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Cleaning up nitrogen pollution may reduce future carbon sinks

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Cleaning up nitrogen pollution may reduce future carbon sinks

Abstract

Biosphere carbon sinks are crucial for reducing atmospheric carbon dioxide (CO_2) concentration to mitigate global warming, but are substantially affected by the input of reactive nitrogen (N_r). Although the effects of anthropogenic CO_2 emission and nitrogen deposition (indicated by N_r emission to atmosphere) on carbon sink have been studied, it is unclear how their ratio (C/N) changes with economic development and how such change alters biosphere carbon sinks. Here, by compiling datasets for 132 countries we find that the C/N ratio continued to increase despite anthropogenic CO_2 and N_r emissions to atmosphere both showing an asymmetric para-curve with economic growth. The inflection points of CO_2 and N_r emissions are found at around $15,000 gross domestic product per capita worldwide. Economic growth promotes the use of N_r and energy, while at the same time increases their use efficiencies, together resulting in occurrences of inflection points of CO_2 and N_r emissions. N_r emissions increase slower but decrease faster than that of CO_2 emissions before and after the inflection point, respectively. It implies that there will be relatively more anthropogenic CO_2 emission but less N deposition with economic growth. This may limit biosphere carbon sink because of relative shortage of Nr. This finding should be integrated/included in global climate change modelling. Efforts, such as matching N deposition with carbon sequestration on regional scale, to manage CO_2 and N_r emissions comprehensively to maintain a balance are critical.

Key words: Carbon sink, CO_2 emission, Climate change, Economic development, Nitrogen deposition, Stoichiometry

1. Introduction

Emission of carbon dioxide (CO_2) from human activities to the atmosphere is the most important driver of global warming, and the increase of atmospheric CO_2 concentration is responsible for ~64% of the radiative forcing from well-mixed greenhouse gases (Thompson et al., 2016). Hence, stabilizing and ultimately reducing the atmospheric CO_2 concentrations is one of the principal mechanisms to mitigate anthropogenic climate change. Over half of current global anthropogenic CO_2 emissions are taken up by terrestrial ecosystems (30%) and oceans (25%), and the rest is accumulating in the atmosphere (Reay et al., 2008). Increasing CO_2 absorption in both terrestrial ecosystems and oceans is therefore a critical topic for the United Nations Framework Convention on Climate Change (UNFCCC) to reduce CO_2 accumulation in the atmosphere. External forces, such as human disturbances and climate change, could create a disequilibrium to alter global carbon (C) cycle (Luo and Weng, 2011). For instance, elevated CO_2 concentrations can increase the C sink of natural ecosystems in many Free-Air CO_2 Enrichment (FACE) experiments (Drake et al., 2011; Talhelm et al., 2014). However, these C sinks are often limited by the availability of nutrients such as nitrogen (N) and water (Hungate et al., 2003; Luo and Weng, 2011). Recent research showed that the rate of CO_2 uptake in Amazonian rainforests have decreased due to deficiency in nutrients such as N (Corlett, 2014). This suggests that the relative abundance of N input to ecosystems compared to atmospheric CO_2 concentration is crucial for the future increase in biosphere C sinks.

Economic drivers are crucial for determining future trends in CO_2 emissions and reactive N (N_r) use/loss (Chow and Li, 2014; Zhang et al., 2015; Li et al., 2016). For example, emissions of CO_2 and N oxides (NO_x) are closely related to fossil fuel combustion driven by economic development (Fig. S1). To meet human demand for food and energy, over 100 Tg N yr^{-1} (mainly ammonia (NH_3) and NO_x) have been emitted to the atmosphere in 2010, and about 70% of the N_r emitted is deposited on land surface, with the remainder deposited onto oceans (Fowler et al., 2013). This large amount of man-made CO_2 and N_r emissions substantially alters global C and N biogeochemical cycles through changing the C/N ratios (CO_2 to N_r emission) on multiple scales (Erisman et al., 2011; Erisman et al., 2013). Elevated levels of N deposition have been found to increase C sinks in many terrestrial and oceanic ecosystems because of the removal of N shortage, as shown up in a reduced C/N ratio (Reay et al., 2008). Besides deposition, the input N can also be transferred to aquatic ecosystems through runoff from agriculture and human settlements, etc., which might increase the C sink in aquatic ecosystems (Erisman et al., 2011). However, these increased N_r fluxes have exceeded the “safe operating space” for global societal development, adversely affecting human health, ecosystems and the environment (Erisman et al., 2013; Steffen et al., 2015). N_r pollution is estimated to cost €70–320 billion per year in the European Union (Sutton et al., 2011), and $81–441 billion per year in the United States (Sobota et al., 2015). Activities that improve N use efficiency

(NUE), i.e. to produce more food and energy with less N_r loss to the environment, have been proposed and implemented in many regions; these activities may increase the C/N ratio and affect C sinks in the biosphere (Chen et al., 2014; Lassaletta et al., 2014).

Environmental Kuznets Curves (EKC) have been widely applied to identify relationships between economic development and anthropogenic CO_2 emission, as well as N_r pollution, especially N_r loss from cropland (Chow and Li, 2014; Zhang et al., 2015; Li et al., 2016). However, little attention has been paid to changes in C and N stoichiometry of emissions to the atmosphere and its relevance for the global C sink, i.e., how climate change will be affected by changes in the C/N ratio of emissions under future economic development. Here, we analyzed global spatio-temporal changes in anthropogenic N_r inputs and losses, and CO_2 emissions as a result of economic development, using a panel data model for 132 countries from 1961 to 2008 (Fig. 1). Using these long-term data, we attempted to understand how C/N ratios change with economic development, predict their effect on terrestrial C sink capacity preliminarily and analyze the socioeconomic mechanisms behind the changes of the C/N ratio. In this paper, the C/N ratios refer to the ratios of anthropogenic emissions of CO_2 and N_r to atmosphere, including NH_3 and NO_x that is related to the N deposition to land surfaces. We put these in context with N_r losses and NUE in food and energy production, as well as related environmental issues.

2. Methods

2.1. Data sources

We compiled annual data on population and urbanization levels for 132 nations for the period of 1961–2008 from the FAOSTAT database (FAO, 2016) and GDP (gross domestic product, expressed in real 1990 international dollars, using purchasing power parity, PPP) from the Total Economy Database (GGDC, 2008). Data on cropland area, weighted yield of up to 275 crop types, and N and P fertilizer use were compiled from the FAOSTAT database. Per-capita fertilizer use and per-area fertilizer use were calculated as the annual N fertilizer use of a nation divided by the total population and cropland area, respectively. Cultivated biological N fixation (CBNF) was calculated for each nation based on the area of crop legumes, pasture and fodder legumes, and rice with N fixation rates of 115, 168 and 33 kg N ha^{-1} yr^{-1}, respectively (Herridge et al., 2008).

Per-capita fossil fuel energy consumption values and CO_2 emission from fossil fuel use combustion and cement production in the nations from 1961 to 2008 were obtained from the World Development Indicators dataset of the World Bank (http://databank.worldbank.org/). CO_2 emission per energy consumed (CO_2E) was calculated from CO_2 divided by the total fossil energy consumption of each nation. Total NO_x emissions from fossil fuel combustion and NH_3 emissions for the nations from 1970 to 2008 were collected from the Emission Database for Global Atmospheric Research (EDGAR, 2016). NO_x emission per energy consumed (NO_xE) was calculated from NO_x divided by the total fossil energy consumption of each nation.

2.2. Inflection point in a country

A piecewise linear regression approach, which has been widely used in many previous studies (e.g., (Piao et al., 2011)), was applied to CO_2 emission or Nr use/loss on per-capita GDP series for each nation from 1961 to 2008.

$$\mathrm{y} = \begin{cases} \beta_0 + \beta_1 x + \varepsilon, & x \le \alpha \\ \beta_0 + \beta_1 x + \beta_2 (x - \alpha) + \varepsilon, & x > \alpha \end{cases}$$

where x is per-capita GDP; y is CO_2 emission or Nr use/loss; α is the inflection point of per-capita GDP; and β_0, β_1, and β_2 are regression coefficients; ε is the residual of the fit. The CO_2 emission or Nr use/loss trend is β_1 before the inflection point, and β_1+ β_2 after it. All coefficients were determined by least-squares linear regression. We also confined α to within the period 1965 to 2004 (1974 to 2004 for NO_x) to avoid a linear regression in one period having too few data points. A probability level of $P<0.05$ was considered significant. To check whether there could be more than one inflection point, we plotted the data from each country into a line chart to make sure that the inflection point detected is corrected.

2.3. Panel cointegration analysis

We applied recently developed panel unit root and panel cointegration techniques (Li et al., 2016) to estimate the cointegration relationships between economic growth and C/N ratios (emission of CO_2 to the emission of NH_3-N+NO_x-N) (Table S1 & S2). We performed panel cointegration analysis as follows: *Step1*, we computed the summary panel unit root test on the levels of the series, along with a summary of the results, using individual fixed effects or both fixed effects and trends as regressors, and automatic lag difference term and bandwidth selection (using the Schwarz criterion for the lag differences, and the Newey-West method and the Bartlett kernel for the bandwidth). Most of the results indicated the presence of a unit root, since the Levin–Lin–Chu (LLC), Breitung, Im–Pesaran–Shin (IPS), and Augmented Dickey–Fuller–Fisher (ADF–Fisher) tests reject the null of a unit root. *Step2*, performing panel cointegration tests to determine whether per capita N_r use and loss, and their climatic effects had a cointegration relationship with socioeconomic development. We chose the deterministic trend specification according to the type of exogenous regressors and used the Schwarz criterion for the lag differences, and the Newey-West method and the Bartlett kernel for the bandwidth. Fisher/Johansen cointegration tests showed that the long-term cointegration relationship exists. *Step3*, Running the cointegration regression. After the panel unit root test and cointegration tests, we regressed per capita N_r use and loss, and their climatic effects on socioeconomic development.

2.4. Panel data model

Considering multiple interactions, we built a panel model to quantify the effect of per-capita GDP on N input, and N_r and CO_2 emissions. Classic empirical studies on EKC have been criticized because of concerns regarding statistical analyses of time series data that may be non-stationary. Therefore, we examined the stationarity of our data and used the ADF–Fisher test, which is the most frequently used method for the co-integration test in EKC empirical studies (SI text, Table S1 & S2). The panel model compiles data on both the temporal and spatial scales simultaneously (48 years for 132 nations in this study, totaling 6,336 samples), also known as 'time-series cross-sectional data'. The panel model can solve unobservable time-invariant regional differences and omitted variable problems. We tested the EKC for CO_2, N input/emission on the global scale based on the pool data of 132 countries from 1961 to 2008 by using a panel model with the test of stationarity to eliminate spurious regression results. The panel model used in this study was constructed as follows:

$$Y_{it} = c + PGDP_{it}\beta_1 + Dummy\beta_2 + PGDP_{it} \times Dummy\beta_3 + \sum_j Ctrl_{itj}\beta_j + \mu_i + \varepsilon_{it}$$

where Y_{it} is the N input, and N_r and CO_2 emission in year t in country i; $PGDP_{it}$ is the annual per-capita GDP; $Dummy$ is a binary parameter (0, 1) introduced to test whether the inflection points of N_r fluxes and CO_2 emission exist with growth of per-capita GDP. For N Fertilizer, the group dummy is 0 for per-capita GDP <\$14,000 and 1 for GDP per capita ≥\$14,000; for NO_x, NH_3, and CO_2, the critical per-capita GDP is

\$15,000 for the group dummy. $Ctrl_{itj}$ is a group of control variables, including population, urbanization, etc., which may affect the N input, and N_r and CO_2 emission; $\beta_1, \beta_2 ... \beta_j$ are the coefficients of these influencing factors; c is the intercept; the effect of per-capita GDP on the N input, and N_r and CO_2 emission, calculated as $\partial Y/\partial pl = \beta_1$; μ_i, is the unobservable individual effect in country *i* such as the time invariant geographical situation; and ε_{it} is random error. Contemporaneous correlation, heteroskedasticity and serial correlation are controlled to calculate asymptotically efficient parameters with a Prais-Winsten regression in Stata12 (Wooldridge, 2010).

2.5. Simulation of C sink changes

The *Integrated BIosphere Simulator* (IBIS) is used to simulate the C sink changes in 2050 globally (Lu et al., 2016). The basic parameterization of climate, including air temperature, precipitation, CO_2 concentration, etc. follows storyline IPCC B1, under which the air temperature increase is assumed to remain below 2°C and CO_2 concentrations lower than 500 ppm in 2050 (Stocker et al., 2013). We included the effect of managing N_r into the simulation, considering a relatively lower N deposition with economic development in 2050. Considering the economic growth to 2050, a 20-50% relative reduction of N deposition compared to that of CO_2 emission is assumed to estimate the impact on the C sink, i.e., the net ecosystem productivity (NEP) changes.

In fact, economic growth has complex effects on both the CO_2 and N_r emissions and their impacts on the C sink on regional and global scale. Meanwhile, the spatial distribution of N deposition and N-saturation issue also affect the C sink. Therefore, to simplify the estimation we just assumed a 20-50% lower N deposition input to the IBIS model in this paper. Future work is needed to investigate the potential of C sink changes, and quantify these uncertainties in the context of changes in C sink capacity derived from C/N ratio.

3. Results

3.1. Changes of C/N ratio and their effect on C sink

C/N ratios of total emissions per capita significantly increase with the growth of per-capita GDP without an inflection point across all countries (Fig. 2, Table 1). Each 1% increase in per-capita GDP resulted in a 0.47% increase of the C/N ratio of emissions. This suggests that the relative availability of N to terrestrial ecosystems and oceans through N deposition will be reduced with economic development, relative to the increase in anthropogenic CO_2 emissions. We estimate that global C sinks will be reduced by 5-10% under the IPCC B1 emissions scenario, when a 20-50% relative reduction of N deposition compared to that of CO_2 emission is assumed (Fig. 3). This reduction of C sink capacity mainly occurs in sub-tropical areas in Eastern Asia and North America, where the hotspots of N_r emissions are in close proximity to natural forest ecosystems. This reduction is subject to large uncertainties due to variations in both temporal and spatial scales.

3.2. Driving forces of C/N ratio change on global scale

To further assess the driving forces of C/N ratio changes, we analyzed how specifically CO_2 emission, N_r emission and their sources were affected by economic development. Generally, per-capita anthropogenic CO_2 emissions significantly increased with the growth of per-capita GDP before reaching an inflection point with an increasing rate of 0.57% per 1% increase of per-capita GDP (Table 1). Energy consumption increased with per-capita GDP, but leveled off when per-capita GDP reaches around \$20,000 (Fig. 4). CO_2E declined when per-capita GDP increased beyond \$10,000, which suggests that energy use efficiency increases with economic development (Fig. 4). The changes in energy consumption and CO_2E with the growth of per-capita GDP resulted in an inflection point for CO_2 emissions at around \$15,000. Nevertheless, CO_2 emissions did not decline drastically after the inflection point but remained relatively stable (Fig. 2).

An inflection point was observed for per-capita NO_x emissions from fossil fuel combustion in relation to the growth of per-capita GDP at around \$15,000 (Fig. 2). Before the inflection point, a 0.31% increase of NO_x emission per 1% increase of per-capita GDP was found; while after the inflection point, a 0.99% decrease of NO_x emission per 1% increase of per-capita GDP was found. Economic growth appeared to have a larger effect on the reduction of NO_x emissions after the inflection points, when compared to that of CO_2 emission (Table 1). Economic development significantly increased energy use, but reduced NO_xE once per-capita GDP reached approximately \$10,000 (Fig. 4).

There was no significant effect of per-capita GDP on NH_3 emission due to substantial variations in NH_3 emissions across countries while per-capita GDP was lower than \$5,000 (Table 1, Fig. 2). Although not significant, the inflection point test was still positive, and the potential inflection point of NH_3 emission was found with a per-capita GDP around \$15,000 (Fig. 2d). Similar to that of NO_x, the rate of increase in NH_3 before the inflection point was lower while the rate of decrease after the inflection point was higher compared to that of CO_2 emission (Table 1).

An asymmetric para-curve relationship of N fertilizer use with economic development was found at the global scale (Table 1). The inflection point of N fertilizer use was observed at a value of per-capita GDP of around \$14,000 (SI text, Fig. 5), lower than the per-capita GDP for the inflection of NH_3 emission (around \$15,000) (Fig. 7). This suggests that it is harder to reduce NH_3 emission than N fertilizer use with economic development. Substantial variations were seen in the maximum level of per-capita and per-area N fertilizer use in different countries (Fig. 2). Countries with a large population density and PGDP (e.g., the Netherlands and Denmark) typically have intensive food production systems, high food production per land area and strict regulations on fertilizer use and Nr emission abatement (Table 1).

Globally, cultivated biological N fixation (CBNF) played a subordinate role in food production compared to N_r input from mineral fertilizers during the period 1961-2008 (Fig. 4). No inflection points were observed for CBNF with the growth of per-capita GDP (Table 1). Instead, per-capita CBNF decreased with the increase in per-capita GDP, although the ranges of per-capita CBNF varied widely among countries (Fig. 2b).

3.3. Analysis of inflection points on national scale

To further understand the underlying mechanism of the changes in C/N ratios with economic development, we analyzed the inflection points of N_r use/loss on national scale. We classified the countries into two categories: Type 1 and Type 2, based on whether there is an inflection point. Generally, Type 1 countries were the relatively rich/developed, and Type 2 countries the relatively poor/developing (Fig. 8, Fig. 5; see SI for the list of countries). The inflection points of N fertilizer use in Type 1 countries occurred at an average per-capita GDP of \$14,200 ± 800 for the period 1973 to 2003 (average in 1986, Fig. 5). In contrast, the average per-capita GDP of Type 2 countries was only \$4,300 in 2006-2008, far below the per-capita GDP identified as an inflection point at the global scale. For Type 1 countries, the per-capita N fertilizer use decreased by 39% while the crop yield and PFP_N (partial factor productivity of N fertilizer = kg grain yield per kg N fertilizer input) increased by 27% and 53%, respectively, from the year when the inflection point occurred relative to the reference period 2006-2008 (Fig.

5). In contrast, the per-capita N fertilizer use and yield were still increasing in Type 2 countries without an inflection point, but both significantly lower than those of Type 1 countries, except that PFP_N was higher than that in Type 1 countries. Higher PFP_N in Type 2 countries suggests low N input/supply and thereby also low crop yield. Per-capita GDP was positively related to crop yield (Fig. 4). The PFP_N followed a U-shaped pattern in relation to growth in per-capita GDP and increased substantially beyond the inflection point (Fig. 4).

Similar to per capita N fertilizer use, per area N fertilizer use of Type 1 countries declined from 170 to 115 kg N ha^{-1} yr^{-1} from the year when inflection occurred to 2006-2008, but was still much higher than the average value of 82 kg N ha^{-1} yr^{-1} in 2006-2008 for Type 2 countries (Fig. 6). Nevertheless, among the Type 2 countries, six countries (Chile, China, Colombia, Costa Rica, Egypt, and Malaysia) used more than 200-400 kg N ha^{-1} yr^{-1}. These six countries share no common natural conditions, geographic location or climatic conditions, but a significant share of the agricultural area is cultivated intensively for domestic consumption or export. Also, these countries had a per-capita GDP similar to the inflection per-capita GDP of the Type 1 countries, except for Egypt. Through a cluster analysis we found that these six countries were separated from Type 2 countries to form a new group of Type 3 countries based on the average value of per-capita GDP, N fertilizer use per area and crop yield from 2006 to 2008. Type 3 countries had a significantly higher per area N fertilizer use, lower PFP_N and intermediate per-capita GDP compared to the other two groups of countries (Fig. 6). It suggests that, in the near future, the inflection points of N fertilizer use are more likely to occur in Type 3 countries than in the other Type 2 countries.

At national scale, inflection points of NO_x emissions were found in 24 countries (Type 1 countries, see SI for the list) with an average per-capita GDP of \$15,500 ± 500 from 1973 to 1999 (average in 1988, Fig. 5). Although the NO_x emissions of Type 1 countries decreased from 13.4 to 8.6 kg N $capita^{-1}$ yr^{-1} from the year of the inflection point to 2006-2008, they remained much higher than the NO_x emissions of Type 2 countries (3.6 kg N $capita^{-1}$ yr^{-1}). From the year of the inflection point to 2006-2008, per-capita energy consumption increased by 7% while NO_xE decreased by 38% in Type 1 countries, as their per-capita GDP increased by 47% (Fig. 5). This indicates a decoupling of NO_x emissions and energy consumption.

Furthermore, we found inflection points of NH_3 emissions and phosphorus (P) fertilizer use in 20 countries, the majority of which also had inflection points for N fertilizer use. The inflection year was indeed late for NH_3 emissions but early for P fertilizer use compared to that of N fertilizer use (Fig. 7). Accordingly, the inflection point per-capita GDP was larger for NH_3 emissions but smaller for P fertilizer use compared to that of N fertilizer use.

4. Discussion

4.1. C/N ratios of emissions

Natural terrestrial ecosystems such as forests, grasslands, wetlands and oceans are important C sinks (Ciais et al., 2010; Quéré et al., 2013). Nitrogen deposition plays a critical role in increasing C sequestration in natural ecosystems (Reay et al., 2008; Luo and Weng, 2011). Previous studies have suggested an over 2-fold increase in N deposition by the end of this century using either IPCC projections or the RCP approach (Ciais et al., 2013; Winiwarter et al., 2013). However, some models used by the IPCC and in some other studies have been criticized for their lack of constraint on terrestrial N balances (Houlton et al., 2015). This study provides an alternative perspective to understand the effects of N_r input on global C cycles through the relationships between economic development and CO_2 and N_r emissions and the C/N ratios of the emissions.

The use of EKC to analyze the relationships between CO_2 and N_r emissions has been tested in several studies (Zhang et al., 2015; Li et al., 2016). Our findings concur with previous findings suggesting that the EKCs of CO_2 and N_r emissions indeed exist (Fig. 1). This indicates that continuing economic growth after inflection points can reduce CO_2 emissions and N_r pollution through socioeconomic changes, such as better management and increased NUE. However, the reduction of N_r emissions after the inflection point is much larger than that of CO_2. Therefore, if economic growth increases beyond about $15,000, the C/N ratio of the emissions rapidly grows. In comparison to managing N_r uses and losses, the reduction of CO_2 emissions seems to be far more difficult as noted in our study. Emissions are tightly coupled with the energy supply methods and associated with economic growth (Liu et al., 2015). Reducing CO_2E appears to be quite difficult unless large scale energy saving is introduced, or clean and renewable energy technologies such as solar, wind and hydropower generation are adopted widely (Liu et al., 2015). Therefore, per-capita CO_2 emissions may not decrease significantly with economic development, unless energy efficiency increases, or sustainable energy sources replace fossil fuels to a large extent. This indicates that the per-unit-N_r emission will accompany a higher CO_2 emission with economic development in both rich and poor countries. The lifetime of N_r in the atmosphere is in the order of days to weeks and the majority of N_r emitted to the atmosphere will deposit on the land surface (Fowler et al., 2013; Liu et al., 2013), while the rest will end up in the oceans (Kim et al., 2014). The lifetime of CO_2, however, is in the order of years to decades (Solomon et al., 2009). Therefore, the strong cumulative effect of CO_2 compared to that of N_r in the atmosphere and biosphere will further increase C/N ratios and thereby may affect C sink capacities on a global scale.

Although the overall change in global C sink as function of N deposition depends on economic development, large variations across global regions occur, with hotspots of C sinks changing on both temporal and spatial scales. From the 1970s to the 1990s, inflection points for N uses and losses were found in Type 1 countries, mainly located in the Europe and North America, where the level of N deposition was the highest (Galloway et al., 2008; Townsend and Howarth, 2010). Significant C sinks were mostly located in Type 1 countries, probably because of the elevated N deposition as well as land use and land cover changes occurred during the last century (Luyssaert et al., 2010; Erisman et al., 2011; Pinder et al., 2012). After inflection points have been reached, N uses and losses were reduced by 20-40% in Type 1 countries (Fig. 5), which decreased C sinks in natural terrestrial systems in these regions recently (Piao et al., 2011). The hotspots of N deposition and C sinks have more recently switched from Type 1 countries to Type 2 countries in East and South Asia (Galloway et al., 2008; Reay et al., 2008), such as China, which appears to be close to reaching an inflection point on N_r use/losses (Fig. 8). Meanwhile, we can still identify many other Type 2 countries which would further increase their N_r uses and losses before reaching an inflection point, mainly in Africa and tropical regions, where soil N availability currently limits agricultural yields. Moreover, mining of soil N occurs in many low-input agricultural systems in Africa (Vitousek et al., 2009; Sutton et al., 2013). Our findings relate also to the discussions about the “4 per 1000” (4p1000) initiative, launched at the COP21 conference in Paris (Van Groenigen et al., 2017). Nevertheless, we foresee future hotspots of N deposition to emerge, once Asian countries have passed their inflection points (Fig. 8).

4.2. Nitrogen related analysis

Many uncertainties and confounding factors still need to be addressed to further understand the effects of C/N ratios, because terrestrial C sinks show rather complex responses to N availability. At sites with excessive N_r input, e.g. croplands and adjacent natural vegetation, the reduction of N_r input may in fact increase net primary productivity (NPP) and C sinks (Lu et al., 2016). This mainly occurs due to N saturation in ecosystems such as forests reducing plant growth and at times resulting in forest death with high N deposition rates (Sutton et al., 2011). This has been observed in Europe and Northeastern United States in areas suffering from high N deposition (Sutton et al., 2011). Similar effects have been noted in some regions in China and India with the highest N deposition rates currently (Lu et al., 2016). At the same time, the reduction of NO_x and NH_3 emissions may have positive impacts on C sinks, because NO_x and NH_3 are precursors for tropospheric ozone and particle matters (PM) pollution, which reduces plant productivity (Erisman et al., 2011).

The reduction of N input and losses can also reduce the emission of N_2O, which is the third most important greenhouse gas (GHG), although no significant correlation or inflection point of N_2O emissions with economic development was detected in our analyses (Fig. S2). Compared to N_r inputs, N_2O emission processes are more complex, with multiple emission sources affected by substrate availability and natural factors, such as soil redox potential and microbial processes (Davidson and Kanter, 2014; Sutton et al., 2014). This complexity is apparent from the large variations in N_2O emissions at per-capita GDP lower than \$5,000 (Fig. S2). Nevertheless, inflection points of N_2O emissions were still found in many countries at a per-capita GDP of around \$15,000. Under current levels of N_r uses and losses, the climatic effect of N_r is balanced with both warming and cooling effects (Erisman et al., 2011; Pinder et al., 2012). However, with further economic development, the reduction of N_r availability could limit the growth of C sinks. Reducing N_r input would be expected to reduce N_2O emissions, but at least part of the climate benefit could be reduced by the negative impact on C sinks. This may shift the climate balance of N_r, reducing CO_2 sequestration potential, thereby offsetting part of the climate change benefit of reducing N_r. The overall effects of the EKC of N input/loss and the increasing trend of C/N ratios with economic development on climate change are complex, and further research is warranted.

Besides N losses through N_r emission to the air, a substantial proportion of N input is lost to water systems (Galloway et al., 2008). Although N_r released to water bodies may also increase C sinks in aquatic ecosystems such as wetlands or coastal ecosystems due to N_r-emission fueled primary production in aquatic systems, the majority of this input N_r is denitrified (Schlesinger, 2009; Zhao et al., 2015). Once other forms of N_r are converted to nitrate, the denitrification process will likely reduce nitrate to N_2 or N_2O (Zhao et al., 2015). However, owing to the substantial variations of denitrification on spatiotemporal scales (Kulkarni et al., 2008), the effect of N leaching to water systems on the C sink has not, to our knowledge, been quantified.

Next to N losses and inputs to air and water bodies, N input via N fertilizer and CBNF to cropland may also have impact on C sink. However, different with natural ecosystems, croplands are not typically regarded as major C sinks, but are rather regarded to be C neutral (Ciais et al., 2010; Quéré et al., 2013), depending also on crop rotation, soil cultivation and crop residue return to soil. Input of N into cropland commonly increases crop yield and crop residue production, and thereby may enhance C sequestration (Sun et al., 2010). However, excessive N input to cropland results in nitrate accumulation rather than C sequestration (Zhou et al., 2016). Residual nitrate in cropland soils may in part by used by the next crop, but often leaches to groundwater, and thus does not affect the capacity of C sinks (Zhou et al., 2016). Meanwhile, the EKC of N fertilizer use suggests that the N input to croplands can be reduced, while yield will increase, with economic development. This could reduce N accumulation in soils as well, and is consistent with recent findings on the EKCs of N surplus in cropland (Zhang et al., 2015).

4.3. Policy implications

Integrated management of N and C is essential for sustainable development, environmental quality and climate change mitigation in the future (Maione et al., 2016). Firstly, the development and adoption of clean energy supply systems and improvement in energy use efficiency via advanced technologies and management is crucial to the reduction of both CO_2 and N_r emissions to the atmosphere. We found that it is easier/faster to reduce N_r emission than CO_2 emission, after the inflection point. A developed economy can benefit from the implementation of emission control technologies, such as selective catalytic reduction (SCR), to reduce the NO_x emissions (Walters et al., 2015). It is also true for the reduction of NH_3 emission through emission abatement technology, improved agricultural practices and management (Sutton et al., 2011; Van Grinsven et al., 2013). It appears more difficult to reduce CO_2 emissions, although improvements in fossil energy use efficiency, energy savings, and increased use of renewable energy help greatly (Liu et al., 2015). Increased energy use efficiency and energy savings can also benefit the reduction of NO_x and NH_3 emissions (Gu et al., 2015). Therefore, aligning technologies and policies related to clean energy supply and improving energy use efficiency is crucial (Omri, 2013; Sutton et al., 2014). This can reduce both CO_2 and N_r emissions, benefit the balance of C/N ratio of these emissions, and thereby maximize C sink capacity.

Secondly, integrated management of C and N at landscape and regional scales is vital. CO_2 concentration is generally uniform at global scale with little spatial variation, but N_r emission and deposition vary a lot on at regional scale. Thus, managing the C sink should also be at regional scale to maximize the use N_r emission and deposition. The land sharing theory suggests that integrating farmlands, urban lands and natural lands (e.g. forest, grassland) in the same region can benefit C sinks in natural ecosystems through the use of N_r emitted from nearby farmlands and urban lands (Phalan et al., 2011; Paustian et al., 2016). In some circumstances, land sharing can also be beneficial to the conservation of biodiversity, e.g. by using tree shelter belts to protect sensitive habitats from excess N deposition near intensive farming locations (Bealey et al., 2016). Thus, coupling N_r emission/deposition with C sequestration at regional scale can maximize the use of N_r emission to mitigate global warming. It is critical to determine how far N_r emissions can be transported to areas downwind of the sources. Integrated modelling of air pollution derived from N_r emissions and C emission/sequestration at regional scale will help address the triple challenges of food security, environmental degradation and climate change.

Finally, N management in agriculture through precision farming and agro-ecological practices can potentially reduce N inputs in high input systems, and thereby reduce N_2O emissions and increase the agricultural C-sink potential. Agriculture is the largest source of N emission to natural terrestrial ecosystems, resulting in adverse effects on the environment and human health (Erisman et al., 2013). Besides the C sink goal, management of agricultural N_r use can increase crop yield and NUE in countries with economic development (Gu et al., 2015; Zhang et al., 2015). In addition to more stringent environmental regulations, improved agricultural production processes can shift yield responses to N input rate to produce more food with less N inputs (Chen et al., 2014; Lassaletta et al., 2014). This can benefit our society substantially through improved food security and environmental sustainability. The occurrences of inflection points as PGDP increases, suggests the potential to achieve a better management and use of N in agriculture, which also reduces N_2O emissions and thereby contribute to the mitigation of global warming. Although croplands have relatively low potential for C sequestration (Lam et al., 2013), measures such as minimum tillage, crop residue return, and perennial cropping still may increase the C sequestration in croplands. Future measures on promoting C sink need to take the N management into consideration to maximize both the N use and C sink while reducing their adverse effects on the environment and global climate warming (Van Groenigen et al., 2017).

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Table 1 Nonlinear effects of socioeconomic development on N_r input/loss, CO_2 emission and C/N ratio in the fixed effects panel model

Variables	**N Fertilizer**	**NO_x**	**CBNF**	**NH_3**	**CO_2**	**$CO_2/(NO_x+NH_3)$**
Per-capita GDP	**0.630*****	**0.309*****	**-0.733*****	0.043	**0.571*****	**0.468*****
	(0.185)	**(0.064)**	**(0.013)**	(0.088)	**(0.084)**	**(0.090)**
Population	**0.875*****	0.126	**-0.031*****	-0.200	0.153	**0.477*****
	(0.225)	(0.094)	**(0.012)**	(0.115)	(0.147)	**(0.130)**
Urbanization	-0.011	0.000	MC	0.011*	-0.001	0.000
	(0.009)	(0.004)		(0.005)	(0.005)	(0.005)
Group dummy	**2.547****	**3.472*****	NA	**0.043**	**1.460*****	NA
	(0.864)	**(0.399)**		**(0.064)**	**(0.342)**	
PGDP × dummy	**-1.002*****	**-1.295*****	NA	**-0.358**	**-0.571*****	NA
	(0.307)	**(0.136)**		**(0.184)**	**(0.125)**	
Intercept	-1.271***	0.291	3.222***	2.737***	0.394	3.395
	(0.330)	(0.174)	(0.030)	(0.223)	(0.263)	(0.283)
N	5595	3842	4336	3347	3811	3347
R^2-within	0.223	0.550	0.457	0.264	0.540	0.280

Note: all data of variables in this table have been transformed logarithmically, and all the variables have been tested for stationary to make sure all the panel data are balanced by using ADF-Fisher test in the analysis. Group dummy is a binary parameter (0, 1) introduced to test whether or not the inflection points of N_r fluxes/CO_2 emissions exist with per capita GDP. Cluster-robust standard errors (cluster at country level) were used for estimations. $^{***}P<0.001$, $^{**}P<0.01$, $^{*}P<0.05$; NA, not applicable; MC, multicollinearity with per capita GDP; R^2-within was estimated based on the group deviation method.

Figure captions

Fig. 1. Conceptual models of N_r use/loss and CO_2 emission with increasing level of economic development. (a) Conceptual models; (b) Evolution of the relationships among N inputs, N production and N use efficiency (NUE) with increasing economic development. (I), (II) and (III) refer to Type 1, 2 and 3 countries, respectively.

Fig. 2. Per-capita N_r use/loss, CO_2 emission and C/N ratio in relation to per-capita GDP across 132 countries. (a) N fertilizer; (b) CBNF; (c) NO_x emission from fossil fuel combustion; (d) NH_3 emission; (e) CO_2; (f) $CO_2/(NO_x+NH_3)$. CBNF, cultivated biological N fixation. Green data points represent Type 1 countries with an inflection point, and grey data points represent Type 2 countries without an inflection point. There was no inflection point for CBNF, and we applied the list of Type 1 countries of N fertilizer for CBNF due to the supplementary role of CBNF in food production compared to that of N fertilizer. See SI for the list of different types of countries.

Fig. 3. Spatial variation of global net ecosystem productivity (NEP) considering N_r deposition changes with economic development in 2050. The storyline used in this simulation is the IPCC B1 scenario.

Fig. 4. The potential explanation pathways of N_r uses and losses and CO_2 emission across 132 countries. (a) Grain yield with per-capita GDP; (b) PFP_N with GDP per capita; (c) CBNF with N fertilizer use; (d) energy use and GDP per capita; (e) NO_xE with GDP per capita; (f) CO_2E with GDP per capita. PFP_N, partial factor productivity of N fertilizer = kg grain yield per kg N fertilizer; CO_2E, CO_2 emission per unit of energy supply; NO_xE, NO_x emission per unit of energy consumed. Green data points represent Type 1 countries with an inflection point, and grey data points represent Type 2 countries without an inflection point. We applied the list of Type 1 countries of N fertilizer for yield, PFP_N and CBNF, and we applied the list of Type 1 countries of NO_x for energy, NO_xE and CO_2E. R^2 is the determining factor of regression curve. *$P<0.001$* is for all the regression curves. See SI for the list of different types of countries.

Fig. 5. Comparisons of N fertilizer and NO_x and their related factors for Type 1 and Type 2 counties. (a) Per area N fertilizer (Fertilizer/A), per capita N fertilizer (Fertilizer/P), cropland yield (Yield), crop production per N fertilizer (PFP_N) and PGDP

(per-capita GDP); (b) Per capita NO_x emissions via fossil fuel combustion (NO_x), per capita energy consumption (Energy), NO_x emission per energy consumption (NO_xE), and per-capita GDP; (c)-(d) changes of Type 1 countries from inflection year to 2006-2008. Type1_Inflection represents the countries in the year when inflection occurred, Type1_Current represents the current status (average value from 2006 to 2008) of Type 1 countries, and Type2_Current represents the current status (average value of 2006 to 2008) of Type 2 countries. See SI for the list of different types of countries.

Fig. 6. Cluster analysis for Type 1, 2 and 3 countries. (a) Cluster analysis based on per-capita GDP, fertilizer per hectare and yield; (b) quantitative comparisons of the three types in (a). The list of countries for the cluster analysis can be found in SI. Although Type 3 countries are transition countries between Type 1 and Type 2 countries in terms of economic development and N_r use, the much higher Fertilizer/A and lower PFP_N of Type 3 countries compared to the other two types of countries suggest serious N_r pollution in Type 3 countries. Units: Fertilizer/P (kg N capita^{-1} yr^{-1}), Fertilizer/A (10 kg N ha^{-1} yr^{-1}), Yield (100 kg rice milled equivalent ha^{-1} yr^{-1}), PFP_N (g rice milled equivalent g^{-1} N fertilizer), PGDP ($1,000 capita^{-1} yr^{-1}). See SI for the list of different types of countries.

Fig. 7. Comparisons of the inflection points on N fertilizer use, NH_3 emissions and P fertilizer use in Type 1 countries. (a) The year (+1900) and per-capita GDP (×100 capita^{-1} yr^{-1}) of the inflection points on N fertilizer use (kg N capita^{-1} yr^{-1}), NH_3 emissions (kg NH_3 capita^{-1} yr^{-1}) and P fertilizer use (kg P_2O_5 capita^{-1} yr^{-1}), and changes from the inflection year to the present (average data from 2006 to 2008); (b) Relationships between N fertilizer and NH_3 emissions and P fertilizer use in the inflection point year. See SI for the list of Type 1 countries.

Fig. 8. Typical illustrative examples for both Type 1 and Type 2 countries on the relationships of N fertilizer use and NO_x emission with per-capita GDP. The open circles in each panel are the calculated N fertilizer use or NO_x emissions in a particular year between 1961 and 2008 as a function of per-capita GDP. The solid lines are the regression curves except the one for China that is moved average. (a) Inflection of N fertilizer. (b) No inflection point for N fertilizer use; (c) Inflection point for NO_x emissions. (d) No inflection point for NO_x emissions.

Fig. 1.

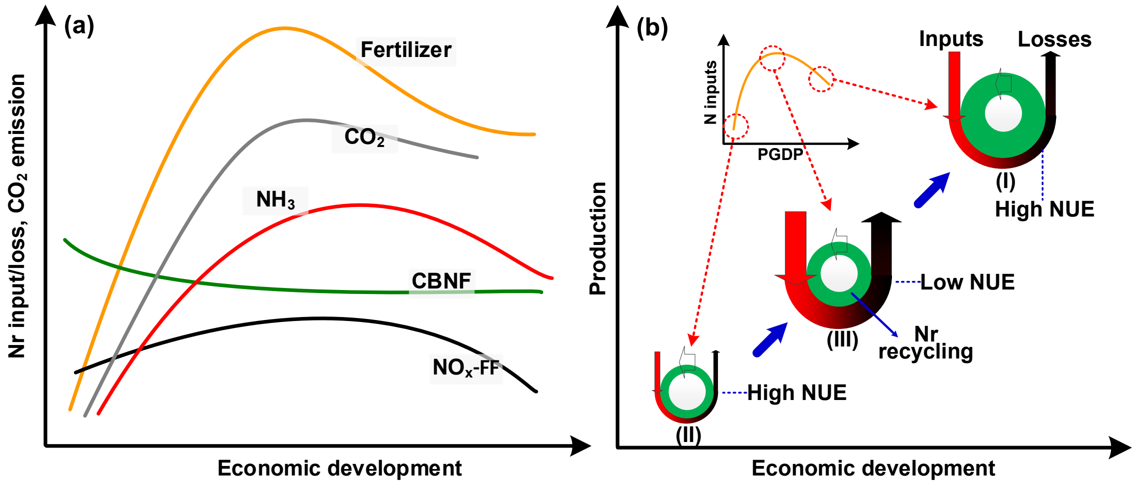

(a)
Nr input/loss, CO₂ emission
Fertilizer
CO₂
NH₃
CBNF
NOx-FF
Economic development
(b)
Production
N inputs
PGDP
Inputs
Losses
(I)
High NUE
Low NUE
(III)
Nr recycling
High NUE
(II)
Economic development

Fig. 2.

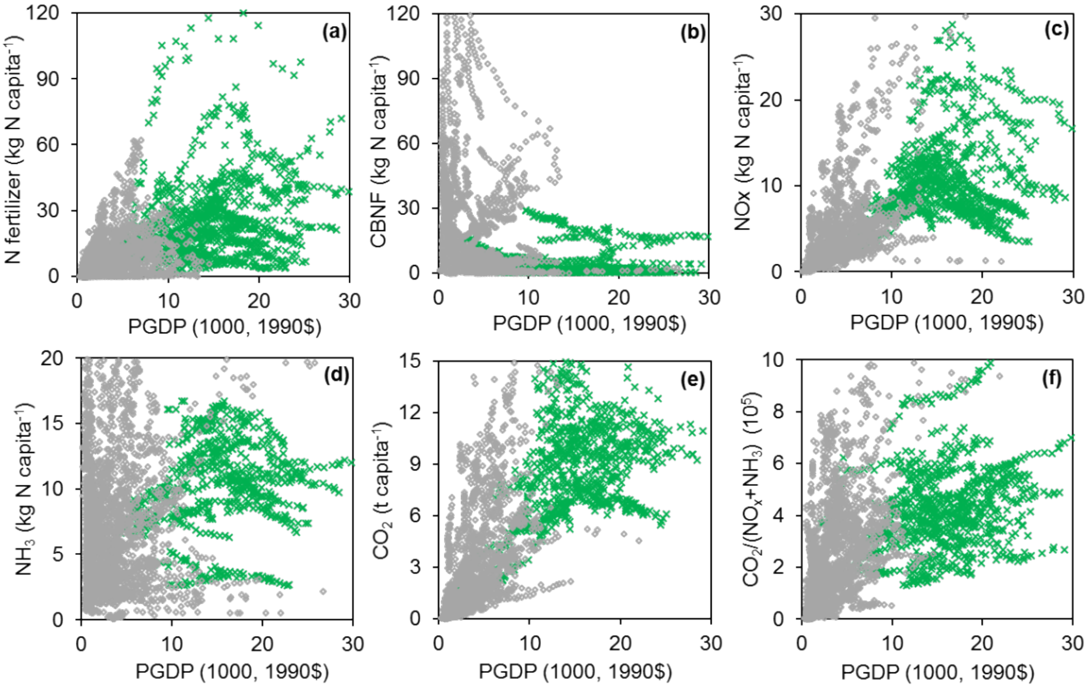

(a)
N fertilizer (kg N capita^{-1})
0
30
60
90
120
PGDP (1000, 1990$)
0
10
20
30
(b)
CBNF (kg N capita^{-1})
0
30
60
90
120
PGDP (1000, 1990$)
(c)
NOx (kg N capita^{-1})
0
10
20
30
PGDP (1000, 1990$)
(d)
NH$_3$ (kg N capita^{-1})
0
5
10
15
20
PGDP (1000, 1990$)
(e)
CO$_2$ (t capita^{-1})
0
3
6
9
12
15
PGDP (1000, 1990$)
(f)
CO$_2$/(NO$_x$+NH$_3$) (10^5)
0
2
4
6
8
10
PGDP (1000, 1990$)

Fig. 3.

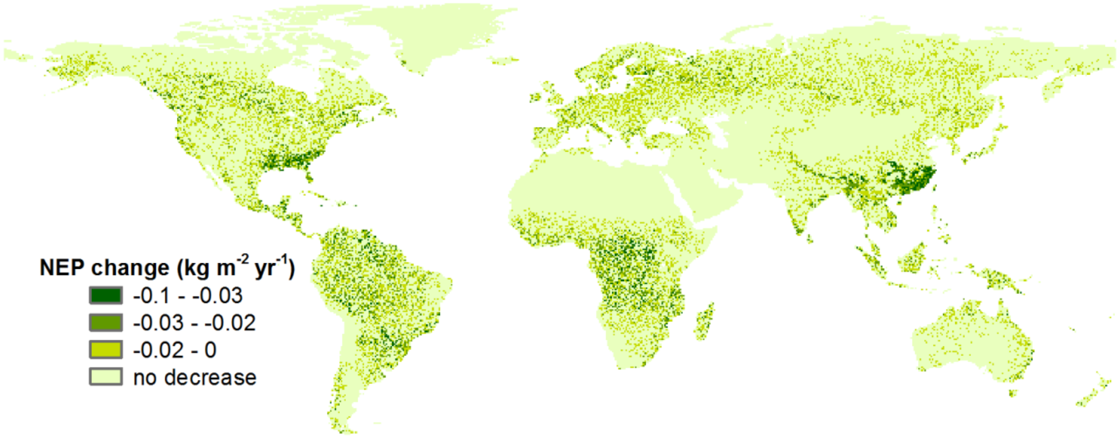


Fig. 4

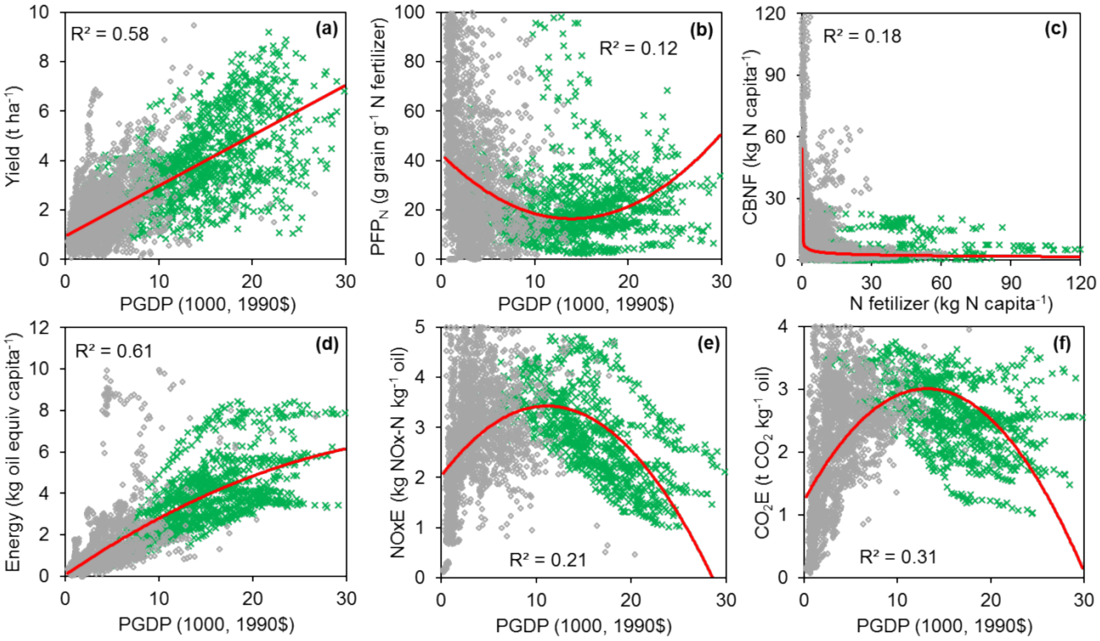

(a)
R² = 0.58
Yield (t ha^{-1})
PGDP (1000, 1990$)
(b)
R² = 0.12
PFP$_N$ (g grain g^{-1} N fertilizer)
PGDP (1000, 1990$)
(c)
R² = 0.18
CBNF (kg N capita^{-1})
N fetilizer (kg N capita^{-1})
(d)
R² = 0.61
Energy (kg oil equiv capita^{-1})
PGDP (1000, 1990$)
(e)
R² = 0.21
NOxE (kg NOx-N kg^{-1} oil)
PGDP (1000, 1990$)
(f)
R² = 0.31
CO$_2$E (t CO$_2$ kg^{-1} oil)
PGDP (1000, 1990$)

Fig. 5

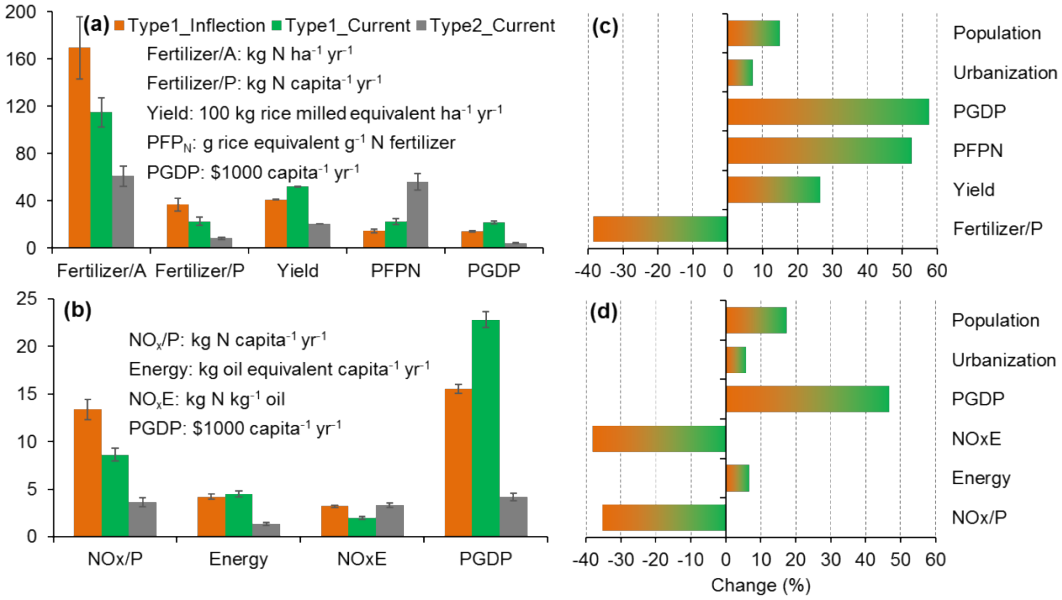

(a)
Type1_Inflection
Type1_Current
Type2_Current
Fertilizer/A: kg N ha-1 yr-1
Fertilizer/P: kg N capita-1 yr-1
Yield: 100 kg rice milled equivalent ha-1 yr-1
PFPN: g rice equivalent g-1 N fertilizer
PGDP: $1000 capita-1 yr-1
200
160
120
80
40
0
Fertilizer/A
Fertilizer/P
Yield
PFPN
PGDP
(b)
NOx/P: kg N capita-1 yr-1
Energy: kg oil equivalent capita-1 yr-1
NOxE: kg N kg-1 oil
PGDP: $1000 capita-1 yr-1
25
20
15
10
5
0
NOx/P
Energy
NOxE
PGDP
(c)
Population
Urbanization
PGDP
PFPN
Yield
Fertilizer/P
-40 -30 -20 -10 0 10 20 30 40 50 60
(d)
Population
Urbanization
PGDP
NOxE
Energy
NOx/P
-40 -30 -20 -10 0 10 20 30 40 50 60
Change (%)

Fig. 6

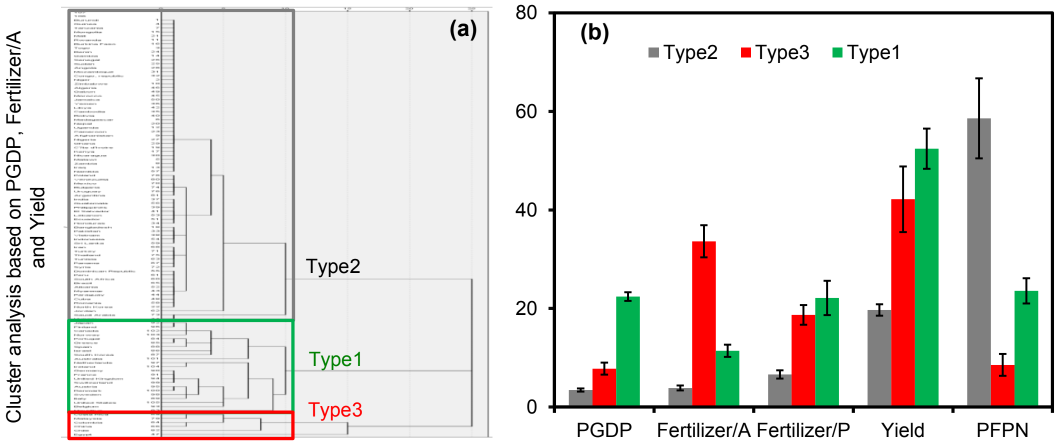

Cluster analysis based on PGDP, Fertilizer/A and Yield
(a)
Type2
Type1
Type3
(b)
■ Type2 ■ Type3 ■ Type1
80
60
40
20
0
PGDP
Fertilizer/A
Fertilizer/P
Yield
PFPN

Fig. 7

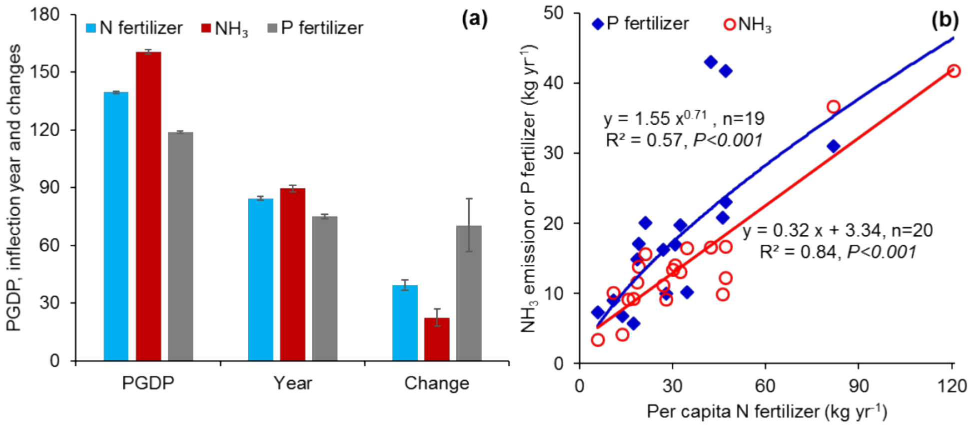
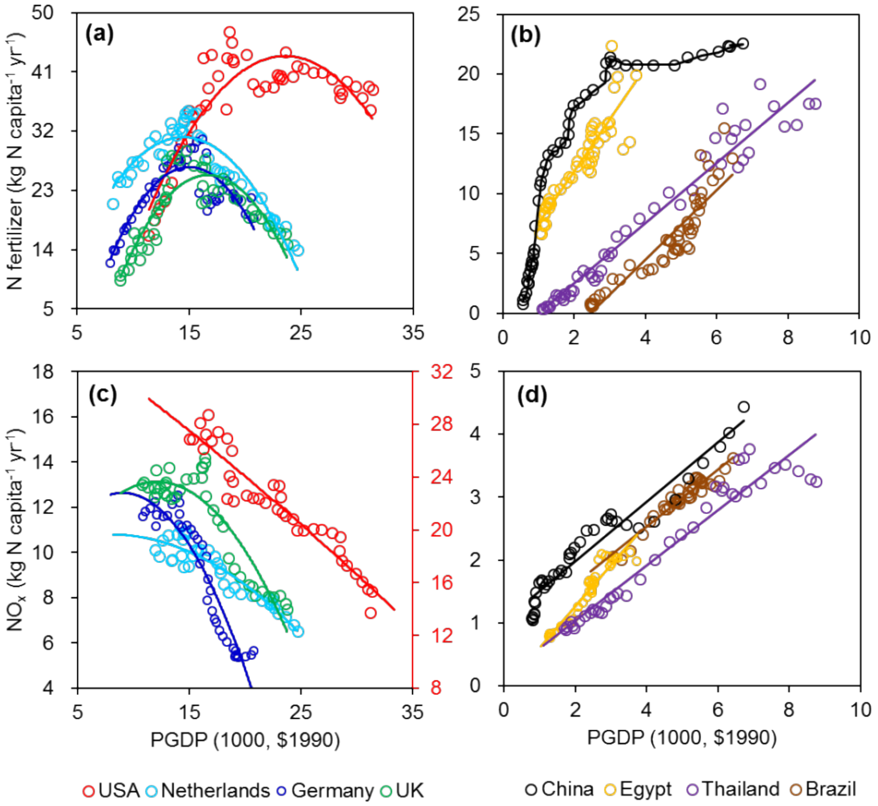

(a)
N fertilizer
NH_3
P fertilizer
PGDP, inflection year and changes
0
30
60
90
120
150
180
PGDP
Year
Change
(b)
P fertilizer
NH_3
$y = 1.55\ x^{0.71}$, n=19
$R^2 = 0.57$, $P<0.001$
$y = 0.32\ x + 3.34$, n=20
$R^2 = 0.84$, $P<0.001$
NH_3 emission or P fertilizer (kg yr^{-1})
0
10
20
30
40
50
0
30
60
90
120
Per capita N fertilizer (kg yr^{-1})

Fig. 8


(a)
(b)
(c)
(d)
N fertilizer (kg N capita^{-1} yr^{-1})
NO$_x$ (kg N capita^{-1} yr^{-1})
PGDP (1000, $1990)
PGDP (1000, $1990)
50
41
32
23
14
5
25
20
15
10
5
0
18
16
14
12
10
8
6
4
32
28
24
20
16
12
8
5
4
3
2
1
0
5
15
25
35
0
2
4
6
8
10
○ USA ○ Netherlands ○ Germany ○ UK
○ China ○ Egypt ○ Thailand ○ Brazil

Supplementary Information

SM text

Inflection points hypothesis

To increase food production, N fertilizer and legume cultivation are used to maximize N input to agricultural lands (Fig. S1). In order to supply energy, the burning of fossil fuel inevitably increases NO_x and CO_2 emissions to the atmosphere (Sutton et al., 2013). Zhang et al. (Zhang et al., 2015) showed that the N loss from cropland follows a bell-shaped relationship with economic growth. Here we expand this relationship to anthropogenic N_r creation. On the one hand, greater income increases demand for more food and energy consumption, which in turn increases the N_r input to agricultural lands as nutrient for plant growth, and the emission of NO_x and CO_2 to the atmosphere through fossil fuel combustion (Tilman et al., 2011). On the other hand, a higher income is often accompanied by a societal demand for improved environmental quality, such as clean water and air, and the mitigation of climate change (Zhang et al., 2015). Consequently, governments may impose regulatory policies or offer subsidies and incentives to reduce local/regional N pollution and mitigate global warming by increasing resource use efficiencies and C sinks.

Therefore, we hypothesize that N_r creation, loss and CO_2 emissions follow a pattern similar to an environmental Kuznets curve (EKC): N_r creation and loss (NH_3, NO_x, and N_2O), and CO_2 emissions increase with income growth and the quest for food and energy at the early stages of economic development, but then decrease with further income growth at a more affluent stage (Fig. S2). Future climate change is tightly linked with CO_2 emissions and changes in C sinks that are dependent on N supply to the ecosystems through N deposition (Hungate et al., 2003). Thus, the ratio of CO_2 emissions to N_r emissions, including NH_3 and $NO_{x,}$ to the atmosphere is crucial for the future climate change. Although both CO_2 and N_r emissions may follow the EKC with economic development, CO_2 emission is more tightly coupled with the energy supply by fossil fuel combustion. Therefore, we hypothesized that the C:N ratio (emissions of CO_2 to NH_3 and NO_x) would continue to increase with the economic growth, compromising the potential increase of C sinks under future elevated CO_2 concentration.

References:

Hungate, B.A., Dukes, J.S., Shaw, M.R., Luo, Y., Field, C.B., 2003. Nitrogen and climate change. Science 302, 1512-1513.

Sutton, M.A., Reis, S., Riddick, S.N., Dragosits, U., Nemitz, E., Theobald, M.R., Tang, Y.S., Braban, C.F., Vieno, M., Dore, A.J., Mitchell, R.F., Wanless, S., Daunt, F., Fowler, D., Blackall, T.D., Milford, C., Flechard, C.R., Loubet, B., Massad, R., Cellier, P., Personne, E., Coheur, P.F., Clarisse, L., Van Damme, M., Ngadi, Y., Clerbaux, C., Skjoth, C.A., Geels, C., Hertel, O., Wichink Kruit, R.J., Pinder, R.W., Bash, J.O., Walker, J.T., Simpson, D., Horvath, L., Misselbrook, T.H., Bleeker, A., Dentener, F., de Vries, W., 2013. Towards a climate-dependent paradigm of ammonia emission and deposition. Philosophical Transactions of the Royal Society B: Biological Sciences 368, 20130166.

Tilman, D., Balzer, C., Hill, J., Befort, B.L., 2011. Global food demand and the sustainable intensification of agriculture. Proceedings of the National Academy of Sciences 108, 20260-20264.

Zhang, X., Davidson, E.A., Mauzerall, D.L., Searchinger, T.D., Dumas, P., Shen, Y., 2015. Managing nitrogen for sustainable development. Nature 528, 51-59.

Table S1 Testing for Stationary in unbalanced panel data

H0: all Panels contain unit roots	Fisher-ADF Test: drift term included	
	Modified inverse chi-squared Statistic	p-value
Ln(N Fertilizer)	48.3663	0.0000
Ln(NO_x)	21.4483	0.0000
Ln(NH_3)	33.1388	0.0000
Ln(CO_2)	23.3914	0.0000
Ln($CO_2/(NO_x+NH_3)$)	28.3823	0.0000
Ln(GDP per capita)	21.7810	0.0000
Ln(Population)	79.0207	0.0000
Urbanization	99.4736	0.0000
Ln(Yield)	43.5265	0.0000
Ln(Energy)	32.0228	0.0000

Table S2 Panel cointegration analysis for the C/N ratio with PGDP

	Coefficient	Std. Error	t-Statistic	Prob.
Constant	4.8278	0.0053	917.7913	0.0000
LnPGDP	0.3288	0.0920	3.5733	0.0004
$(\text{LnPGDP})^2$	1.0602	0.2951	3.5922	0.0003

Note, all data of variables in this table have been transformed logarithmically.

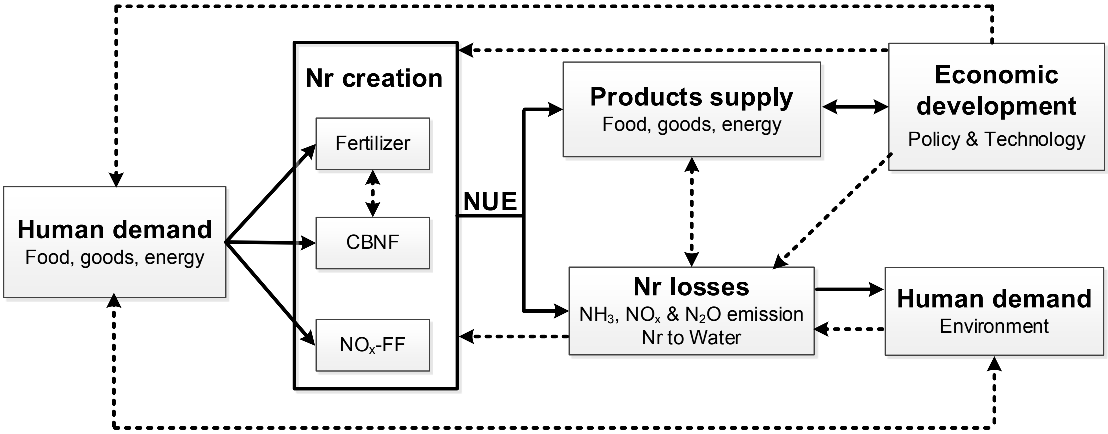


Fig. S1 Framework of the economic driving changes on N_r use and loss. Solid lines represent the drivers that may increase the amount or level of the objectives such as N fertilizer; dashed lines represent the regulations that may decrease the amount or level of the objectives such as N losses. The interactions among these two sets of variables would finally result in the emergences of inflection points for N_r use and loss with economic development.

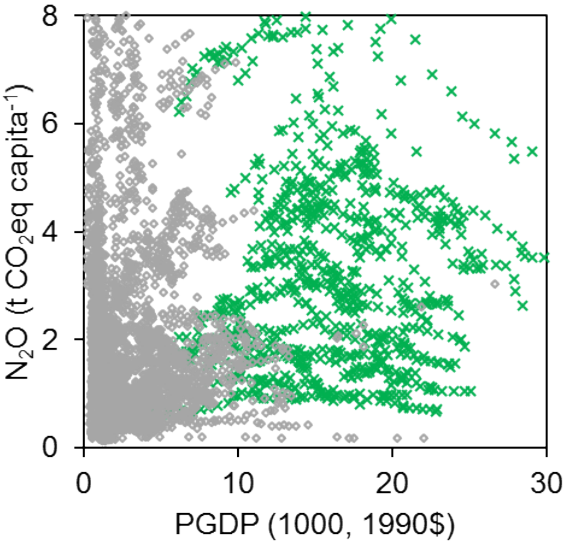


Fig. S2 Per capita N_2O emission in relation to GDP per capita across 132 countries. Green data points represent Type 1 countries with an inflection point, and grey data points represent Type 2 countries without an inflection point.

Country list with N_r inflection points

For per capita N fertilizer use

Type 1 countries	Australia, Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Israel, Italy, Japan, Mexico, The Netherlands, New Zealand, Norway, Portugal, South Korea, Spain, Sweden, Switzerland, The United Kingdom, The United States
Type 2 countries	Afghanistan, Albania, Algeria, Angola, Argentina, Bahrain, Bangladesh, Benin, Bolivia, Botswana, Brazil, Bulgaria, Burkina Faso, Burundi, Cambodia, Cameroon, Canada, Cape Verde, Central African Republic, Chad, Chile, China, Colombia, Comoro Islands, Congo, Democratic Republic of Congo, Costa Rica, Côte d'Ivoire, Cuba, Djibouti, Dominican Republic, Ecuador, Egypt, El Salvador, Equatorial Guinea, Gabon, Gambia, Ghana, Guatemala, Guinea, Guinea Bissau, Haïti, Honduras, Hungary, India, Indonesia, Iran, Iraq, Jamaica, Jordan, Kenya, Kuwait, Laos, Lebanon, Lesotho, Liberia, Libya, Madagascar, Malawi, Malaysia, Mali, Mauritania, Mauritius, Mongolia, Morocco, Mozambique, Myanmar, Namibia, Nepal, Nicaragua, Niger, Nigeria, North Korea, Oman, Pakistan, Panama, Paraguay, Peru, The Philippines, Poland, Qatar, Romania, Rwanda, São Tomé and Principe, Saudi Arabia, Senegal, Seychelles, Sierra Leone, Singapore, Somalia, South Africa, Sri Lanka, Sudan, Swaziland, Syria, Tanzania, Thailand, Togo, Trinidad and Tobago, Tunisia, Turkey, Uganda, United Arab Emirates, Uruguay, Venezuela, Vietnam, Yemen, Zambia, Zimbabwe

Type 1 countries have an inflection point, while Type 2 countries have not.

For per capita NO_x emission from fossil fuel combustion

Type 1 countries	Australia, Austria, Belgium, Canada, Denmark, Finland, France, Germany, Greece, Ireland, Israel, Italy, Japan, The Netherlands, New Zealand, Norway, Portugal, Singapore, South Korea, Spain, Sweden, Switzerland, The United Kingdom, The United States
Type 2 countries	Afghanistan, Albania, Algeria, Angola, Argentina, Bahrain, Bangladesh, Benin, Bolivia, Botswana, Brazil, Bulgaria, Burkina Faso, Burundi, Cambodia, Cameroon, Cape Verde, Central African Republic, Chad, Chile, China, Colombia, Comoro Islands, Congo, Democratic Republic of Congo, Costa Rica, Côte d'Ivoire, Cuba, Djibouti, Dominican Republic, Ecuador, Egypt, El Salvador, Equatorial Guinea, Gabon, Gambia, Ghana, Guatemala, Guinea, Guinea Bissau, Haïti, Honduras, Hungary, India, Indonesia, Iran, Iraq, Jamaica, Jordan, Kenya, Kuwait, Laos, Lebanon, Lesotho, Liberia, Libya, Madagascar, Malawi, Malaysia, Mali, Mauritania, Mauritius, Mexico, Mongolia, Morocco, Mozambique, Myanmar, Namibia, Nepal, Nicaragua, Niger, Nigeria, North Korea, Oman, Pakistan, Panama, Paraguay, Peru, Philippines, Poland, Qatar, Romania, Rwanda, São Tomé and Principe, Saudi Arabia, Senegal, Seychelles, Sierra Leone, Somalia, South Africa, Sri Lanka, Sudan, Swaziland, Syria, Tanzania, Thailand, Togo, Trinidad and Tobago, Tunisia, Turkey, Uganda, United Arab Emirates, Uruguay, Venezuela, Vietnam, Yemen, Zambia, Zimbabwe

Cluster analysis based on per-capita GDP, N fertilizer use per hectare cropland, and crop yield

Type 1 countries	Mauritius, Portugal, Greece, Israel, South Korea, Spain, Italy, Germany, France, Japan, Belgium, The United Kingdom, Austria, Finland, The Netherlands, Sweden, Switzerland, Denmark, Australia, Canada, Norway, Ireland, The United States
Type 2 countries	Burundi, Niger, Togo, Guinea, Malawi, Madagascar, Tanzania, Zambia, Afghanistan, Zimbabwe, Rwanda, Uganda, Iraq, Gambia, Mongolia, Burkina Faso, Kenya, Bangladesh, Côte d'Ivoire, Nepal, Mali, North Korea, Cameroon, Benin, Sudan, Senegal, Nigeria, Angola, Ghana, Nicaragua, Mozambique, Democratic Republic of Congo, Pakistan, Honduras, Cambodia, Yemen, India, Vietnam, Philippines, Bolivia, El Salvador, Libya, Myanmar, Paraguay, Morocco, Algeria, Cuba, Gabon, Jamaica, Ecuador, Albania, Lebanon, Indonesia, Dominican Republic, Guatemala, Namibia, Romania, Sri Lanka, South Africa, Peru, Jordan, Tunisia, Brazil, Panama, Iran, Mexico, Turkey, Syria, Saudi Arabia, Bulgaria, Thailand, Uruguay, Hungary, Poland, Venezuela, Argentina
Type 3 countries	Egypt, Colombia, China, Costa Rica, Malaysia, Chile

Type 3 countries are similar to Type 2 countries, but are close to an inflection point.

Inflection point analysis for ammonia (NH_3) emission and phosphorus (P) fertilizer use

Countries with inflection points for NH_3 emissions	Austria, Belgium, Denmark, Finland, France, Germany, Greece, Ireland, Israel, Italy, Japan, Mexico, The Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, The United Kingdom, The United States
Countries with inflection points for P fertilizer use	Australia, Austria, Belgium, Canada, Denmark, Finland, France, Germany, Greece, Hungary, Israel, Italy, Japan, Mexico, The Netherlands, Norway, Sweden, Switzerland, The United Kingdom, The United States