Report

Human-caused increases in reactive nitrogen burial in sediment of global lakes

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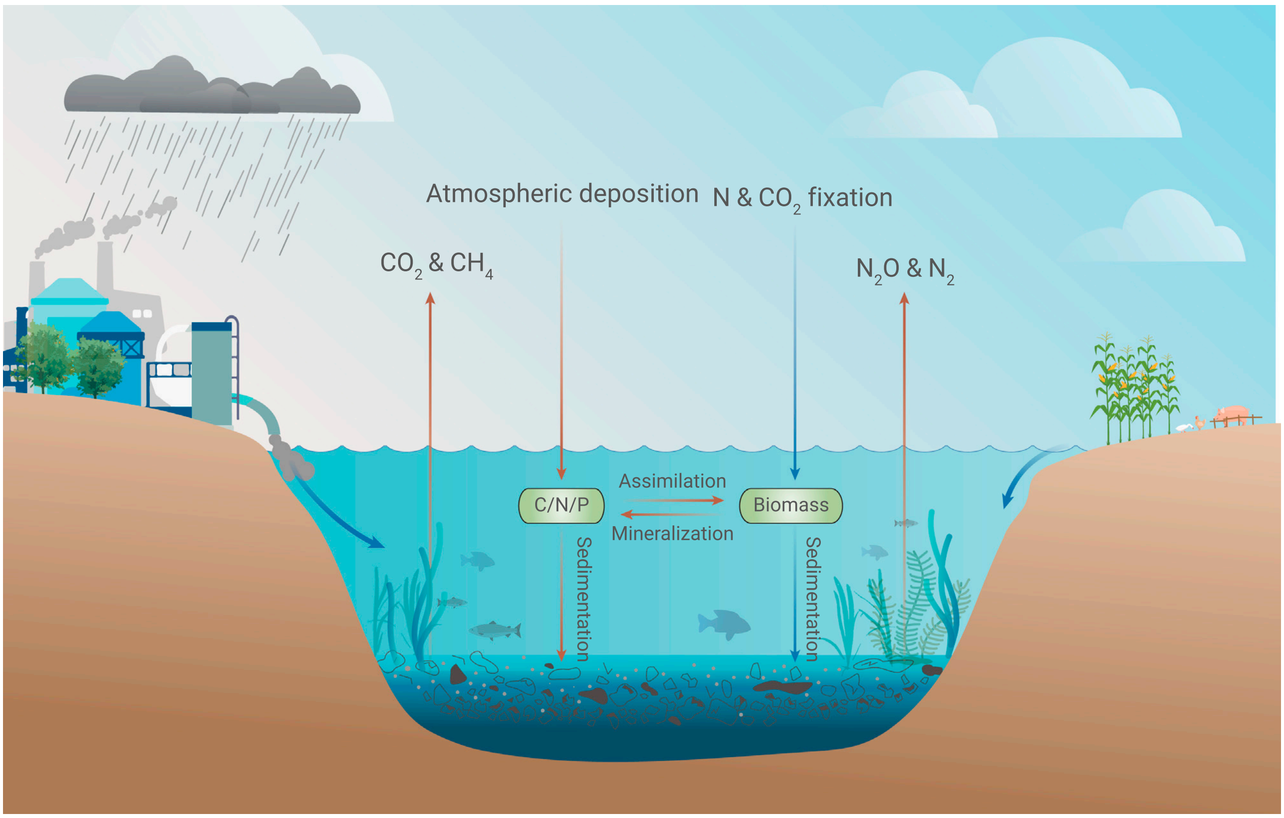
Received: February 28, 2021; Accepted: August 22, 2021; Published Online: August 12, 2021; https://doi.org/10.1016/j.xinn.2021.100158

Graphical abstract



Public summary

- Ten million tons of nitrogen was buried in lake sediment annually during 2000–2010
- Lake nitrogen burial rate is increasing since the 1860s
- Nitrogen burial is highly correlated with carbon burial rate in lakes
- Nitrogen burial in lakes can explain part of the global missing nitrogen sink

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Received: February 28, 2021; Accepted: August 22, 2021; Published Online: August 12, 2021; https://doi.org/10.1016/j.xinn.2021.100158

Citation: Wang M., Houlton B.Z., Wang S., et al., (2021). Human-caused increases in reactive nitrogen burial in sediment of global lakes. The Innovation **2(4)**, 100158.

Human activities have increased reactive nitrogen (Nr) input to terrestrial ecosystems compared with the pre-industrial era. However, the fate of such Nr input remains uncertain, leading to missing sink of the global nitrogen budget. By synthesizing records of Nr burial in sediments from 303 lakes worldwide, here we show that 9.6 ± 1.1 Tg N year^{-1} (Tg = 10^{12} g) accumulated in inland water sediments from 2000 to 2010, accounting for 3%–5% of global Nr input to the land from combined natural and anthropogenic pathways. The recent Nr burial flux doubles pre-industrial estimates, and Nr burial rate significantly increases with global increases in human population and air temperature. Sediment ratios of C:N decrease after 1950 while N:P ratios increase over time due to increasingly elevated Nr burial and other related processes in lakes. These findings imply that Nr burial in lakes is overlooked as an important global sink of Nr input to terrestrial ecosystems.

Keywords: **biogeochemistry; nitrogen accumulation; sink; eutrophication; carbon**

INTRODUCTION

Nitrogen is important to marine and inland water primary production.[1] When excess nitrogen enters aquatic ecosystems it causes a cascade of negative influences on the environment, including eutrophication, hypoxia, and fish kills.[1,2] Increased input of reactive nitrogen (Nr, all nitrogen species other than inactive nitrogen gas) to water bodies due to terrestrial fertilizer applications and anthropogenic nitrogen deposition has resulted in severe eutrophication globally, threatening the safety of drinking water and the health of aquatic ecosystems, and harming local economies.[3,4] Compared with the pre-industrial era, human activities have tripled the total Nr input to terrestrial ecosystems,[5,6] but the fate of such Nr input remains uncertain.[7,8] Nitrogen measurements in the atmosphere and estuaries reveal that less than half of Nr input to land surface is exported to the ocean via air and/or water flows, or emitted to the atmosphere[7] as nitrogen oxides (NO_x) and ammonia (NH_3).[9] The remaining Nr either accumulates in ecosystems or is denitrified to nitrous oxide (N_2O) or dinitrogen (N_2),[8,10] albeit with significant uncertainty surrounding magnitudes.[11,12] Understanding Nr accumulation in inland water is crucial for constraining the global nitrogen cycle and providing a scientific basis for maintaining environmental and public health.[13,14]

Despite substantial scientific progress, the total magnitude of sedimentary Nr burial in inland water lakes and its changes over time are rarely systematically examined, particularly at the global scale.[5,15] Sediments can be either net sources or sinks depending on biological and geological controls on nitrogen cycling (Figure 1). Deposition of elements into lake sediments causes them to subsequently be either immobilized or released as Nr.[12] Nitrogen burial in lakes could be leading to a globally significant nitrogen sink with implications for Nr's influence on aquatic health.[16,17] Thus, quantifying changes of Nr burial in lakes is critical to improved understanding of fates and recovery of Earth's ecosystems to anthropogenic Nr loadings (Figure 1). In this study, we aim to (1) quantify the Nr burial in lakes globally from 1850 to 2010; (2) identify the driving forces of the temporal dynamics of lake Nr burial ; and (3) demonstrate the role of lake Nr burial in the global nitrogen cycle.

RESULTS

Increase of Nr burial in lakes

We find evidence for substantial increases in sedimentary Nr burial in most lakes, with higher rates of Nr burial during the period 1950–2010 than 1850–1950 (Figure 2). We excluded the Nr burial after 2010 from analysis given the effect of post-depositional mineralization on the temporal variability of Nr burial. The global average Nr burial rate was 2.7 g N m^{-2} year^{-1} over the period 1950–2010, representing a 55% increase compared with 1850–1950 rates (i.e., 1.8 g N m^{-2} year^{-1}). The year 1950 demarcates the approximate time in which global anthropogenic alterations of the nitrogen cycle grew considerably via fertilizer applications to land for food production.[18] Indeed, a significant increase in Nr burial in lakes is observed after 1950, consistent with widespread use of nitrogen-based fertilizers for agriculture and fossil fuel combustion for energy, which increases Nr in deposition.[15,19]

China has used and released approximately one-third of cumulative global anthropogenic Nr over the past decade.[20] Given such dominant influence over the global nitrogen cycle, we separated China from other countries to analyze temporal changes of Nr burial in more detail. We note that the trend of increase in Nr burial since 1850 is consistent between China and most other regions worldwide (Figure 3A). In China and other global regions, Nr burial rates generally increased from 1850 to the 21st century, but with several short-lived deviations (Figure 3A) linked to changes in human activities (Figure 3B). In particular, we find evidence for a significant increase in Nr burial rates around 1910, a pivotal time point that maps with the advent of Haber-Bosch nitrogen fixation (HBNF) in 1908. This innovation substantially increased access to cheap synthetic fertilizer, which boosted food production and anthropogenic Nr input to the aquatic environment.[21]

Similar trends of change in Nr burial rate are observed across all global regions, albeit at different magnitudes (Figure 3A). The highest Nr burial rate in Oceania might due to its warming and dry climate,[22] leading to a concentrated Nr content in surface water that further contributes to the high Nr burial rate in lakes. Other high Nr burial rates are found in South America, Asia, and North America, all of which have both high air temperature and intensive Nr use in agriculture and industries.[7] The lowest Nr burial rates are found in Europe (low average air temperature) and Africa (little Nr used in agriculture and industries). However, due to the limited data in the southern hemisphere, we may overestimate or underestimate the Nr burial rate in these regions,

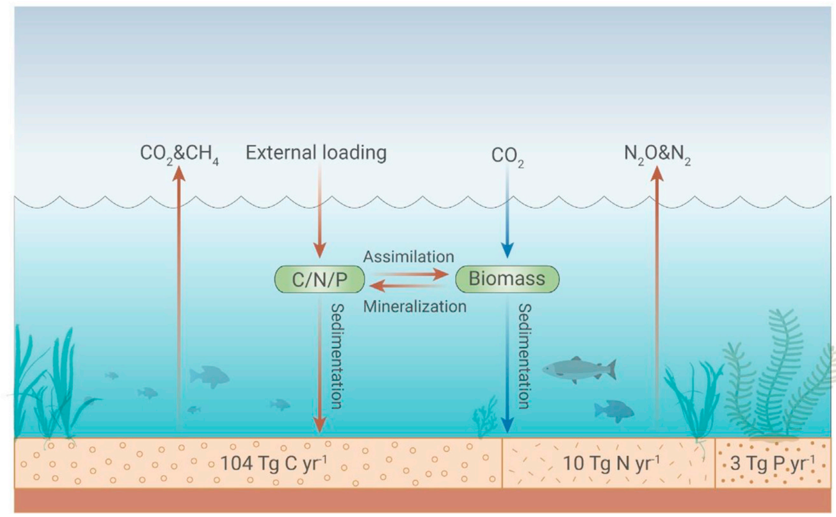

Figure 1. A conceptual model of nitrogen, carbon, and phosphorus burial in lakes. Orange arrows represent nutrient related processes; blue arrows represent biomass related processes. Numbers at the bottom represent the accumulation amount per year during 2000-2010.

especially Oceania (Figure 2). More sampling data from these regions would refine the estimation in the future.

The consequent increase of Nr burial was interrupted during the World War II (Figure 3A), when production and export of synthetic fertilizer was hampered and had to compete with industrial HBNF for production of ammunition. The use of explosives releases more inert nitrogen gas (N_2) than Nr, which reduced the Nr concentrations in the environment,[23] leading to decline of Nr burial in lakes. After World War II, nitrogen fertilizer use boomed, dramatically increasing global Nr burial rates in lake sediments.[24] The global Nr burial, as extrapolated from the set of 303 lakes, increased from 4.5 ± 0.5 (mean ± standard error) Tg N year^{-1} during 1850–1860 to 9.6 ± 1.1 Tg N year^{-1} during 2000–2010 (Tg = 10^{12} g), with a progressively increasing trend overall (Figure 3B). This finding demonstrates that global Nr burial rates are accelerating, and are mainly derived from the increase in sediment mass burial rate rather than the changes of nitrogen concentration in the sediment. This suggests that nitrogen burial in lakes may be mainly derived from organic matter, which has relatively consistent nitrogen concentration and varies slightly over time (Figure S2).

Changing ratios of carbon to nitrogen and phosphorus

To understand the processes of Nr burial in lake sediment, we also included carbon and phosphorus burial in our analysis. Consistent with coherent social-economic-environmental controls over temporal variations in Nr burial in lake sediments, we observe nonlinear trends of carbon burial rates, from 44.5 ± 4.1 Tg C year^{-1} during 1850–1860 to 104.2 ± 10.3 Tg C year^{-1} during 2000–2010 (Figures 3B, S5, and S6). Global carbon burial rates are significantly

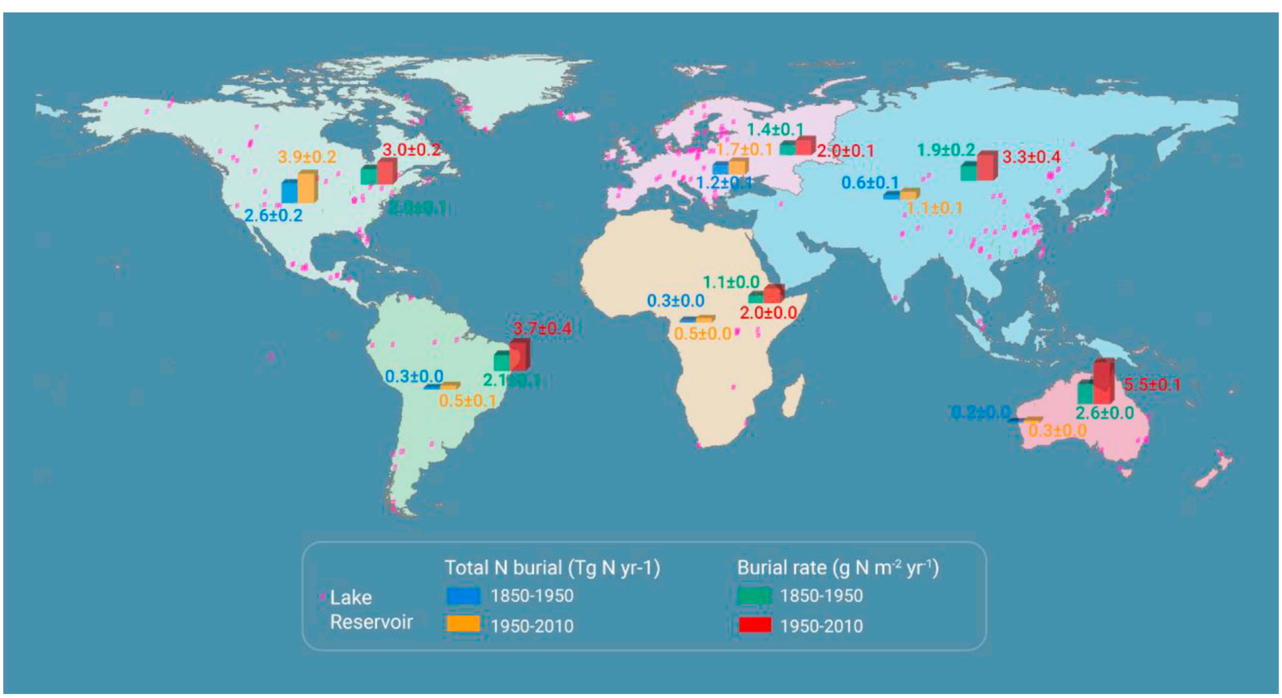

Figure 2. Distribution of sampled lakes and their Nr burial rates for 1850–1950 and 1950–2010 in different global regions Numbers ± standard error are shown for two time periods. Nr burial rate increases between these two periods. Upscaling from local to continent-level nitrogen (N) burial rate further takes continent and time consistency into consideration, based on global-level upscaling (for more details see material and methods). For the categories with insufficient samples, we then consider the sensitivity of three factors of N burial rate to be replaced. The total N burial at continent level is calculated using upscaling burial rate multiplied by the total continental lake area in each period. Finally, the average continent-level N burial rate is the ratio of the total N burial to the total lake area.

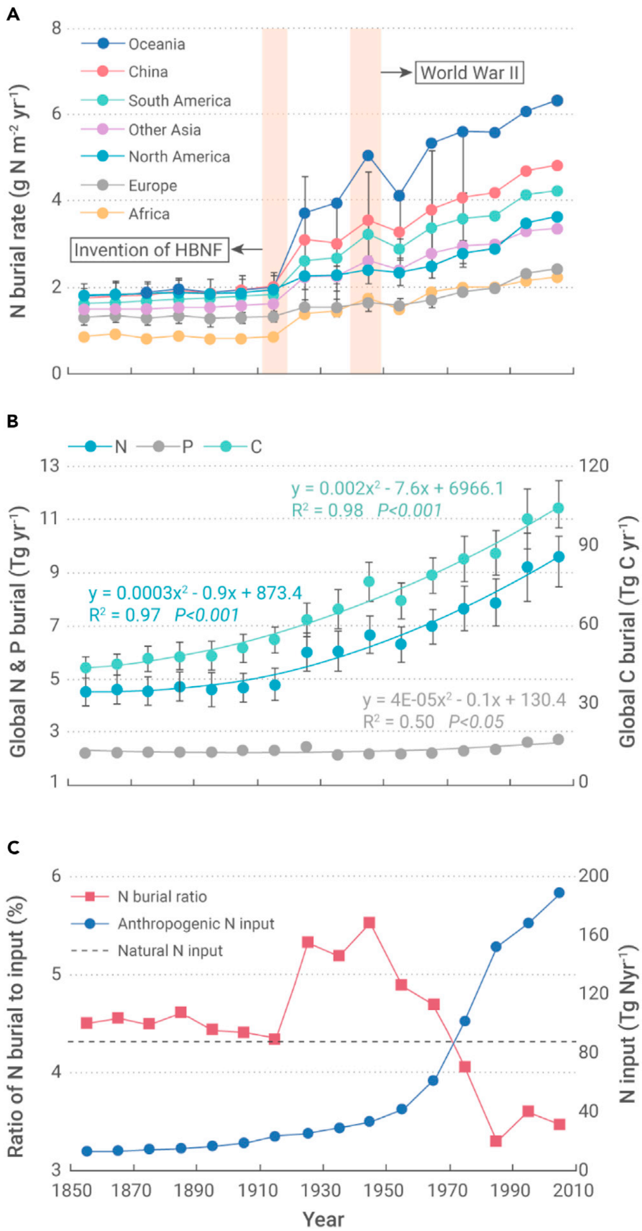

Figure 3. Changes of nitrogen, carbon, and phosphorus burial in global lakes since 1850 (A) Nitrogen burial rate in China and other global regions. (B) Total nitrogen, carbon, and phosphorus burial in global lakes. (C) Ratio of nitrogen burial in lakes to total nitrogen input to terrestrial ecosystem including both natural sources (e.g., lightning in Table 1) and anthropogenic sources (including HBNF, cultivated biological nitrogen fixation, and nitrogen oxide emission from fossil fuel combustion). HBNF refers to Haber-Bosch nitrogen fixation. The equations presented in (B) describe the temporal trends. Data source of anthropogenic nitrogen input is derived from Galloway et al.[55]

correlated with Nr burial rates temporally (Figure 4A), reflecting fundamental stoichiometric couplings between carbon and nitrogen in biomolecules.[25,26] Plankton and other autotrophs in lakes incorporate carbon and inorganic nitrogen in their biomass through photosynthesis, and this matter can be recycled and buried in lake sediments (Figure 1). Meanwhile, soil erosion can transport organic nitrogen that is integrated with carbon in organic matter, such as that derived from soil microbes. Despite these coupling processes, other confounding factors such as methane emission and nitrogen loss due to denitrification can decouple the linkage between carbon and nitrogen. Furthermore, global carbon and Nr burial rate and their total burial amount are estimated on the basis of the same suit of metrics such as lake area (see material and methods), which may bring uncertainties to our estimations. Similar to the findings of nitrogen, the carbon and phosphorus burial rates are also mainly determined by sedimentation rates rather than changes in concentration of the carbon and phosphorus in the sediment (Figure S2).

To further understand the coupling of nitrogen with carbon in lake sediments, we examined carbon and Nr burial in decadal segments using the data from all the sampled lakes and observe a substantial correlation among these constituents, which increases substantially from 1850 to 2010 (Figure S3). Although the overall correlation between carbon and nitrogen burial is strong (slope = 11.6, R^2 = 0.99, and $p < 0.001$) (Figure 4A), large variations are found on the temporal scale. This result suggested that nitrogen-carbon input and controls on burial vary across decades and lakes, with decomposition potentially affecting the relationships between carbon and nitrogen burial. This microbial process can alter the stoichiometry of carbon via nitrogen interactions (Figure 1).

In contrast, phosphorus burial rates are less synchronized with Nr burial and display overall lower correlations with carbon burial than nitrogen over time (Figures 3B and 4). Despite increases in sedimentary phosphorus accumulation rates equal to 2.2 ± 0.01 to 2.7 ± 0.05 Tg P $year^{-1}$ during 1850–2010, the growth rate of phosphorus burial did not vary coherently with nitrogen and carbon fluxes (Figure 3B). These results suggest that the dominant processes of phosphorus burial in lakes are different from those for carbon and nitrogen (Figure 1), including biological, physical, and geological processes.[1,27] Phosphorus fluxes to the sediment (burial) and back (release) to the water heavily depends on oxygen concentrations; thus, a net burial appears with oxygen-rich systems and a net release with anoxic systems. Phosphorus is also an important nutrient for the growth of plankton and other autotrophs in lakes;[1] hence, a positive correlation is found between Nr and phosphorus burial rates albeit less strongly than for carbon and Nr (Figure 4B). This could reflect preferential mineralization of phosphorus via phosphatase enzymes, which are not coupled to carbon and nitrogen.[17] Due to eutrophication, oxygen concentrations may have decreased in lakes, with the sediment releasing phosphorus instead of burying it. Furthermore, erosion and transport of mineral-bound phosphorus via precipitation runoff and wind are important pathways that differ from carbon and nitrogen delivery to lakes.[27] This inorganic pathway changes the overall coupling process of phosphorus with carbon and nitrogen through biological processes (Figure 1).

We examined C:N:P ratios to understand controls on lake sedimentary burial rates. While stoichiometry does not provide a direct assessment of the mechanism, the relative balance among C:N:P offers generalized constraints to eliminate controls and their net effect on burial fluxes, and helps us to devise working hypotheses for direct inquiry. In the case of aquatic ecosystems, the Redfield ratio (i.e., C:N:P molar ratio) of plankton biomass is normally distributed around 106:16:1,[28] such that deviations from Redfield are used to identify such mechanisms as denitrification.[29] This Redfield ratio is markedly different from the C:N:P ratio from terrestrial ecosystems (567:27:1), which normally have much higher C:N and N:P ratios.[30] Our pooled data from 1850 to 2010 exhibited a C:N:P ratio at 29:3:1 in global lake sediments (Figure 4C), suggesting an intermediate level of C:N ratio and much lower N:P ratio compared with that of terrestrial ecosystems and Redfield. It seems that point source pollution and the erosion of land are important in transporting a large amount of phosphorus to the lake sediment, leading to a much lower N:P ratio compared with both terrestrial and aquatic ecosystems. The decomposition of organic matter will remove carbon and denitrification will further convert Nr into N_2 while phosphorus is conserved, contributing to the lower N:P ratio.

Compared with the terrestrial ecosystems, the lower C:N ratio suggests a lower carbon fixation in aquatic ecosystems indicated by the Redfield ratio.[28] The C:N ratio in lake sediment is close to the C:N ratio in terrestrial soils,[31] suggesting that the erosion processes from land contribute substantially to the lake sediment. Furthermore, Nr could be lost from the sediments through poor water denitrification or anammox processes,[32] owing to lower oxygen in sediments than in overlying water. A higher C:N ratio compared with the Redfield ratio suggests that, on average, Nr is more rapidly lost from sediments than carbon, which is also supported by sedimentary burial N:P ratios that are lower than Redfield, pointing to microbial denitrification.[28] Nitrogen is

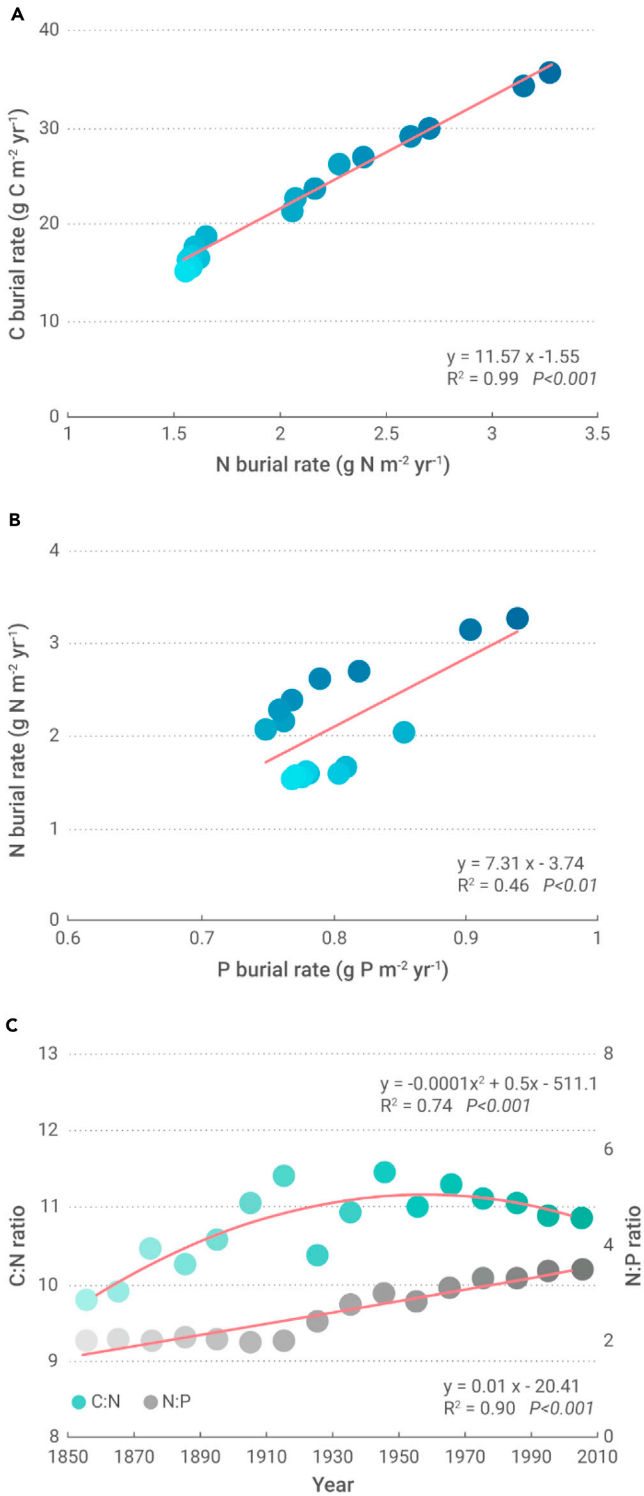

Figure 4. Stoichiometric couplings of carbon, nitrogen, and phosphorus (A) Correlation between carbon and nitrogen burial rates. (B) Correlation between nitrogen and phosphorus burial rates. (C) Changes of C:N and N:P ratios from 1850 to 2010. The year of each data point represents the middle year of a 10-year time period, with the darker color closer to the present day. The regression analyses of (A) and (B) are listed in Table S9.

mineralized 50% more rapidly than carbon in sediments during diagenesis.[33] This can be conservatively accounted for by excluding the most recent sections of sediment where diagenetic processes are likely the primary driver of changes in concentration.[34]

With the increased use of Nr for food and energy production, we found evidence for an increasing trend in C:N ratios. This suggests that the additional Nr lost from terrestrial environments to lakes promotes higher rates of carbon fixation by phytoplankton. The invention of HBNF sharply reduced C:N ratio in 1910 due to the quick increase of Nr availability up until World War II, after which the rapid increase in Nr use in agricultural and industrial sectors lowered the C:N ratio. The increasing N:P ratios from 1850 to 2010 reveal a substantial increase in human-derived Nr compared with phosphorus in lake burial (Figure 4C).

Overall, our results are consistent with previous studies (Table S11). However, our analysis of C:N:P burial rates still might be biased due to limited lake sediment ^{210}Pb data and the lack of bulk density data for a portion of lake sediment cores in our synthesis. Moreover, the estimation of limnic burial rates at regional and national scales can be biased by the limitations of upscaling based on either small datasets or variable regression relationships between controlling factors and elemental burial rates. Given the range in rates observed among lakes within the regions, the real uncertainty may be underestimated and is an important area for further work.

DISCUSSION

Human and natural driving forces

Human activities dominate the total input of Nr, and climate changes affect both biological (e.g., eutrophication) and geophysical (e.g., erosion) processes that determine nitrogen cycling.[3,35] We found a significant increase in both carbon and Nr burial rates in lakes related to global population through a Granger causality test and cointegration analysis (Table S1). Human activities directly increase the Nr input to aquatic ecosystems by enriching Nr in runoff, leaching, and deposition, which are traced to agricultural production, domestic wastewater, transportation, and industrial production.[5] This elevated nitrogen input promotes lake productivity under nitrogen-limited conditions. Therefore, we found a cointegration of carbon and nitrogen burial in lakes (Table S1), which is driven by the increase of anthropogenic Nr input to lakes.

However, Nr burial in lakes was progressively decoupled from population growth over recent decades. The strength of the trend of Nr burial with population growth decreased slightly during 1980–1990 (Figure 5A), when the majority of global Nr use occurred in developed regions such as the United States and Europe.[36] New agricultural practices and improved energy use efficiency of fossil fuel resulted in reductions of the nitrogen intensity in food and energy production in these regions.[37] With increasing use of nitrogen fertilizers in emerging economies such as China and India, which encapsulated post-1990 global nitrogen hotspots,[36,37] a sharp increase in Nr burial in lakes following the 1990s is apparent in our global analysis.

Beyond direct human effects, we identify a secondary role of changes in air temperature in determining Nr and carbon burial rates in global lake sediments (Table S1). Higher air temperature is associated with higher Nr and carbon burial rates. Increasing air temperatures can increase ecosystem productivity, facilitating carbon dioxide (CO_2) uptake from the atmosphere and Nr and phosphorus removal from the water.[38]

In contrast, while precipitation was not significantly correlated with either carbon or nitrogen burial, sedimentary phosphorus burial rates were statistically linked to precipitation (i.e., Granger causality test). Higher precipitation directly transports phosphorus to lakes, suggesting that erosion and particulate transport are more important for phosphorus than nitrogen.[39]

The missing global Nr sink

Our findings may partially explain the missing nitrogen sink in the terrestrial biosphere.[7,8] Previous studies have quantified Nr accumulation in plant biomass and soil, such as forests,[40] nitrate accumulation in ground water,[41] nitrogen transported to the ocean through atmospheric deposition and river flow,[42] and N_2O losses to the atmosphere.[8,43] However, the sum of these well-known nitrogen fates is much smaller than current estimates of Nr input to the land (Table 1). We estimate that 4.5 Tg N year^{-1} accumulated in global lakes during the pre-industrial era, accounting for 5% of total global nitrogen input (Table 1 and Figure 3C). Following marked increases in human Nr creation, total Nr burial in global lakes increased to about 9.6 Tg N year^{-1} in the

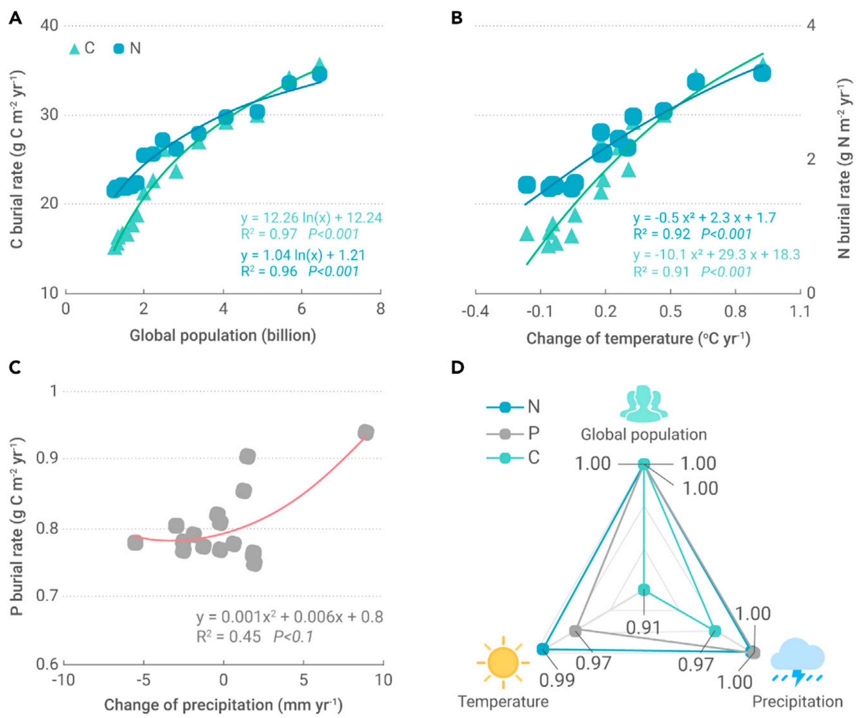

Figure 5. Correlations of carbon, nitrogen, and phosphorus burial rates in lakes with natural and anthropogenic factors
(A) Human population.
(B) Air temperature.
(C) Precipitation.
(D) Probability of burial rates driven by these factors. Each data point represents the average value of carbon, nitrogen, and phosphorus burial rates in global lakes in a 10-year time period.

2010s. Considering the contribution from natural sources, we estimate a 6.6 Tg N year^{-1} burial in global lakes derived from anthropogenic sources, accounting for 3% of total anthropogenic nitrogen input to terrestrial ecosystems. The proportion of Nr burial in lakes to total Nr input to terrestrial ecosystems increased dramatically since 1910, then declined after World War II (Figure 3C). Inclusion of global Nr burial in the Nr budget provides a relatively closed biogeochemical budget (Table 1) and resolves discussions about missing sinks. It offers a better understanding the global nitrogen cycles, especially within the context of an increasingly eutrophic world.[39]

Once Nr enters a given water body, it has the potential to be denitrified to nitrogen gases, exported downstream in dissolved or particulate form, or buried in sediments through biological processes such as uptake by algae and sedimentation (Figure 1). Nitrate in drinking water derived from surface waters can lead to the formation of *N*-nitrosamines and *N*-nitrosamides that damage (human and natural organism) DNA, threatening the safety of drinking water.[41] Nr burial in lake sediments provides an important sink through which nitrogen losses from the land are temporarily immobilized and secured from imposing risks on humans and biodiversity, fishing, and recreational use. However, this Nr burial can also be released into the water in large quantities through disturbances such as storms and other natural disasters, acting as a potential "time bomb" for future Nr release.

MATERIAL AND METHODS

Lake sites

The data from 303 lakes used in this study were derived from published sources (see Table S12 and Figures S4–S6). Studied lakes differ greatly in terms of local climate, catchment topography, size, and water depth (Table S12). These lakes are located in 57 different countries, with a latitudinal range from −53.48 N to 70.55 N, a longitudinal range from −159.17 E to 174.77 E, and an elevation range from −9 m to 4,854 m above sea level (Figure S1). The mean annual temperature ranges from −13.4°C to 28.6°C and the mean annual precipitation varies from 39 mm to 3,089 mm. Lake size ranges from less than 1 km^2 to 81,844 km^2, and the catchment area ranges from less than 1 km^2 to 99,531 km^2. Lakes also cover a wide range of water depths, with mean water depth varying from 1 m to 170 m (Table S12).

Data collection

We chose lakes based on all the following criteria: (1) having ^{210}Pb and ^{137}Cs sediment dates with 1–2 cm slice resolution and temporal scale surpassing 50 years; (2) having at least one of the three data types (dry bulk density [DBD], organic carbon concentration [TOC], and mass accumulation rate [MAR]); and (3) having total nitrogen (TN) concentration data. Among the chosen lakes, 266 and 92 lakes have total carbon (TOC) and total phosphorus (TP) concentration data, respectively (Table S12). These raw data are presented as either tables or figures in the published papers. The table data can be directly obtained, and the figure data were obtained through Getdata Graph Digitizer. Sediment accumulation rate (SAR, mm year^{-1}) was calculated on the basis of sediment layers between the dated depths of the profiles. SAR was multiplied by DBD to generate MAR; MAR was then multiplied by TN, TP, and TOC to generate nitrogen accumulation rate (NAR), phosphorus accumulation rate (PAR), and carbon accumulation rate (CAR). Direct DBD data are not available for 108 lakes (Table S12), which were generated according to the widely used empirical relationship with TOC concentration.[44]

It is well known that sediment accumulates nonuniformly in lakes.[45,46] Therefore, a single sediment core from a lake is unlikely to provide an unbiased estimate of the average mass burial rate.[47] Although multiple cores from a single lake can be used to improve the estimation of the mean lake mass burial rate, such an extremely labor-intensive approach cannot be realistic, especially for global studies. An alternative approach is to estimate the sediment focusing factor using the ratio of the mean sediment ^{210}Pb flux to the regional atmospheric ^{210}Pb flux.[47] We used the focusing factor to adjust the NAR, CAR, and PAR estimate of lakes with a single core. In total, 219 lake cores have either multiple cores or ^{210}Pb flux data for focusing correction, which are used for further analysis. Those single-core lakes without ^{210}Pb flux data for focusing correction were excluded from further analysis except for analysis of the correlation between CAR, NAR, PAR, and their ratios of C:N and N:P. The atmospheric ^{210}Pb flux is assumed to be proportional to rainfall and calculated according to the relationship of 100 Bq kg^{-1} per year per 1,000 mm of precipitation.[48] Precipitation was derived from the world climate data interpolation at 1-km spatial resolution (www.worldclim.org).

Post-deposition mineralization is another issue that needs to be addressed when estimating nitrogen, carbon, and phosphorus accumulation rates in lakes. Although sediment degradation can take place over centuries or thousands of years, the degradation rate decreases over time.[49] A study that tracked organic carbon and nitrogen over 27 years indicated that 86%–87% of sedimentary carbon and nitrogen loss occurred within the first 5 years and that the older sediments lost less than 1% of sediment mass per year.[33] To eliminate the effect of post-depositional mineralization on the temporal variability of NAR, CAR, and PAR, sediments younger than 10 years were excluded from calculations, as in earlier studies.[34,44,50]

Focusing-corrected NAR, CAR, and PAR of each lake were calculated at 10-year bins by averaging the rates for each period to study the temporal pattern. The average NAR, CAR, and PAR in different continents including Oceania, South America, North America, Europe, Africa, Asia (China and other Asia), and worldwide was used to generate the continental and global mean NAR, respectively, with the error bar (standard error) representing differences in the rates among different lakes within a

Table 1. The contribution of lake Nr burial to global nitrogen budget

	Pre-industrial	Human derived
Nitrogen input		
Biological nitrogen fixation	40–100	40–50
Lightning	2–10	0
Industrial N fixation	0	120
Fossil fuel combustion	0	30
Rock weathering	19–31	0
Totals	88	195
Nitrogen output		
River runoff to oceans	18[a]	40[c]
Atmospheric transport to the oceans	39	48[b]
Denitrification	28	63[d]
Accumulation		
Increment in biosphere	0	9[b]
Accumulation in human settlement	0	5[e]
Groundwater accumulation	0	15[b]
Nr burial in lakes	3	7
Output + accumulation	88	186
Input − (output + accumulation)	0	9

Based on the results of this study, we separated the Nr burial in lakes from the total nitrogen loss through hydrologic processes for the pre-industrial nitrogen budget derived from Vitousek et al.[6] Calculation of Nr burial in lakes in human-derived nitrogen budget was based on the total Nr burial in lakes and share of nitrogen input from anthropogenic source to total nitrogen input to terrestrial ecosystems. Units of all numbers in this table are Tg N year^{-1}.
Data sources of the pre-industrial nitrogen budget (column 2) are mainly from Vitousek et al.,[6] Houlton et al.,[54] and Fowler et al.,[5] and data sources of human-derived nitrogen input (column 3) are mainly from Fowler et al.,[5] Schlesinger,[8] and Gu et al.[23]
[a]Vitousek et al.[6]
[b]Schlesinger.[8]
[c]23% of total anthropogenic nitrogen input (Schlesinger[8]).
[d]Estimated according to nitrogen loss through river runoff currently and the ratio of denitrification to total nitrogen loss through hydrologic processes in the pre-industrial era.
[e]Gu et al.[23]

continent. The average NAR of each continent weighted by its lake area was used to generate the continental mean NAR. The NAR, CAR, and PAR of each lake was also calculated for two periods (1850–1950, 1950–2010) by averaging the rates within each period, with the error bar (standard error) representing differences in the rates among different lakes within a continent. Bootstrapping is a systematic and generally used way to evaluate estimation errors and was used to estimate the errors for NAR, CAR, and PAR at 10-year bins and different periods. Historical changes of global air temperature and precipitation are derived from Thorpe and Andrews,[51] and historical population data are from the United Nations World Population Prospect.[52]

Analysis of Nr burial rate

Each lake in our sampling dataset was identified by coordinates through the WWF HydroLAKES database (https://www.hydrosheds.org/pages/hydrolakes), and the lake characteristics (e.g., lake area, watershed area, average depth, total volume) were extracted. We used stepwise multiple linear regression to identify the three most sensitive parameters that effect Nr burial rates and divided both 219 sample lakes and 1,427,688 global lakes into groups (for nitrogen, carbon, and phosphorus burial rate, the number of groups are 216, 216, and 48, respectively). Upscaling from local to global Nr burial rate was calculated by multiplying mean sample value of Nr burial rates in each sample group and areas of global lakes and reservoirs in each group.[53] For the categories with insufficient samples, we used the mean value of a similar category as replacement. The replacement principle of similar category is determined by the sensitivity of factors. For example, the mean value of Nr burial rate under the same groups of watershed area of lakes and lake area will be replaced preferentially, because they are the two most sensitive factors of Nr burial rate. Summary statistics of these sample lakes are presented in Table S5. Numbers of sample lakes in each upscaling group are listed in Tables S7 and S8. Uncertainties are estimated on the basis of the variations of Nr burial rates in each group. More details about the estimations of burial rate of nitrogen, carbon, and phosphorus can be found in supplemental information.

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ACKNOWLEDGMENTS

This study was supported by the National Natural Science Foundation of China (41773068, 42061124001, 31901163, 32171583, and 41822701), Fund of Guangdong Provincial National Science Foundation (2018A030310518, 2019A1515011568), and General Project of Guangzhou Scientific Research Program (201904010160). This work contributes to the "Toward International Nitrogen Management System" funded by the United Nations Environment Program (GEF project ID: 5400-01142).

AUTHOR CONTRIBUTIONS

B.G. designed the study; M.W., C.R., S.W., and B.G. conducted the research; B.G. wrote the first draft; all authors contributed to the discussion and revision.

DECLARATION OF INTERESTS

The authors declare no competing interests.

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at https://doi.org/10.1016/j.xinn.2021.100158.

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