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Spatial and decadal variations in inorganic nitrogen wet deposition in China induced by human activity

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Atmospheric nitrogen (N) deposition, an important component in the global N cycle, has increased sharply in recent decades in China. Here, we constructed national-scale inorganic N wet deposition (N_{dep}) patterns in China based on data from 280 observational sites and analysed the effects of anthropogenic sources and precipitation on N_{dep}. Our results showed that the mean N_{dep} over China increased approximately 25%, from 11.11 kg ha^{-1} a^{-1} in the 1990s to 13.87 in the 2000s. N_{dep} was highest over southern China and exhibited a decreasing gradient from southern to western and northern China. The decadal difference in N_{dep} between the 1990s and 2000s was primarily caused by increases in energy consumption and N fertiliser use. Our findings conformed that anthropogenic activities were the main reason for the N_{dep} increase and provide a scientific background for studies on ecological effects of N deposition in China.

Atmospheric nitrogen (N) deposition has dramatically increased worldwide in the past few decades[1,2]. Although these N inputs to ecosystems can increase food production[3] and stimulate plant growth[4–6], particularly in N-limited regions, excessive N has caused a cascade of detrimental effects on natural ecosystems and humans, including the acidification of soils and waters[7], negative impacts on the buffering capacity of soil[8], a loss of biological diversity[9,10], and risks to human health[11]. Simultaneously, N deposition can influence the balance of greenhouse gases (N oxide and ozone in addition to carbon dioxide)[1,12]. Therefore, an understanding of the spatio-temporal patterns and the factors controlling N deposition is essential for evaluating its ecological effects, and can provide the scientific background for research into global changes.

Both natural and anthropogenic sources contribute to atmospheric N deposition, but anthropogenic reactive N (N_r) emissions dominate N deposition at both continental and regional scales[13,14]. Anthropogenic N_r is released into the atmosphere either as N oxides ($NO_x = NO + NO_2$), primarily from fossil fuel combustion, or as ammonia (NH_3), primarily from agriculture[2,13]. After a series of chemical conversions and physical transport, the ultimate fate of NO_x and NH_3 is removal by wet scavenging and dry deposition on terrestrial and aquatic ecosystems[2,15]. Although the process of N deposition is relatively clear in theory and has been applied in models, knowledge of how anthropogenic sources influence N deposition at the regional scale using observational or statistical data is lacking.

Anthropogenic N_r emissions have increased significantly in China due to rapid agricultural production and industrial development in the past three decades[16], and progress has been made with regard to research into N deposition in China over the last decade, particularly wet (bulk) deposition. Three important aspects of this research are as follows. Observations of N deposition in situ have been more extensively studied across China from 1980 to the present[17,18]. Meta-analyses based on this massive observational dataset have been implemented in recent years to evaluate nationwide N deposition dynamics and spatial patterns[19,20]. Simultaneously, atmospheric chemistry transport models have also been implemented to simulate current and future N deposition in China[2,21]. Such studies have all indicated high N deposition in China. Nonetheless, it remains unclear how the pattern of N deposition is determined and what role anthropogenic sources play, an issue that requires further investigation.

This study established nationwide datasets of inorganic N wet deposition (abbreviated as N_{dep}) from 280 observational sites and amassed data on N fertiliser use, energy consumption, and precipitation in China between

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the 1990s and 2000s. Based on these data, this study attempts to address the following questions: (1) What are the spatial patterns of N_{dep} in the 1990s and 2000s in China? (2) How do anthropogenic sources and precipitation influence the spatial pattern of N_{dep}? (3) Do the factors controlling the spatial pattern of N_{dep} also determine decadal variation?

Results

Spatial patterns of N_{dep} in the 1990s and 2000s. In China, the magnitude and spatial pattern of N_{dep} differed significantly according to region and decade (Fig. 1). In the 1990s, N_{dep} exhibited a decreasing gradient from southern to western and to northern China. N_{dep} was >30 kg ha^{-1} a^{-1} in some provinces of southern China, such as Hunan, Hubei, Guangdong, and Guangxi, whereas N_{dep} in other provinces of southern China was approximately 20–25 kg ha^{-1} a^{-1}. N_{dep} over northern, northeastern, and northwestern China was approximately 10–20, 5–15, and 0–10 kg ha^{-1} a^{-1}, respectively (Fig. 1a).

Compared to the 1990s, N_{dep} increased across nearly all of China in the 2000s (Fig. 1b), and mean N_{dep} over China increased by approximately 25% from the 1990s (11.11 kg ha^{-1} a^{-1}) to the 2000s (13.87 kg ha^{-1} a^{-1}). N_{dep} over most parts of southern China reached 30 kg ha^{-1} a^{-1} and also increased to some extent in other

regions. It is worth noting that North China also gradually became a centre for N_{dep}, with a value of 25 kg ha^{-1} a^{-1}.

The effect of metropolitan areas on N_{dep} in natural forest ecosystems. Cities are centres of human activity that emit large quantities of N pollution and can influence surrounding ecosystems through atmospheric transport. N emissions from natural forest ecosystems are mainly due to the decomposition of soil and animal excreta, although the magnitude of N deposition induced by these emissions is quite limited. We analysed the relationship between the N_{dep} of natural forest ecosystems and the distance from a forest site to the nearest large city with more than 2 million inhabitants. The results indicated that N_{dep} at a forest site was significantly related to the distance to the nearest city using a power model ($R^2 = 0.57$, $P < 0.001$) (Fig. 2). The distance within a city affected forest N_{dep} was approximately 200 km (shadow in Fig. 2). This result indicates that anthropogenic sources influence N_{dep} primarily at the local scale and vice versa, i.e., N_{dep} was primarily influenced by local anthropogenic sources. This finding was the basis for studying the relationship between anthropogenic sources (N fertiliser use and energy consumption) and the spatial pattern of N_{dep} by province in the ensuing analyses.

The effects of anthropogenic sources and precipitation on the spatial pattern of N_{dep}. N fertiliser use, energy consumption, and precipitation were significantly correlated to N_{dep} by province in the 1990s (Fig. 3), contributing 60%, 51%, and 63%, respectively, to the spatial variation of N_{dep} ($P < 0.001$). N_{dep} was strongly logarithmically related to N fertiliser use and energy consumption (Fig. 3a,b) and linearly related to precipitation (Fig. 3c). It is worth noting that there was a large dispersion of N_{dep} in provinces where N fertiliser use was approximately 3–8 t km^{-2} a^{-1} (shadow in Fig. 3a). We found that precipitation played an important role in this dispersion; for example, given similar magnitudes of N fertiliser use in different provinces, high precipitation usually accompanied high N_{dep} (Supplementary Table S1). A similar situation was observed for the relationship between energy consumption and N_{dep}. Conversely, in the case of relatively consistent precipitation, those provinces with high N_{dep} also had high N fertiliser use or energy consumption (shadow in Fig. 3c and Supplementary Table S1). Therefore, anthropogenic sources and precipitation need be considered together when studying the factors that control the spatial patterns of N_{dep} at the regional scale.
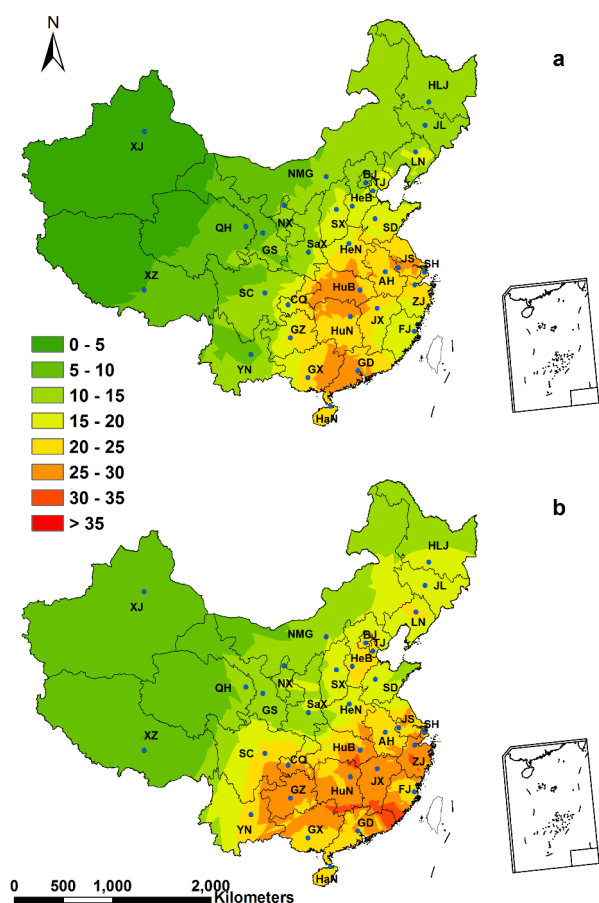

Figure 1 | Spatial patterns of N_{dep} in China (kg ha^{-1} a^{-1}). Spatial distribution maps of N_{dep} in the 1990s (a) and 2000s (b) were obtained from 136 and 144 monitoring sites, respectively, by Kriging interpolation. The letters denote the abbreviations of province names in China, and the full names of the provinces are listed in Supplementary Table S1. The dots indicate the location of the capital of each province. The distribution of monitoring sites is shown in Supplementary Figure S2. The maps were generated using ArcGIS 10.0 software.
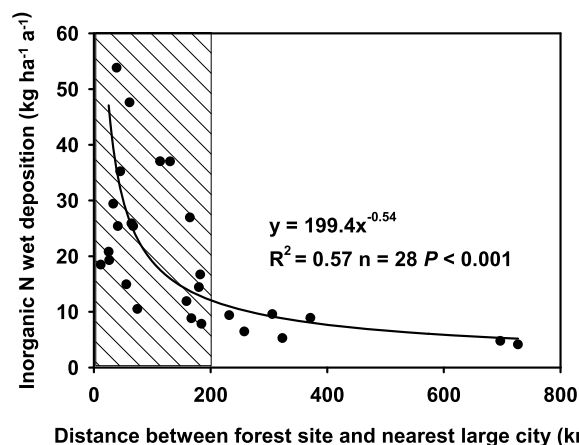

Figure 2 | Correlation between N_{dep} in natural forest ecosystems and the distance between the forest site and nearest large city. All natural forest monitoring sites in the 2000s were selected from our datasets. Large cities were selected if they contained more than 2 million inhabitants. Note: the area of shadow indicates the distance to which the N_{dep} of a natural forest was strongly influenced by the nearest city.

Subsequently, we analysed the combined effect of N fertiliser use, energy consumption, and precipitation on the spatial variation of N_{dep} in China. The result showed that these three factors combined contributed 79% of the spatial variation of N_{dep} (equation (1)).

$$N_{dep} = 3.3366 \ln[(F_N \times 18.5\% + E \times 0.24\%) \times P] \\ - 5.0317 \; (R^2 = 0.79 \; P < 0.001) \tag{1}$$

where N_{dep} is inorganic N wet deposition (kg ha^{-1} a^{-1}), F_N is N fertiliser use (expressed as the volume of effective component) (t km^{-2} a^{-1}), E is energy consumption (as standard coal) (t km^{-2} a^{-1}), P is precipitation (mm), 18.5% is the average NH$_3$-N emission factor for N fertiliser[22], and 0.24% is the average NO$_x$-N emission factor for energy consumption[23].

We used data from the 2000s to test whether equation (1), established using data from the 1990s, can reflect the spatial variation of N_{dep} in China. The results showed that N_{dep} calculated using equation (1) was strongly related to the N_{dep} values for the 2000s interpolated by Kriging (Fig. 4), indicating that equation (1) can well describe the spatial patterns of N_{dep} in China. Assuming that the existing emission factors for N fertiliser and energy consumption do not change much, we can use N fertiliser use, energy consumption, and precipitation as the driving factors to simulate and predict future trends in N_{dep}.
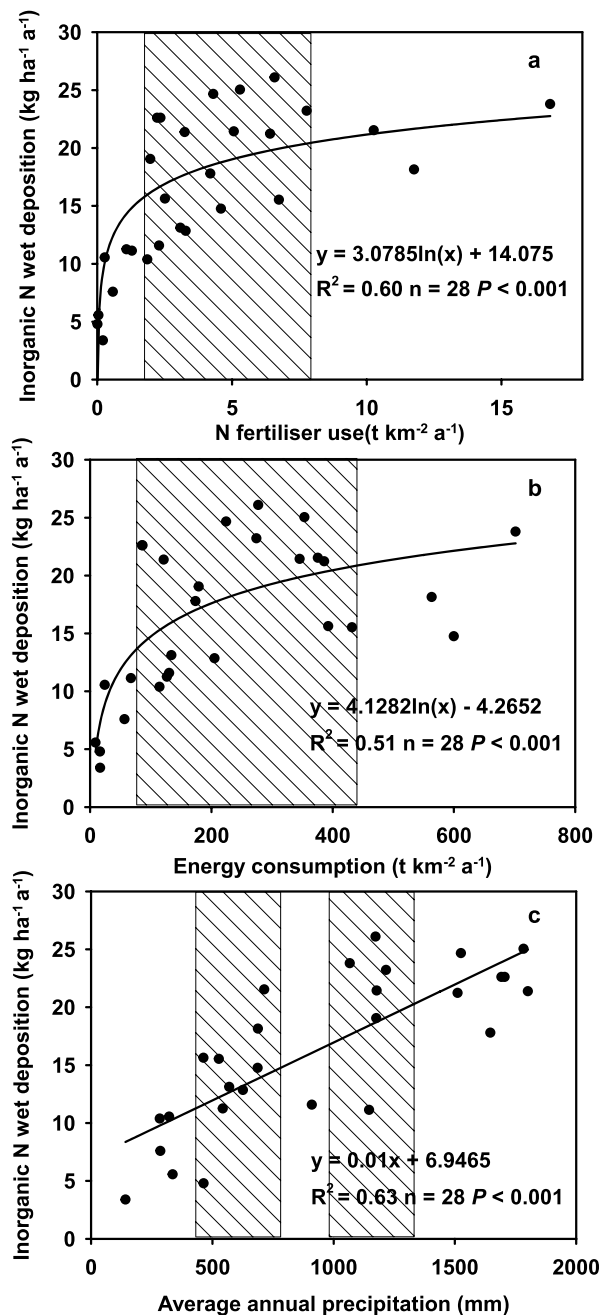
The effects of anthropogenic sources and precipitation on the decadal variation in N_{dep}. The changes in N fertiliser use, energy consumption, precipitation, and N_{dep} from the 1990s to the 2000s in China were quite different (Fig. 5). Compared to the 1990s, N fertiliser use increased by approximately 10%, energy consumption nearly doubled, precipitation decreased slightly, and N_{dep} increased by approximately 25% in the 2000s. We used equation (1) to calculate the individual effects of N fertiliser use, energy consumption, and precipitation on the decadal variation of N_{dep} (see Data and Methods). The results indicated that an increase in energy consumption dominated the change in N_{dep} from the 1990s to the 2000s in China, contributing approximately 80% of this decadal variation. N fertiliser use also contributed to the decadal increase in N_{dep}, but its role was relatively small. Precipitation had little effect on the decadal increase of N_{dep}, although there was an adverse effect, as shown in Fig. 6; this effect was quite small and may be produced from the deviation of equation (1).

Discussion

Atmospheric N deposition in China has dramatically increased in recent decades. In the present study, N_{dep} averaged 11.11 and 13.87 kg ha^{-1} a^{-1} in the 1990s and 2000s, respectively. Other studies

Figure 3 | The effects of N fertiliser use, energy consumption, and precipitation on the spatial pattern of N_{dep} in the 1990s. (a) N fertiliser use (expressed as the volume of effective component). The data on N fertiliser use in provinces were obtained from the China Statistical Yearbook[43]. (b) Energy consumption (expressed as standard coal). The data on energy consumption in provinces were primarily from the China Energy Statistical Yearbook[44]. (c) Precipitation. The data on the annual amount of precipitation were obtained from the China Meteorological Administration and included 756 weather stations (Supplementary Fig. S3). The mean N_{dep} in provinces was obtained from spatial maps of N_{dep} over China using the Zonal Statistics tool of ArcGIS 10.0 software. Notes: the area of shadowing indicates the provinces with large variations in N_{dep} when the x-axis variable was relatively constant.
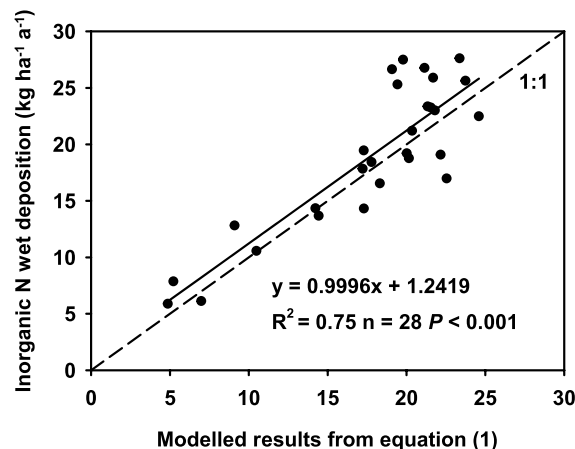
Figure 4 | Test of equation (1) using data from the 2000s. The x-axis variable was modelled results of N_{dep} in provinces, as obtained by equation (1) and data on N fertiliser use, energy consumption, and precipitation in provinces in the 2000s. The y-axis variable was interpolated results of N_{dep}, as obtained by Kriging interpolation.
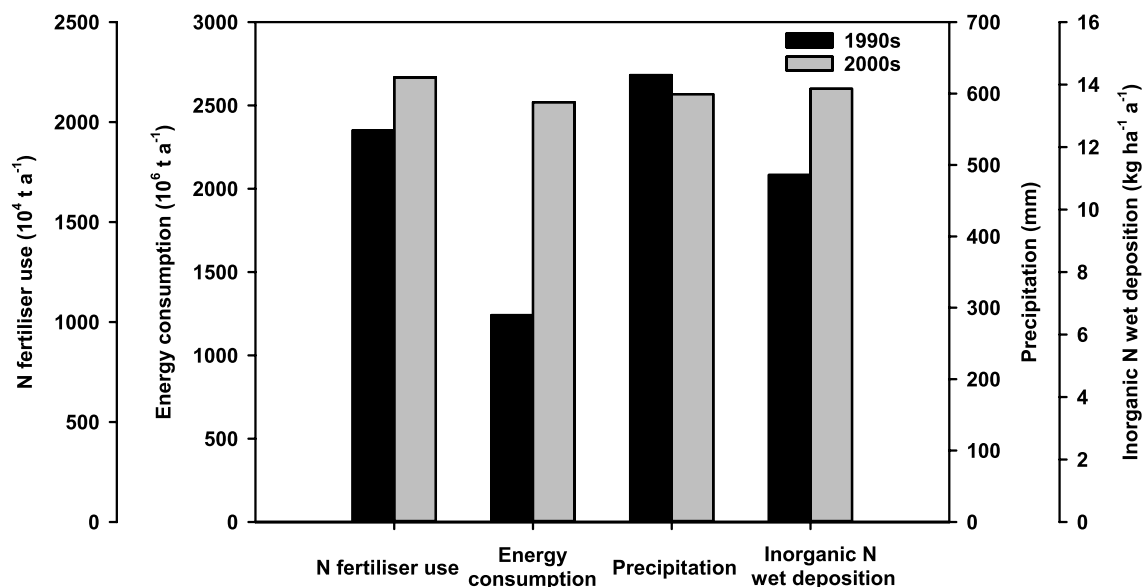
Figure 5 | N fertiliser use, energy consumption, precipitation, and N$_{dep}$ in China in the 1990s and 2000s. The data on N fertiliser use in provinces were obtained from the China Statistical Yearbook[43]. The data on energy consumption in provinces were primarily from the China Energy Statistical Yearbook[44]. The annual amount of precipitation was obtained from the China Meteorological Administration and included 756 weather stations (Supplementary Fig. S3). N$_{dep}$ was obtained by Kriging interpolation.

have also assessed wet deposition (the same N deposition species as in this study, including NH_4^+ and NO_3^-) in China using different approaches. Lü and Tian employed the Kriging technique to characterise spatial pattern of wet deposition, arriving at an average of 9.88 kg ha^{-1} a^{-1} during 1980–2005 throughout China[20], a value that is slightly lower than the results of our study. One possible reason for this discrepancy is the different time scales used in the two studies. Ti et al. noted that annual wet deposition averaged 5.37–9.1 kg ha^{-1} a^{-1} over China during 1985–2007 (ref. 3), which was based on the relationship between wet deposition from monitoring and N emissions, with a resolution of 0.5° × 0.5°; however, as the gridded emissions were obtained based on regional emission data and population data[24], that estimate of wet deposition may have a large uncertainty. By summarising monitoring data from publications and measurements, Liu et al. reported that annual bulk deposition (wet deposition) averaged 13.2 and 21.1 kg ha^{-1} a^{-1} in the 1980s and 2000s, respectively, over China[19]. Their results were considerably higher

than those of other studies, and a possible reason for this being that the mean wet deposition over China reported by Liu et al. was obtained directly from the arithmetic average of monitoring data, which did not consider the spatial differences in wet deposition in China. In summary, due to the lack of an extensive long-term monitoring network, the assessment of N deposition in China still has a large uncertainty. Nonetheless, the results from geostatistical methods considering spatial differences may be reliable[20,25].

Although the values of critical loads in different ecosystems have a given amount of uncertainty, it is currently believed that the values of terrestrial ecosystems are approximately 10–20 kg N ha^{-1} a^{-1} (ref. 10,25–27). Considering only N$_{dep}$ in this study (Fig. 1), we evaluated the possible regions where N deposition exceeded the critical loads (Supplementary Fig. S1). Based on our results, 53% and 16% of land in China received N deposition exceeding 10 and 20 kg N ha^{-1} a^{-1} in the 1990s, and these proportions reached 61% and 24%, respectively, in the 2000s. However, according to the results from their models, Dentener et al. reported that 11% of the world's natural vegetation received N deposition exceeding the critical load of 10 kg N ha^{-1} a^{-1} (ref. 2), and Zhang et al. noted that 35% of US land received N deposition exceeding this load[14]. These results indicated that the potential risk of N saturation may be higher in China than in other regions, which may cause a cascade of detrimental effects on natural ecosystems and humans, such as the acidification of soils and waters[7], a negative impact on the soil's buffering capacity[8], the loss of biological diversity[9,10], and human health risks[11]. Consequently, it is important and urgent to study the impact of N saturation on terrestrial ecosystems in China.

Previous studies have suggested that anthropogenic activities are the main cause of atmospheric N deposition[2,13,14,19,28,29] and that the pattern of wet deposition depends on precipitation and N emissions[14,15]; however, these results were obtained primarily from model simulations and isotope techniques. For example, Zhang et al. used the GEOS-Chem global chemical transport model to separate the contributions to N deposition from anthropogenic and natural sources[14]; the results showed that anthropogenic and natural sources account for 84% and 16%, respectively, of N deposition in the US. Russell et al. combined back trajectories and isotope techniques to reveal that the dominant sources of N deposition were most likely
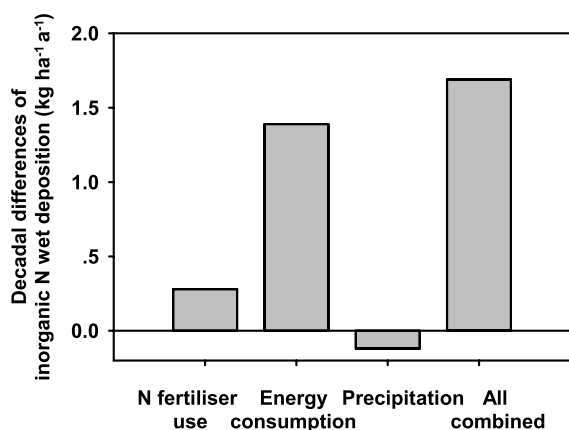

Figure 6 | The effects of N fertiliser use, energy consumption, and precipitation on the decadal variation in N$_{dep}$. The individual effects of these three factors on the decadal variation of N$_{dep}$ was obtained by equation (1) and data on N fertiliser use, energy consumption, and precipitation in provinces in the 1990s and 2000s (see Data and Methods).

fertilisers, soil, animal excreta, and fossil fuel combustion[28]. Based on isotope techniques, Hastings et al. distinguished nitrate from anthropogenic emissions and from lighting in different seasons[29].

In the present study, we used monitoring data and statistical information to demonstrate how anthropogenic sources and precipitation influence the spatial pattern and decadal variation of N_{dep} in China (Fig. 3, 4, and 6). The combination of anthropogenic sources (N fertiliser use and energy consumption) and precipitation contributed 79% of the spatial variation of N_{dep} (equation (1)), and energy consumption contributed approximately 80% of the decadal variation in N_{dep} (Fig. 6). The findings further demonstrated in a new way that anthropogenic sources have played an important role in the increase of N_{dep} and that N_{dep} is primarily influenced by regional human activities. These findings suggest that it is possible for regions to control their N deposition by decreasing their emissions.

Currently, it is generally considered that forest ecosystems have the greatest carbon uptake potential under increasing N deposition[4]. Based on the interpolated results of this study, we evaluated the possible contribution of N_{dep} to forest carbon sequestration in China. For this, we assumed the following: the forest average of N_{dep} was equivalent to the national average over China (11.11 and 13.87 kg ha^{-1} a^{-1} in the 1990s and 2000s); the net ecosystem production (NEP) response to N deposition would be approximately 45 kg C per kg N in forest ecosystems[30–37] (Supplementary Table S2); and the area of forests in China were 146 and 185 million ha (ref. 38) and that these forests may take up an additional 72 and 115 Tg C each year owing to N_{dep}, respectively, in the 1990s and 2000s. According to the results of Gregg et al., CO_2 emissions from fossil fuel combustion and cement manufacturing reached approximately 1500 Tg C a^{-1} in 2006 in China[39], suggesting that forests may have absorbed approximately an additional 7% of anthropogenic CO_2 emissions owing to N_{dep} in the 2000s in China. Because there was no consideration of some factors, such as forest type, forest age, and N saturation, that can influence the response of forest carbon sequestration to N deposition, the above assessment has a large uncertainty. Nonetheless, this preliminary assessment does suggest that anthropogenic N emissions can offset anthropogenic CO_2 emissions to some extent, particularly in China, which has a large forest area and high N deposition.

Our findings are still uncertain to some extent due to the use of derived data, although we conducted rigorous data screening and quality control. There were four plausible contributors to the uncertainty. First, error is involved in the preservation of samples and in measuring instruments. The samples of inorganic wet deposition were obtained from precipitation collected in open rain collectors by different researchers, and some errors were introduced by the researchers using different methods of sample preservation and different measuring instruments. Second, errors may be contributed by the number and distribution of monitoring sites. Due to the lack of an extensive long-term monitoring network, the temporary monitoring sites were limited and distributed unevenly; therefore, uncertainty from the interpolated results was induced by the limited amount and uneven distribution of the monitoring sites. Third, only N fertiliser use and energy consumption were considered as anthropogenic sources. In China, the largest sources of NH_3 and NO_x are due to N fertiliser use and energy consumption[40–42], respectively, which is why we selected these two human activities as anthropogenic sources; however, other N sources, such as livestock and biomass burning, may impact the study results because of regional differences in these sources. Furthermore, some uncertainties are attributed to the data from governmental statistical yearbooks. The data from governmental statistical yearbooks were collected not only from professional statistical departments but also from competent departments of various industries, which may affect the neutrality of data for some subjective needs. Therefore, it is necessary and urgent to establish an extensive long-term monitoring network with consistent quality control in China to reduce the uncertainty associated with N deposition estimates and evaluation of the ecological effects of N deposition.

Methods

Data sources. Datasets on N_{dep} were summarised from published sources for the 1980–2010 period (see Supplementary Information for a complete reference list) and the National Acid Deposition Monitoring Network (NADMN) established by the China Meteorological Administration. After rigorous data screening and quality control, we obtained a total of 620 site-year data for N_{dep}. The contents of our datasets included the following: the name of the monitoring site, location of the monitoring site, monitoring period, monitoring method, ecosystem type, annual precipitation, concentration and deposition of NH_4^+-N, and NO_3^--N, total inorganic N wet deposition, and the literature source. To study the spatio-temporal pattern of N_{dep}, we divided the datasets into three sections according to decade (1980s, 1990s, and 2000s). The mean N_{dep} was calculated per site and decade, which represented the regular status of N_{dep} at the site for the period. After the above integration, the monitoring sites distributed in China numbered 25, 136, and 144 for the 1980s, 1990s, and 2000s, respectively. The number of monitoring sites in the 1980s was insufficient to study the spatial pattern of N_{dep} by geostatistical methods; therefore, we focused on the N_{dep} occurring in the 1990s and 2000s. The monitoring sites were distributed throughout China (Supplementary Fig. S2) and were used to produce maps of N_{dep} for China in the 1990s and 2000s using a geostatistical method.

The data on N fertiliser use (expressed as the volume of effective component) in provinces were obtained from the China Statistical Yearbook[43]. The data on energy consumption (expressed as standard coal) in provinces were predominantly from the China Energy Statistical Yearbook[44], which primarily consists of the consumption of coal, crude oil and their products, and natural gas. Nevertheless, as there were no energy data for 1991–1994 in the national yearbooks, we obtained the energy data for those years from provincial yearbooks. Due to the lack of energy data in Tibet province, we assumed that the per capita energy consumption was similar between the Tibet and Xinjiang provinces, which are both located in western China, and deduced data on energy consumption in Tibet province from the Xinjiang province data (the populations in these two provinces were known). The mean annual N fertiliser use in provinces in the 1990s and 2000s was calculated from annual data for 1990–1999 and 2000–2009, respectively. The mean annual energy consumption in provinces was treated in the same way.

Data on the annual amount of precipitation were obtained from the China Meteorological Administration, comprising 756 weather stations (Supplementary Fig. S3). The mean annual precipitation in provinces in the 1990s and 2000s was calculated based on the annual precipitation between 1990–1999 and 2000–2009, respectively, from the weather stations in each province.

Geostatistical method and cross-validation. A geostatistical method was used to produce spatially continuous estimates from discrete field measurements. For this study, we constructed national-scale N_{dep} maps for the 1990s and 2000s using the Kriging interpolation technique. Kriging is a statistical method of providing unbiased estimates of variables in regions where the available data exhibit spatial autocorrelation, and "Kriging estimates" are obtained in such a way that they have minimum variance[25]. Prior to Kriging interpolation, the Explore Data tool of ArcGIS 10.0 software was employed to conduct a data analysis, including the data's distribution, outlier identification, and trend analyses; the optimal variogram model and parameters were then determined. Because the original data did not follow a normal distribution, we transformed the data through Box-Cox transformation ($\lambda = 0.6$) using the transformation tool in ArcGIS 10.0 software. The normal distribution plots and normal distribution tests are shown in Supplementary Figure S4 and Table S3. Subsequently, the transformed data were used for Kriging interpolation, and the data are back-transformed before the final map was created. Additionally, a cross-validation analysis was implemented to evaluate the results of the Kriging interpolation. In this analysis, every monitoring site was individually removed, and its value estimated via Kriging based on the other surrounding sites was compared to the original observed value. The results of the cross-validation analysis and prediction errors are shown in Supplementary Table S4 and Figure S5, including the mean prediction error, root-mean-square standardised error, root-mean-square error, average standard error, R^2, and regression coefficient. From these parameters, we found that the Kriging method worked well to predict the spatial patterns of N_{dep} in China although the predicted values for some sites were not perfect due to relatively higher root-mean-square error and low R^2. These results indicated that the results of interpolation can express the trend of spatial patterns of N_{dep} in the 1990s and 2000s in China.

The effect of metropolitan area on N_{dep} in natural forest ecosystems. We selected all of the natural forest monitoring sites in the 2000s from our datasets and used the Near tool in ArcGIS 10.0 software to calculate and record the distance between each forest site and the nearest large city with more than 2 million inhabitants. These distances were fitted to the N_{dep} of natural forests by a power model that depended on the distribution of the scatter diagram and the correlation coefficient and P value. These data were analysed using SPSS (Statistical Package for the Social Sciences) 11.0 statistical software. Through the above analysis, we determined the maximum possible distance at which anthropogenic sources influence atmospheric N deposition, which was the basis for studying the relationship between anthropogenic

sources (N fertiliser use and energy consumption) and the spatial pattern of N_{dep} by province in the analyses described below.

The effects of anthropogenic sources and precipitation on the spatial pattern of N_{dep}. Based on our Kriging interpolation, we obtained maps of N_{dep} over China in the 1990s and 2000s with a resolution of 10×10 km. The Zonal Statistics tool of ArcGIS 10.0 software was employed to obtain the mean N_{dep} by province. N fertiliser use, energy consumption, and precipitation in provinces were fitted with the mean N_{dep} using a linear or nonlinear regression model. Subsequently, we analysed the combined effect of these three factors on the spatial variation of N_{dep} in China (equation (1)). It should be noted that Beijing, Tianjin, and Shanghai are three economically developed municipalities with a total area of 34,400 km², less than 0.4% of the national area, but their energy consumption is significantly higher than the other provinces (Supplementary Table S1). Therefore, these three municipalities did not follow the same relationship between energy consumption and N_{dep} (Supplementary Fig. S6) and were excluded when we explored the effects of anthropogenic sources and precipitation on the spatial pattern of N_{dep}. Moreover, due to a lack of data from Taiwan, Hong Kong, and Macao, these three provinces were also not considered. A total of 28 provinces were considered in the analysis; aside from Hainan province, with an area of 34,000 km², the area of every other 27 province is more than 60,000 km² (Supplementary Table S1).

The effects of anthropogenic sources and precipitation on the decadal variation in N_{dep}. We used equation (1) to calculate the individual effects of N fertiliser use, energy consumption, and precipitation on the decadal variation in N_{dep}. Specifically, we quantified the individual effects of a given factor by applying three factors in different decades (1990s or 2000s) as input data for equation (1). The specific method was as follows (using N fertiliser use as an example). First, data on N fertiliser use in provinces in the 1990s and energy consumption and precipitation in the 2000s were input to equation (1), and the mean N_{dep} over China (N1) was then calculated for these parameters; i.e., N fertiliser data were held constant in the 1990s, and data for energy consumption and precipitation taken from the 2000s. Second, data on N fertiliser use, energy consumption, and precipitation in provinces in the 2000s were inputted into equation (1), and the mean N_{dep} over China (N2) was calculated for these parameters. Third, the difference between N2 and N1 was the individual effect of N fertiliser use on the decadal change (1990s–2000s) in N_{dep} over China because this difference was obtained for conditions in which only N fertiliser use was varied.

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Author contributions

Y.J. analysed the data and wrote the manuscript. G.Y. and N.H. supervised the project and commented on the contents of the manuscript. X.Z., H.F., W.S., Y.Z. and Y.J. collected the datasets and conducted the data pre-processing. D.Z. and Q.W. revised and edited the manuscript.

Additional information

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