**Carbon Research**

## ORIGINAL ARTICLE

# Applying C:N ratio to assess the rationality of estimates of carbon sequestration in terrestrial ecosystems and nitrogen budgets

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## Abstract

Carbon (C) sequestration in terrestrial ecosystems needs to ensure the reactive nitrogen (Nr) supply. However, the organic C:N ratio is rarely considered in both estimates of C sequestration in terrestrial ecosystems and N budget at regional and global scale. Here we propose an approach for extracting Nr sequestrated in terrestrial ecosystems with C (termed as $N_{rc}$) from N budget and then assessing the rationality of estimates of C sequestration in terrestrial ecosystems and N budgets by using the sequestrated organic C:N ratio as a criterion. We extracted $N_{rc}$ from the N budget of IPCC AR5 at global scale and Chinese N budget (Proc Natl Acad Sci (USA) 112:8792, 2015) at regional scale based on the assumptions that there is no net Nr accumulation in agricultural products and no net Nr (except $N_2O$) accumulation in the atmosphere at annual temporal scale, and $N_2O$ is not involved in biological processes. By taking the C sequestration in terrestrial ecosystems from C budget of IPCC AR5 (2.1 Pg C/yr during 2000–2009) to assess the N budget from the same report, the organic C:N ratios calculated by the $N_{rc}$ extracted from both the upper ranges and averages of Nr input and output in global N budgets of IPCC AR5 fell outside the rational range of organic C:N ratio, but the organic C:N ratio (=31) calculated by the $N_{rc}$ (68.1 Tg N/yr) extracted from the lower ranges of the N budget fell very well into the range of organic C:N ratio in global terrestrial ecosystems. $N_{rc}$ extracted from Chinese N budget was 11.6 Tg N/yr in 2010, which could be combined with C into Chinese terrestrial ecosystems in a range between 0.14–0.66 Pg C/yr with a robust estimate of 0.36 Pg C/yr by applying the global terrestrial ecosystem sequestrated organic C:N ratio to calculation. The results indicate that the proposed approach for extracting $N_{rc}$ from N budget works well. Checking the estimated organic C:N ratio in terrestrial ecosystems will help to assess whether the C and/or N budgets are rational or not.
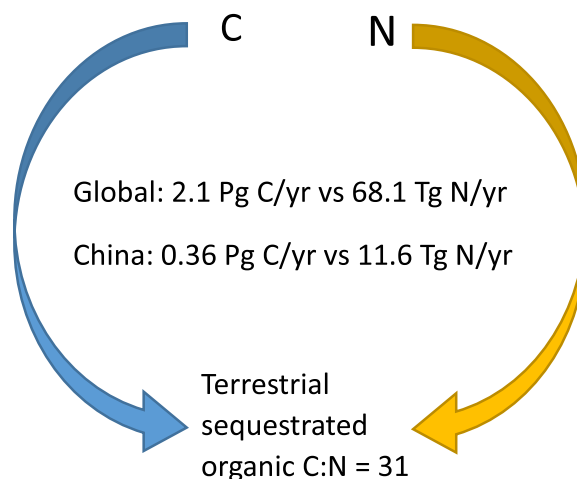
### Highlights

• An approach is proposed to extract Nr sequestrated with C in terrestrial ecosystems.

• The lower ranges of global N budget in IPCC AR5 are rational.

• C sequestration in Chinese terrestrial ecosystems is estimated to be 0.36 Pg C/yr in 2010.

**Keywords:**  Carbon sequestration, Nitrogen budget, Terrestrial ecosystems, Organic C:N ratio, Assessment

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**Graphical abstract**

C     N

Global: 2.1 Pg C/yr vs 68.1 Tg N/yr

China: 0.36 Pg C/yr vs 11.6 Tg N/yr

Terrestrial
sequestrated
organic C:N = 31

## 1 Introduction

Carbon (C) and nitrogen (N) are two elements related to energy and nutrient, respectively, which are the foundation of humankind survival and development. But C and N are also two elements which force humankind to face tremendous challenges on climate change and environmental deterioration. When we use C-based energy, e.g., fossil fuels, $CO_2$ is produced and emitted to the atmosphere, leading to the increase in atmospheric $CO_2$ concentration, driving global warming (Archer et al. 2009). When we enjoy the benefits of Haber-Bosch process fixed N, which improves crop N nutrition and increases crop yields dramatically, the surplus reactive N (Nr) disperses on the Earth surface, leading to N pollution (Galloway et al. 2008).

Terrestrial ecosystems are one of the largest organic C pools and play important roles in both sources and sinks of atmospheric $CO_2$. According to the IPCC AR5 (Ciais et al. 2013), the organic C pools in vegetation and in soil of 1 m profile are estimated to be around 600 Pg C and 1600 Pg C, respectively, being 2.65 times of the atmospheric $CO_2$ pool (*ca* 830 Pg C). The exchange rate between global terrestrial ecosystems and the atmosphere is estimated to be around 120 Pg C/yr, about 10-fold of the $CO_2$ emission from fossil fuel combustion during the period of 2000–2009. The organic C loss from terrestrial ecosystems as $CO_2$ emission to the atmosphere has made a great contribution to the elevation of atmospheric $CO_2$ concentration in human history, resulting mainly from such anthropogenic activities as deforestation and conversion of natural terrestrial ecosystems to agricultural ecosystems

(Le Quéré et al. 2013). On the other hand, the so-called "Earth greening phenomenon" was found by the long-term satellite leaf area index (LAI) records (Zhu et al. 2016; Chen et al. 2019). It is reported that the LAI in growing season increases persistently and widespreadly by over 25% to 50% of the global vegetated area during 1982–2009 (Zhu et al. 2016). The Earth greening phenomenon is mainly attributed to the climate change and continuous elevation of atmospheric $CO_2$ concentration. As the substrate of photosynthesis, atmospheric $CO_2$ concentration increased by 73.1 ppm from 1978 to 2018 (Our World in Data, https://ourworldindata.org/search?q=CO2+concentration+). Based on the review of 250 observations from 186 independent studies of 18 $C_3$ crops, spanning 14 sites and five continents, Ainsworth and Long (2021) concluded that elevation of $CO_2$ concentration by ca. 200 ppm caused a ca. 18% increase in yield under non-stress conditions. The contribution of $CO_2$ concentration elevation is estimated to contribute about 70% of Earth greenness during the period of 1982–2009 (Zhu et al. 2016). Increase in N deposition, land use change (e.g., reforestation and afforestation), and increase in multiple cropping index are also considered as drivers of Earth greening phenomenon (Zhu et al. 2016; Chen et al. 2019).

China is the country making the largest contributions to the Earth greenness and contributes 25% of the global net increase in leaf area from 2000 to 2017 (Chen et al. 2019). Being opposite to continuous decrease of forest coverage rate in the world, the forest coverage rate in China is shifted from decrease to increase in 1980s and continuously increases from 16.74% in 1990 to 23.43% in 2020 under the

government policy incentives, although the current forest coverage rate in China is still lower than the world average (31.16% in 2020) (Our World in Data, https://ourworldindata.org/search?q=forest+coverage). In terms of the forest coverage area, it increases by $6.284 \times 10^5 km^2$ from 1990 to 2020 (Li 2021) and the increment takes place across the whole country of China in the last four decades (Zhao et al. 2021). C storage in both vegetation and soils in forest increases continuously with the area increase (Xu et al. 2019; Zhao et al. 2021). The rapid decrease in rural population due to urbanization and shift of rural energy sources from biomass such as crop residues, litters, and woods to natural gas and coal directly reduces the consumption of photosynthetic products and stimulates the C sequestration in the terrestrial ecosystems as well, particularly in cropland soils (Liu et al. 2018; Shi et al. 2019; Chen et al. 2020; Zhang et al. 2022). Under the government policy incentives and strict ban of field biomass burning, the ratio of straw return to the croplands increased from less than 20% in 1980s to 53.9% in 2016 (Chen et al. 2020). Substantial increase in organic C returning into croplands and associated with the implement of good management practices results in the increases in soil organic C content and storage in croplands in China (Sun et al. 2009; Yan et al. 2011; Zhao et al. 2018). The increasing rate of organic C storage in croplands in China is estimated to be in the range from 23.7 to 25.5 Tg C/yr between 1980s and 2010s (Pan et al. 2010; He et al. 2021).

Regarding of N, since Haber-Bosch process was applied for industrial N fixation in 1913, industrial N fixation increased very rapidly in the world and reached up to around 120 Tg N/yr in 2010 (Fowler et al. 2013). Wide application of Haber-Bosch process to the production of N fertilizers has improved crop N nutrition and increased crop yield dramatically. It is estimated that 44% of the global population is sustained by Haber-Bosch process fixed N by the end of last century and the percentage rises to 48% by 2008 (Erisman et al. 2008). However, the dispersion of Nr from agricultural ecosystems to the Earth surface along with the continuous increase in N oxides (NOx) emissions from fossil fuel combustion (Fowler et al. 2013) has resulted in a series of environmental and ecological problems, which are summarized as WAGES, i.e. the problems in Water (such as eutrophication), Air (such as haze), Greenhouse gas emission ($N_2O$), Ecosystems and biodiversity (such as biodiversity loss), and Soil (such as acidification) (Sutton et al. 2011). The environmental and ecological problems caused by Nr input in China are even more serious than the world average because China now consumes about one third of world N fertilizers (Gu et al. 2015; Shi et al. 2015).

Owing to the very large spatial and temporal variations of C sequestration in terrestrial ecosystems and N dispersion rates and transformation rates, it is a great challenge to estimate C sequestration and to balance N budget at regional and global scale. Numerous effects have been made to get reliable estimates of C sequestration in terrestrial ecosystems and N budget at regional and global scale and many papers, reports and books have been published, however, there still are large uncertainties of C sequestration estimates and N budget. We know that there exist ecological stoichiometrical relationships between C, N, and P in vegetation and soil organic matters. But the ratios are rarely applied to the estimates of C sequestration and N budget. The mean ratio of organic C:N in the terrestrial ecosystems at the global scale is relative stable, although it varies with components of ecosystems and is affected by many other factors (McGroddy et al. 2004; Xu et al. 2016). As shown in Table 1, the mean of soil organic C:N ratio at global scale reported by different authors is almost the same. Although the mean of soil organic C:N ratio varies slightly among countries, if the diversities of soil types and vegetation are sufficient such as in China, the averaged organic C:N ratios in soil and in vegetation at regional scale are very close to those at the global scale (Table 1). Many evidences support that C sequestration in terrestrial ecosystems is regulated by N input, even in N-rich tropical forest (Niu et al. 2010; Lu et al. 2021). Therefore, if we could extract the net Nr accumulation, which is combined with C into organic N in terrestrial ecosystems, from N budget, we may enable to, at least coarse-scaly, assess the rationality of the estimates of C sequestration and N budget at regional and global scale by using net sequestrated organic C:N ratios in terrestrial ecosystems as a criterion. In the paper, we proposed an approach for extracting Nr sequestrated with C in terrestrial ecosystems and then assessed the estimates of C sequestration in Chinese terrestrial ecosystems and global N budgets by applying organic C:N ratios in terrestrial ecosystems as the criterion and discussed the impacts of reduction of Nr input strategies on C sequestration and possible solutions in China.

## 2 Methods and materials

Globally, the primary sources of Nr are relatively simple, including lighting produced NOx, biological fixation of $N_2$ into ammonia ($NH_3$), Haber-Bosch process fixation of $N_2$ into $NH_3$ and fossil fuel combustion emission of NOx. The Nr released from N-rock weathering may be important in preindustrial N budget (Houlton et al. 2018), but in most cases, it is not taken into current N budget. The Haber-Bosch process fixed N is mainly used as N fertilizers and applied into agricultural ecosystems. In addition to crop uptake, fertilized N to agricultural ecosystems remains; in soils, leaches to underground water; is transformed into $N_2$, and is emitted as NOx, $NH_3$, and $N_2O$ to the atmosphere. Emitted nitrogenous gases are mixed

**Table 1** Organic C:N ratios in soil and vegetation of terrestrial ecosystems

| Location | Vegetation/soil | C:N ratio | References |
|---|---|---|---|
| South America | Soil | 9.3–9.7 | Batjes 2000 |
| Eastern Canada | Temperate & boreal forest soil | 9.8–24.9 | Marty et al. 2017 |
| Australia | Soil | 11.1–16.0 | Kirkby et al. 2011 |
| Italy | Total soil | 15 | Rodeghiero et al. 2018 |
| | Organic layer | 19.3 | |
| China | Soil | 12.3 | Tian et al. 2010 |
| China | Peatland soil | 26.72 | Zhang et al. 2017 |
| Global | Humus soil | 12.0 | Lal 2008 |
| Global | Afforest organic soil | 37.32 | Xu et al. 2016 |
| | Afforest mineral soil | 12.02 | |
| Global | Soil | 12.3 | Cleveland and Liptzin 2007 |
| | Soil microbe | 7.4 | |
| China | Fine-root | 41.84 | Wang et al. 2020 |
| Italy | Litter | 29.5 | Rodeghiero et al. 2018 |
| Global | Fine-root | 40.9 | Jackson et al. 1997 |
| Global | Leaf | $37.4 \pm 3.0$ | McGroddy et al. 2004 |
| | Senesced litter | $56.7 \pm 5.4$ | |

in the atmosphere with NOx emitted from fossil fuel combustion and nitrogenous gases emitted from other sources such as livestock, transported, and deposited into oceans, inland waters, and unfertilized terrestrial ecosystems. A large proportion of deposited Nr is transformed into $N_2$ in oceans, inland waters and terrestrial ecosystems. The transformation of Nr into $N_2$ is dominated by denitrification process, although some newly found processes such as anammox (Op den Camp et al. 2007) and Feammox (Yang et al. 2020) also make certain contributions to the production of $N_2$, particularly in the wastewater treatment systems. For extracting net Nr sequestrated in terrestrial ecosystems at global scale, we need to make some assumptions as below.

1) At annual temporal scale, net Nr sequestrated in agricultural products is negligible

As same as that net C fixed in agricultural products is not taken into C sequestration in terrestrial ecosystems because agricultural products are not accumulated substantially at annual and global scale, N fixed in agricultural products is also assumed to release into inorganic N pool and there is no net Nr accumulation in agricultural products at annual and global scale. Therefore, we do not need to consider whether the produced Nr is used as fertilizer or not. But the Nr used for industrial products, if known, is excluded from the net Nr sequestrated with C into terrestrial ecosystems.

2) $N_2O$ is not involved in biological processes and not further transformed in the troposphere

$N_2O$ is a long lifetime nitrogenous gas in the atmosphere and almost not further transformed into other nitrogenous form in the troposphere (Fowler et al. 2013). As an intermediate product of denitrification, $N_2O$ could be further reduced into $N_2$ by denitrifiers in soil and water. It is reported that terrestrial ecosystems are able to use atmospheric $N_2O$, but the substantial use of atmospheric $N_2O$ in land and oceans has not been proven certainly (Jones et al. 2014). Hence, it can be assumed reasonably that $N_2O$ in the atmosphere is not involved in biological processes directly and indirectly. Based on the assumption, when we extract net Nr sequestrated with C in the terrestrial ecosystems from N budget, $N_2O$ is excluded.

3) No net Nr (except $N_2O$) accumulation occurs in the atmosphere

Nitrogenous gases except $N_2O$ are very active in the atmosphere, easily transformed and deposited into terrestrial ecosystems and oceans, and involved in biological processes there after being transformed into biological available forms (Fowler et al. 2013). Their concentrations vary temporally and spatially (e. g. Alghamdi et al. 2014), however, their concentrations at the annual temporal scale can be assumed as unchanged in the atmosphere.

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The assumption is supported by the N budget in IPCC AR5 (Ciais et al. 2013), in which the sum gaseous Nr (except $N_2O$) production (emissions from Earth surface to the atmosphere, 94.7 Tg N/yr and lighting produced, 3–5 Tg N/yr) is roughly equal to the atmospheric N deposition into oceans and terrestrial ecosystems (100 Tg N/yr) during the period of 2000–2009.

Based on the above assumptions, the net Nr combined with C into organic form and sequestrated into terrestrial ecosystems at global scale (termed as $N_{rc}$) can be extracted from N budget as follows:

$$N_{rc} = N_{rt} - N_{de} - N_2O - N_{ri} \tag{1}$$

Where $N_{rt}$ refers to the total natural and anthropogenic Nr production; $N_{de}$ refers to $N_2$ production through denitrification; $N_2O$ refers to the $N_2O$ production among the $N_{rt}$; and $N_{ri}$ refers to the Nr kept in inorganic forms in the oceans, inland water bodies and terrestrial ecosystems. We define the ratio of Nr sequestration in terrestrial ecosystems ($N_{rc}$) to the total production ($N_{rt}$) as the net terrestrial sequestration ratio of Nr production ($N_{rc}/N_{rt}$, %).

In the paper, the global $N_{rc}$ is extracted from the global N budget in IPCC AR5 (Ciais et al. 2013) and Chinese $N_{rc}$ is extracted from the Chinese N budget estimated by Gu et al. (2015). The data on C sequestration at global scale are from the same report of IPCC during the same period.

## 3 Results

### 3.1 Organic C:N ratio sequestrated in terrestrial ecosystems at global scale

IPCC AR5 provided detail data on global C and N budget during the same period of 2000 to 2009. Table 2 shows the data presented in IPCC AR5 (Ciais et al. 2013) on C sequestration and emission as $CO_2$ in the global terrestrial ecosystems during the period of 2000–2009. The global terrestrial ecosystems, including inland water bodies, play as a net sink of $CO_2$ with an average rate of 2.1 Pg C/yr, accounting for 26.9% of $CO_2$ emissions from fossil fuel combustion during the corresponding period. Oceans also play as a net sink with the rate of 1.6 Pg C/yr during the same period (Ciais et al. 2013).

Table 3 shows the average and range of Nr production and denitrification $N_2$ production from IPCC AR5 (Ciais et al. 2013). The Nr production and denitrification of Nr into $N_2$ are estimated to be 436 and 409 Tg N/yr with a range of 387–511 and 301–518 Tg N/yr during the period of 2000–2009, respectively. In the same period, $N_2O$ emission is estimated to be 17.9 Tg N/yr, which cannot be combined with C into organic N. Since IPCC AR5 does not provide the data on inorganic N accumulation in terrestrial ecosystems, we extracted $N_{rc}$ with the assumption

**Table 3** Global reactivated and deactivated nitrogen budget during the period of 2000–2009 (Data cited from IPCC AR5, et al. 2013)*

| Input | Mean, Tg N/yr | Lower range, Tg N/yr | Upper range, Tg N/yr |
|---|---|---|---|
| Industrial fixation | 124 | 117 | 126 |
| Cultivated fixation | 60 | 50 | 70 |
| Fossil fuel combustion | 30 | 27 | 33 |
| Lighting | 4 | 3 | 5 |
| NBF[a] in land | 58 | 50 | 100 |
| NBF in oceans | 160 | 140 | 177 |
| **Total** | **436** | **387** | **511** |
| Output | | | |
| Denitrification in oceans | 300 | 200 | 400 |
| Denitrification in land | 109 | 101 | 118 |
| **Total** | **409** | **301** | **518** |
| Net accumulation | 27 | 86 | −9 |
| $N_2O$ emission | 17.9 | | |
| **Terrestrial sequestrated $N_{rc}$** | **9.1** | **68.1** | **−26.9** |

[a] NBF denotes nitrogen biological fixation

**Table 2** Atmospheric $CO_2$ sources and sinks of global terrestrial ecosystems during the period from 2000 to 2009 (data cited from IPCC AR5, Ciais et al. 2013)

| Carbon sources | | Flux Pg C/yr | Carbon sinks[a] | | Flux Pg C/yr |
|---|---|---|---|---|---|
| Natural | Total respiration and fire | 107.2 | Natural | Gross photosynthesis | − 108.9 |
| | Freshwater out gassing | 1 | | | |
| Anthropogenic | Land use change | 1.1 | Anthropogenic | Photosynthesis strengthened by human activities | −14.1 |
| | Respiration strengthened by human activities | 11.6 | | | |
| Total | | 120.9 | | | − 123 |
| Net effects | | −2.1 | | | |

[a] Sink is marked with minus sign

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that $N_{ri}$ in Eq. (1) is zero at global scale during the period. Based on the data available and assumption that the Nr (except $N_2O$) is not accumulated in the atmosphere, we extracted $N_{rc}$ from the lower ranges, averages, and upper ranges of N budget in Table 3, respectively.

1) $N_{rc}$ from the upper range of global N budget

Because the upper range of Nr production (511 Tg N/yr) is smaller than the upper range of denitrification $N_2$ production (518 Tg N/yr) (Table 3), extracted $N_{rc}$ is negative and the magnitude is $-26.9$ Tg N/yr during the period of 2000–2009. The negative $N_{rc}$ implies that a net Nr loss occurs from terrestrial ecosystems during the period of 2000–2009. If it was true, the global terrestrial ecosystems were a net source rather than a net sink of atmospheric $CO_2$.

2) $N_{rc}$ from the averages of global N budget and sequestrated organic C:N ratio

The average of global Nr production and denitrification $N_2$ production is 436 and 409 Tg N/yr, respectively, during the period of 2000–2009 (Table 3). On the average, the net Nr accumulation is 27 Tg N/yr. After subtracting $N_2O$ emissions of 17.9 Tg N/yr, the $N_{rc}$ is 9.1 Tg N/yr and the net terrestrial sequestration ratio of Nr production is 2.1%. The extracted $N_{rc}$ of 9.1 Tg N/yr is combined with 2.1 Pg C/yr sequestrated into global terrestrial ecosystems and the organic C:N ratio is about 231 during the period of 2000–2009. This ratio is much larger than those in soil and in vegetation (Table 1) and implies that either the C sequestration is overestimated or the $N_{rc}$ is underestimated or both.
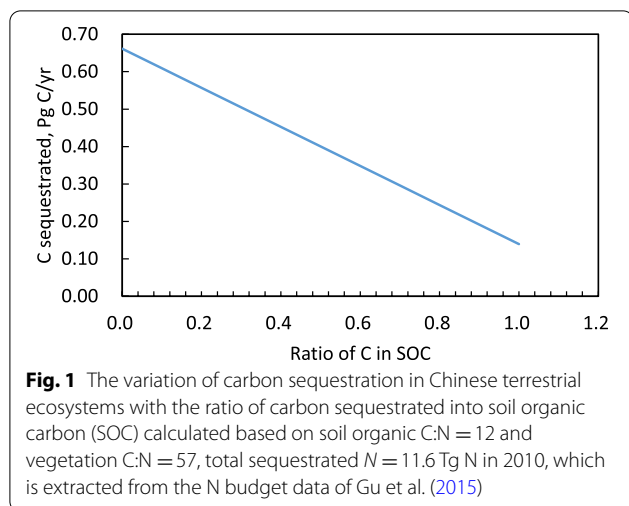
3) $N_{rc}$ from the lower ranges of global N budget and sequestrated organic C:N ratio

If we take both lower ranges of Nr production (387 Tg N/yr) and denitrification $N_2$ production (301 Tg N/yr) in global N budget (Table 3) to extract $N_{rc}$, the net Nr accumulation is 86 Tg N/yr during the period of 2000–2009. Subtracting the emissions of $N_2O$ (17.9 Tg N/yr) from the net Nr accumulation and assuming that $N_{ri}$ is zero, the $N_{rc}$ is 68.1 Tg N/yr during 2000–2009. The net terrestrial sequestration ratio of Nr production is 17.6% and the organic C:N ratio of 68.1 Tg N/yr combined with 2.1 Pg C/yr sequestrated in the global terrestrial ecosystems is 31. The C:N ratio drops very well into the range of organic C:N ratio of soil and vegetation in the terrestrial ecosystems (Table 1).

## 3.2 Nrc and C sequestration capacity in China

According to the estimate of Gu et al. (2015), the natural biological N fixation is 7.1 Tg N/yr, cultivated biological N fixation is 4.6 Tg N/yr, Haber-Bosch process N fixation for synthetic fertilizer use is 32.0 Tg N/yr, and for industrial N use 5.1 Tg N/yr, NOx emission from fossil fuel combustion is 6.6 Tg N/yr, and imported food, feeds, and other agricultural products bring 5.7 Tg N/yr into China in 2010. Thus, the total Nr production reaches up to 61.1 Tg N/yr. In the same year of 2010, the N output includes the denitrification $N_2$ production (24.3 Tg N/yr), exported by air (8.7 Tg N/yr) and by water to oceans (5.5 Tg N/yr). As the consequence, the net of 22.6 Tg N/yr is accumulated in Chinese territory in 2010, accounting for 26.3% of the global net Nr accumulation (86 Tg N/yr) calculated from the lower ranges of N budget in IPCC AR5. Based on the above available data, we need to modify Eq. (1) slightly to extract $N_{rc}$. In Gu's estimate, the emissions of $N_2O$ (around 1.4 Tg/yr) is embedded in the Nr exported by air. The Nr produced by Haber-Bosch process for producing industrial products (5.1 Tg N/yr) is not involved in C sequestration in terrestrial ecosystems. Nr accumulated in underground water (5.9 Tg N/yr), dominated by $NO_3^-$ (Zhang et al. 1996), is not combined with C in terrestrial ecosystems and considered as a part of $N_{ri}$. It has been observed intensively that because of excessive application of N fertilizers, a large amount of nitrate is formed and accumulated in cropland soil profiles, particularly in the soils under plastic-shed cultivation and in the north China where there is no sufficient water transporting nitrate into underground water (Zhou et al. 2016). However, there are no estimates available on $NO_3^-$ accumulation in soil profiles of croplands in China. If we ignore the inorganic N accumulation in cropland soils, and take Nr accumulation in groundwater (5.9 Tg N/yr) alone as $N_{ri}$ in Eq. (1), the $N_{rc}$ is 11.6 Tg N/yr in 2010 in China. The value of $N_{rc}$ could be regarded as the maximum of Nr sequestrated into organic N in Chinese terrestrial ecosystems in 2010 because part of it might exist as inorganic N in soil profile. Compared to the world average estimated from the lower ranges of global N budget, the net terrestrial sequestration ratio of Nr production in China is 19.0%, slightly larger than the global average (17.6%).

Taking 12:1 as soil organic C:N ratio in China and averaged organic C:N ratio (57:1) in senesced litter as C sequestrated in vegetation (Table 1), and based on the $N_{rc}$ extracted from N budget estimated by Gu et al. (2015), the extreme range of C sequestration in Chinese terrestrial ecosystems varies from 0.14 Pg C/yr if all the C is sequestrated into soil organic C (SOC) to 0.66 Pg C/yr if all the C is sequestrated into vegetation (Fig. 1).

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**Fig. 1** The variation of carbon sequestration in Chinese terrestrial ecosystems with the ratio of carbon sequestrated into soil organic carbon (SOC) calculated based on soil organic C:N = 12 and vegetation C:N = 57, total sequestrated $N$ = 11.6 Tg N in 2010, which is extracted from the N budget data of Gu et al. (2015)

## 4 Discussion

The averaged organic C:N ratios in soil and vegetation vary within very narrow ranges as shown in Table 1 at regional scale such as China with very high diversity of terrestrial ecosystems and at global scale. Therefore, the organic C:N ratio calculated by the estimated C and N sequestration in the terrestrial ecosystems at regional such as China and at the global scale could be used as at least a coarse-scale criterion to assess the rationality of these estimates.

### 4.1 Assessment of global N budget in IPCC AR5

The $CO_2$ sources are roughly classified into fossil fuel combustion and land use change, and the sinks include C sequestration in terrestrial ecosystems and oceans and $CO_2$ accumulation in the atmosphere (e.g., Friedlingstein et al. 2020). The C budget is closure at the global scale, meaning that the total sources are equal to the total sinks, although the so-called "missing carbon sinks" might still exist (Houghton 1995). Among $CO_2$ sources and sinks, the uncertainties of fossil fuel source and atmospheric accumulation are relatively small. There are large uncertainties of $CO_2$ fluxes between oceans and atmosphere and between land and atmosphere, however, they are constrained by the global C budget (Houghton 2020). In addition, the advanced techniques, such as eddy covariance technique (Baldocchi 2020), are widely applied to the direct measurements of net C fluxes between land and atmosphere, which can be used to verify the estimate of C sequestration in terrestrial ecosystems. Therefore, the variations of C sequestration in global terrestrial ecosystems estimated by different research teams are relatively small (Ciais et al. 2013; Friedlingstein et al. 2020; Houghton 2020). However, N budget is still open at both regional and global scale. It means that the total

input is not equal to the total output (Table 3). Therefore, although the uncertainties of Haber-Bosch process fixed N and NOx emission from fossil fuel combustion are comparable to those of $CO_2$ fossil fuel sources, the uncertainties of other sources and sinks are not constrained by the balance of Nr input and output. Furthermore, there are still no methods available for direct measurements of biological N fixation and denitrification in field, which are dominant contributors of Nr input and output, respectively (Table 3). Based on these considerations, we took estimated C sequestration in global terrestrial ecosystems to assess global N budget by using the criterion of sequestrated organic C:N ratios in terrestrial ecosystems.

There are many estimates available on C sequestration in global terrestrial ecosystems in literature, for instance, the series estimates firstly published in Earth System Science Data (Le Quéré et al. 2013). But there are very few estimates available made by the same research groups on both C and N budgets during the same period in literature. IPCC AR5 (Ciais et al. 2013) presented the global C budget and global N budget during the same period of 2000–2009. The net C sequestration in global terrestrial ecosystems of 2.1 Pg C/yr during 2000–2009 (Table 2) in IPCC AR5 is comparable to those reported in literature (Le Quéré et al. 2013). Thus, we took the net C sequestration of 2.1 Pg C/yr to respectively assess the rationality of the upper ranges, averages and lower ranges of N budget in IPCC AR5 (Table 3). When we extracted $N_{rc}$ from the upper ranges of Nr production and denitrification $N_2$ production of N budget in IPCC AR5, it was negative, indicating that a net Nr loss takes place in the terrestrial ecosystems. The result was completely contradictory to the common view that global terrestrial ecosystems have played a net sink of atmospheric $CO_2$ in the last four decades (e.g., Chen et al. 2019, Friedlingstein et al. 2020, Houghton 2020). Therefore, the upper ranges of Nr production and denitrification $N_2$ production of N budget in IPCC AR5 are not rational and can be excluded completely.

When we extracted $N_{rc}$ from the averages of Nr production and denitrification $N_2$ production in N budget in IPCC AR5, we got a $N_{rc}$ equal to 9.1 Tg N/yr during the period of 2000–2009 at the global scale, even smaller than that of 11.6 Tg N/yr in 2010 in China. We cannot exclude the possibility completely because Nr input in the world varies spatially and greatly and both too much and too less coexist in the world (Galloway et al. 2008). Net Nr losses take place in the areas where N fertilizers are rarely applied and/or deforestation happens intensively. However, the sequestrated organic C:N ratio of 133 (2.1 Pg C/yr vs 9.1 Tg N/yr) can not support the rationality of averages of N budget during the period of 2000–2009.

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The $N_{rc}$ is 68.1 Tg N/yr extracted from the lower ranges of Nr production and dentirification $N_2$ production of N budget in IPCC AR5. And the organic C:N ratio, sequestrated with 2.1 Pg C/yr, is 31 at global scale during the period of 2000–2009. The ratio falls very well into the organic C:N ratio (Table 1) and can be regarded as an average of organic C:N ratio in global terrestrial ecosystems. The results indicate that the lower ranges of biological N fixation and denitrification in Table 3 are more likely to be rational than those of upper ranges and averages of N budget in IPCC AR5. In deed, in the N budget of Fowler et al. (2013), biological N fixation in oceans (140 Tg N/yr) and in land (58 Tg N/yr), and denitrification $N_2$ in oceans (100–280 Tg N/yr) and in land (100 Tg N/yr) are much close to the corresponding lower ranges of global N budget in IPCC AR5, respectively (Table 3). The extracted $N_{rc}$ of 68.1 Tg N/yr during 2000–2009 is much larger than the estimate by Zaehle (2013), who estimated that the terrestrial ecosystems sequestered 27 Tg N/yr and 1.9 Pg C/yr during 2001–2010. The organic C:N ratio calculated from his estimates is 70, which is larger than the maximum ratio in Table 1 and the average of 17, calculated from his estimates of total C and N pools in global terrestrial ecosystems. His estimate of C sequestration is very close to the estimate in IPCC AR5 (2.1 Pg C/yr), thus, his Nr sequestration (27 Tg N/yr) is more likely to be underestimated. The $N_{rc}$ extracted from the lower ranges of N budget in IPCC AR5 during 2000–2009 is comparable to the estimate by Galloway et al. (2004) that terrestrial ecosystems sequestrates 60 Tg N/yr in 1990s. Considering that the global N fertilizers consumption increases by 14.2% from 1990s to 2000s (data from Our World in Data, https://ourworldindata.org/), the increase in $N_{rc}$ by 13.5% (8.1 Tg N/yr) is a reasonable estimate during the corresponding period.

There are several uncertainty sources for extracting $N_{rc}$ to assess N budget in IPCC AR5. Firstly, the distribution of $N_{rc}$ in oceans and in land is not identified by the proposed approach. The $N_{rc}$ extracted by the proposed approach consists of the net Nr sequestrated both in oceans and in terrestrial ecosystems. It is reported the N deposition also promotes C sequestration in the oceans (Duce, et al. 2008) and inland waters (Liu et al. 2021). Voss et al. (2013) estimated that the burial of Nr in oceans was about 22 Tg N/yr, mainly as organic N form. If the Nr buried in oceans was all sequestrated into organic forms in terrestrial ecosystems and then delivered to ocean, the value of $N_{rc}$ would not be altered. However, if part or all of the Nr buried in oceans was sequestrated directly in oceans, the $N_{rc}$ consisted of the Nr sequestrated with C in terrestrial ecosystems and in oceans. For instance, if all the buried Nr 22 Tg N/yr estimated by Voss et al.

(2013) was sequestrated in oceans directly as an integral part of the net ocean C sink of 1.6 Pg C/yr estimated by IPCC AR5 (Ciais et al. 2013), the Nr sequestrated with 2.1 Pg C/yr in terrestrial ecosystems would actually be 46.1 Tg N/yr, rather than 68.1 Tg N/yr extracted from the lower ranges of N budget in IPCC AR5. In such case, the sequestrated organic C:N ratio was 46 in the terrestrial ecosystems, but it was still within the range of C:N ratio in Table 1.

Secondly, the accumulation of $NO_3^-$ in surface water and groundwater happens commonly where N fertilizers are excessively applied (Singh et al. 2022). The Nr accumulation in Chinese groundwater alone is estimated to reach up to 5.9 Tg N/yr in 2010 (Gu et al. 2015). Therefore, the real $N_{rc}$ combined with C into terrestrial ecosystems N would be less than that extracted from the global N budget of IPCC AR5 assuming that $N_{ri} = 0$ and the sequestrated organic C:N ratio would be overestimated. For instance, after subtracting the Nr accumulated in groundwater in China from the $N_{rc}$ extracted from the lower ranges of N budget in IPCC AR5, the organic C:N ratio is 34. But the C:N ratio still falls very well into the range of global averaged organic C:N ratio in the terrestrial ecosystems (Table 1).

Thirdly, there are at least two Nr sources not taken into current global N budget in IPCC AR5. They are the N releases accompanied with $CO_2$ emissions from land use change and from N-rock weathering, respectively. There are no estimates available in literature on the net N released from land use change. If assuming that the C:N ratio of decomposed organics inductively by land use change was the same of net sequestrated organic C:N ratio of 31 as calculated above, Nr release accompanied with the $CO_2$ emission (1.1 Pg C/yr, Table 2) from land use change was estimated to be 35.5 Tg N/yr. The N release from N-rock weathering was estimated to be between 19 and 31 Tg N/yr (Houlton et al. 2018). If the net Nr terrestrial sequestration ratio of all the Nr released from N-rock and land use change was the same of the Nr from other sources (17.6% calculated based on the lower ranges of N budget), the $N_{rc}$ from land use change and N-rock weathering was between 9.6–11.7 Tg N/yr. Plus the $N_{rc}$ extracted from the averages of N budget in IPCC AR5, the total $N_{rc}$ was between 18.7–20.8 Tg N/yr and the organic C:N ratio sequestrated with 2.1 Pg C/yr in terrestrial ecosystems was between 112 and 101. The range of organic C:N ratios are still much larger than the global averaged organic C:N ratios in the terrestrial ecosystems (Table 1). For the $N_{rc}$ extracted from the lower ranges of N budget, the total $N_{rc}$ was between 77.7–79.8 Tg N/yr and the organic C:N ratio sequestrated with 2.1 Pg C/yr in terrestrial ecosystems was about 27–26, which

still fell into the range of C:N ratios in the global terrestrial ecosystems (Table 1). Taking all these uncertainty sources together into account, in the cases that 22 Tg N/yr was all sequestrated in oceans directly, 5.9 Tg N/yr was kept in inorganic forms, and 9.6–11.7 Tg N/yr was from the land use change and N-rock weathering, the $N_{rc}$ extracted from the lower ranges of N budget in IPCC AR5 was modified into 49.8–51.9 Tg N/yr and the C:N ratio sequestrated with 2.1 Pg C/yr varied between 42 and 40, falling into the C:N ratios in the global terrestrial ecosystems. Therefore, the known uncertainty sources are unlikely to overturn the conclusion that the lower ranges of N budget in IPCC AR5 is rational.

## 4.2 Possible range of C sequestration in Chinese terrestrial ecosystems

Much different from the global scale, the uncertainties of estimated C sequestration in terrestrial ecosystems are unconstrained by C budget at regional scale. In fact, there are no common accepted constraint conditions available for estimating C sequestration in terrestrial ecosystems at regional scale. Without constraint conditions, the estimates could be made more arbitrarily. Although it is a fact that Chinese terrestrial ecosystems play as a net sink of atmospheric $CO_2$ in the last four decades (Sun et al. 2009; Jiang et al. 2016; Zhu et al. 2016; Chen et al. 2019; Zhao et al. 2021), the C sequestration in Chinese terrestrial ecosystems estimated by applying various approaches varies greatly. Wang et al. (2020) compiled some previous estimates in literature and their own work and showed that the estimates of C sequestration in Chinese terrestrial ecosystems varied from 0.179 Pg C/yr to 1.25 Pg C/yr, being about 7 times difference between the smallest and the largest estimates. In order to reach a consensus on the estimate at regional scale, to figure out the constraint conditions that can be used to assess regional estimate is essential. Organic C:N ratios may be a useful criterion to assess the rationality of C and N sequestration in terrestrial ecosystems at regional scale, such as China, if N budget is available.

The $N_{rc}$ extracted from the N budget estimated by Gu et al. (2015) is 11.6 Tg N/yr and the net terrestrial sequestration ratio of Nr production is 19.0% in 2010, slightly larger than the global average (17.6%). It seems reasonable, if considering that the forest coverage rate increases rapidly and the soils for afforestation is dominated by those of N deficient in China. Therefore, the $N_{rc}$ value of 11.6 Tg N/yr could be regarded as the maximum able to be sequestrated with C into terrestrial ecosystems in China since net accumulated Nr kept in inorganic forms in surface water and soils is ignored when extracting $N_{rc}$. Within the reasonable range of organic C:N ratio in the

terrestrial ecosystems, the C sequestration with 11.6 Tg N/yr in Chinese terrestrial ecosystems would not exceed 0.66 Pg C/yr in 2010. If the ratio of global terrestrial ecosystems sequestrated organic C:N $= 31$ is applicable to that of Chinese terrestrial ecosystems, the C sequestration in Chinese terrestrial ecosystems is robustly estimated to be 0.36 Pg C/yr in 2010. Jiang et al. (2016) reported that the C sequestration in Chinese terrestrial ecosystems was estimated to be 0.39–0.51 Pg C/yr with a mean of 0.46 Pg C/yr in 2006–2009 by applying top-down approach and 0.29 Pg C/yr by applying bottom-up approach. The difference between these two estimates is attributed to the delivery of C sequestrated in terrestrial ecosystems to oceans and burial in aquatic sediments. Because the Nr exported outside Chinese territory by water is already taken into account when extracting $N_{rc}$ in China in this work, the estimated 0.36 Pg C/yr includes those sequestrated inland and delivered to inland waters, but does not include those C delivered into oceans. Thus, the C sequestration of 0.36 Pg C/yr estimated from the $N_{rc}$ of 11.6 Tg N/yr and global averaged C:N ratio seems closely consistent with the estimates by Jiang et al. (2016).

It is worth to notice the effects of Nr exported outside Chinese territory by air and water on C sequestration. According to the estimate by Gu et al. (2015), the total Nr of 14.2 Tg N/yr was exported outside Chinese territory in 2010. Exported Nr can also be combined with C into organic N and transformed into $N_2$ outside Chinese territory. By applying the net terrestrial sequestration ratio at global scale (17.6%) to the calculation, the $N_{rc}$ from exported Nr is estimated to be 2.5 Tg N/yr, which is combined with C into organic N outside Chinese territory. Taking the Nr exported outside Chinese territory into together, the total $N_{rc}$ is estimated to be 14.1 Tg N/yr (11.6 + 2.5 Tg N/yr) in 2010 in China and accounts for 20.7% of the world $N_{rc}$ of 68.1 Tg N/yr during the period of 2000–2009.

## 4.3 Impacts of reducing Nr input on C sequestration in Chinese terrestrial ecosystems

Much different from that N deposition contributes 9% of Earth greenness at global scale (Zhu et al. 2016), the contribution of N deposition to the increased C storage in Chinese terrestrial ecosystems is much larger. Tian et al. (2011) estimated that the contribution of N deposition and fertilizer applications together accounted for 61% of the net C storage increment in China's land ecosystems. The increase in atmospheric N deposition from 13.2 kg N/ha in the 1980s to 21.1 kg N/ha in the 2000s was synchronous with the increase in forest coverage rate from 1980s to 2000s (Liu et al. 2013, 2020). Since the soils for afforestation in China are dominated by those of N deficient,

the synchronous increases in N deposition relieved the N stress for plant growth in the early stage of afforestation and made the N deposition contributing C sequestration larger than the world average. It is estimated that the N deposition results in a net C sink of 62 Tg C/yr in the terrestrial ecosystems and 6.51 Pg C in total during the period of 1901–2005 in China (Lu et al. 2012).

However, rapid increase in Nr production has resulted in N pollution seriously (Liu et al. 2020). For controlling N pollution, the national strategies for reducing N production and application were implemented in 2015. The consumption of N fertilizers began to decrease after reaching the peak in 2015 (Fig. 2). With the enforcement of the regulations for controlling N consumption and reducing NOx emissions from fossil fuel combustion, atmospheric N deposition is stabilized after 2010 and expected to be further reduced (Yu et al. 2019; Liu et al. 2020). Since N deposition can even stimulate C sequestration in used recognized N-rich tropical forest (Lu et al. 2021), reduction of Nr input could be expected to increase the degree of N deficiency, and then constrain the C sequestration in Chinese terrestrial ecosystems. The stimulative effects of atmospheric $CO_2$ concentration on vegetation growth would be constrained by N deficiency either.

C sequestration in terrestrial ecosystems is supposed to make a substantial contribution to the achievement of "carbon neutralization" in 2060 in China (Wang et al. 2020). Therefore, it is essential to develop new techniques and management for enhancing N use efficiency. In addition, there are several possible solutions that may be able to relieve the negative impacts of reducing Nr input on C sequestration in terrestrial ecosystems in China.

1) Introduction of leguminous plants. It may be special important at the early stages of afforestation in degraded soils in which organic N contents are low.
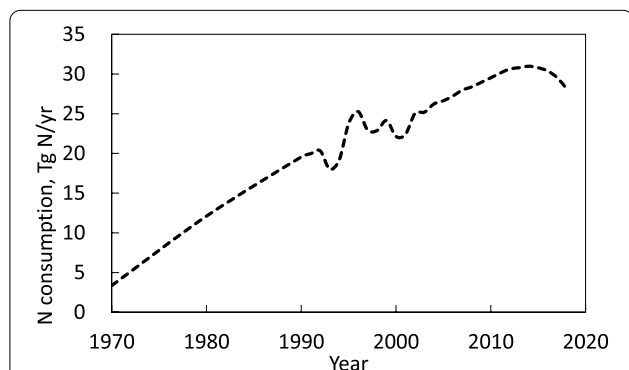
2) N fertilization to N deficient terrestrial ecosystems. The exogenous Nr is dominantly from N deposition, where N is from fossil fuel combustion, N fertilized croplands, and livestock, and transported to the unfertilized ecosystems. For the N deficient soils used for afforestation, N fertilization will stimulate tree growth and promote C sequestration in terrestrial ecosystems.

3) Prevention of excessive N application leading to $NO_3^-$ accumulation in soil and in groundwater. The Nr accumulation rate estimated by Gu et al. (2015) reached up to 5.9 Tg N/yr in groundwater alone in 2010. $NO_3^-$ accumulation occurs in soil profile as well (Zhou et al. 2016). If $NO_3^-$ accumulation in soil and water could be prevented significantly through rational fertilization and the N saved could be applied to the forestlands, the negative impacts of reducing Nr input on C sequestration would be avoided substantially.

## 5 Conclusions

The proposed approach for extracting $N_{rc}$ from N budget works well. Judged with organic C:N ratio at global scale, the upper ranges of Nr production and denitrification $N_2$ production in the N budget of IPCC AR5 is overestimated definitely. The averages of the N budget in the report are more likely to be overestimated as well even if taking Nr releases from land use change and N-rock weathering into account. By taking the lower ranges of the budget, 68.1 Tg N/yr is combined with 2.1 Pg C/yr during 2000–2009. As a consequence, the sequestrated organic C:N ratio of 31 falls very well into the range of organic C:N ratio in the global terrestrial ecosystems. Therefore, the lower ranges of N budget in IPCC AR5 is much more rational than the upper ranges and averages.

The net Nr sequestrated ($N_{rc}$) in Chinese terrestrial ecosystems, extracted from the N budget of Gu et al. (2015), is 11.6 Tg N/yr in 2010. Synchronous increases in Nr input and forest coverage rate makes a great contribution to the C sequestration in Chinese terrestrial ecosystems in the last four decades. However, from the view of net Nr accumulation, the C sequestration rate does not exceed 0.66 Pg C/yr and the robust estimate is 0.36 Pg C/yr in 2010 by applying globally averaged sequestrated organic C:N ratio of 31. With the reduction of Nr input, the Nr available for C sequestration in terrestrial ecosystems is expected to decrease accordingly. Development of novel techniques and management is essential for keeping C sequestration rate in Chinese terrestrial ecosystems through raising Nr use efficiency and preventing $NO_3^-$ accumulation in water and soil.

**Fig. 2** Temporal variations of N fertilizer consumption in China from 1970s to 2018s. Data cited from FAOSTAT

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## Availability of data and materials
Not applicable.

## Declarations

### Ethics approval and consent to participate
Not applicable.

### Consent for publication
All authors read and approved the manuscript to be published in Carbon Research.

### Competing interests
No conflict of interest. The authors of Z.C. Cai and X. Y. Yan are the Editorial Board Members of Carbon Research.

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## Publisher's Note