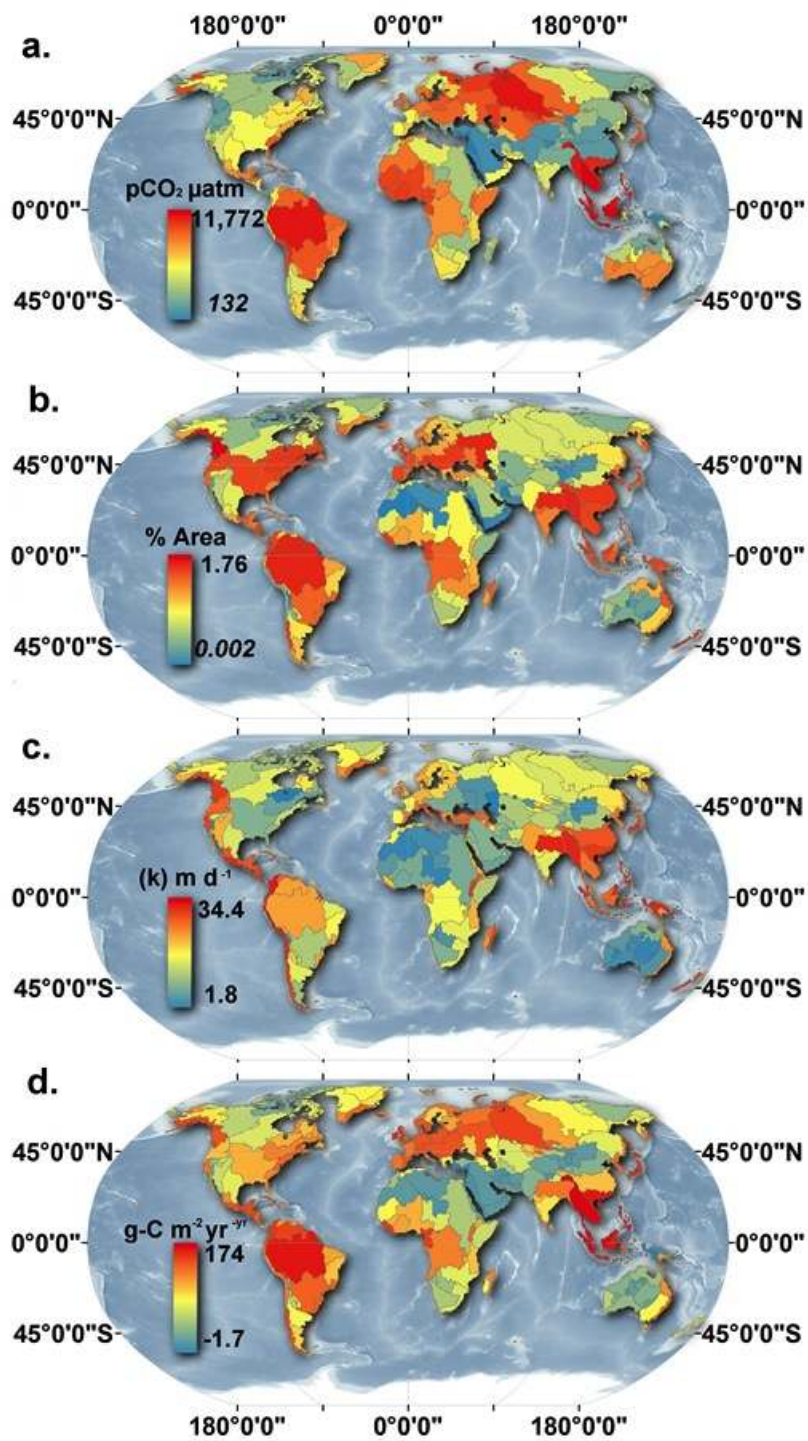
480 Figure 2. Maps of lake/reservoir gas exchange parameters. . Included, from top to bottom, are
481 the $p\text{CO}_2$ of lakes and reservoirs (a; μatm), the effective surface area (b; %), stream gas transfer
482 velocity (c; m d^{-1}), and CO_2 efflux (d; g m^{-2} of land surface yr^{-1}).

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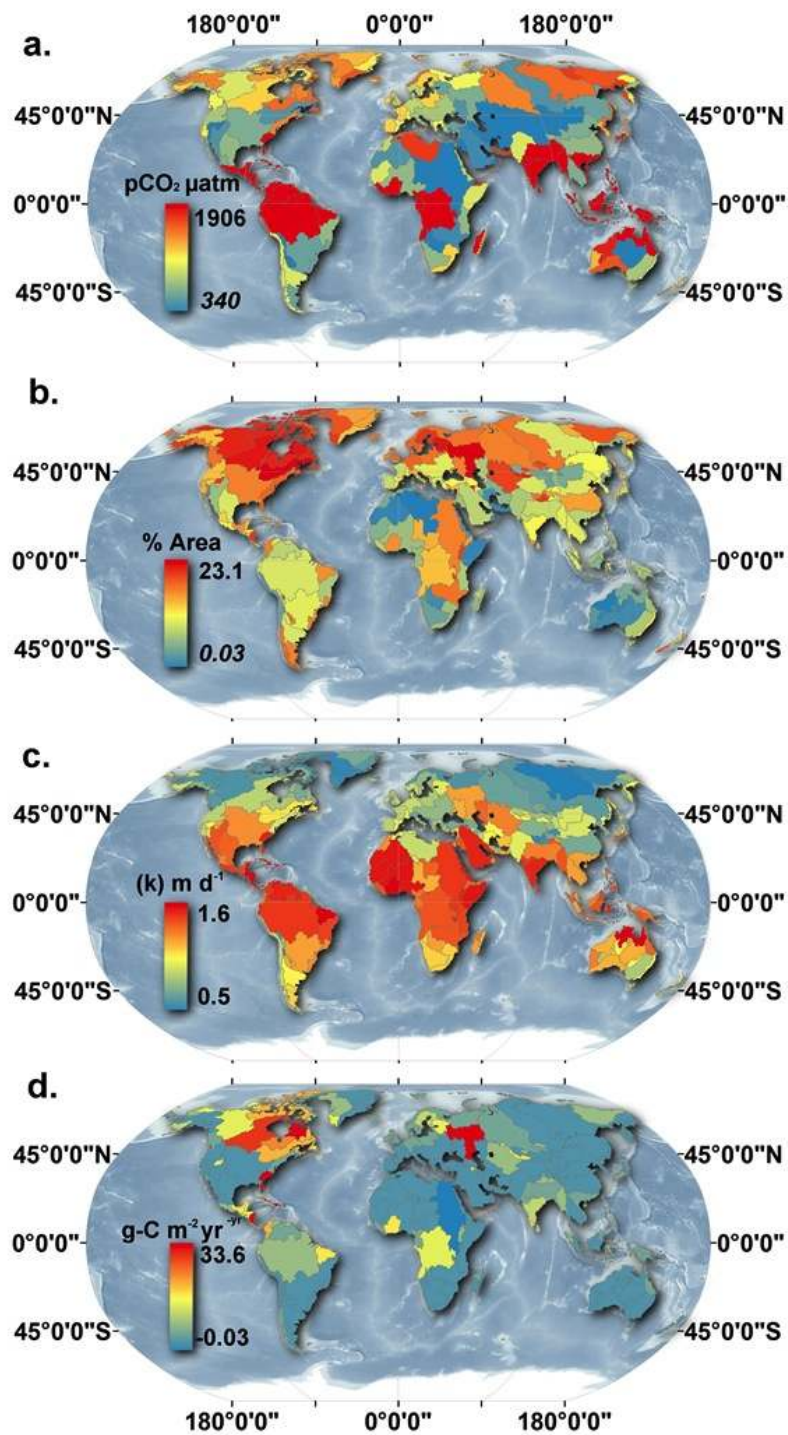
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Figure 1.



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487 Figure 2.



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