LETTER

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Enhanced nitrogen deposition over China

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China is experiencing intense air pollution caused in large part by anthropogenic emissions of reactive nitrogen[1,2]. These emissions result in the deposition of atmospheric nitrogen (N) in terrestrial and aquatic ecosystems, with implications for human and ecosystem health, greenhouse gas balances and biological diversity[1,3–5]. However, information on the magnitude and environmental impact of N deposition in China is limited. Here we use nationwide data sets on bulk N deposition, plant foliar N and crop N uptake (from long-term unfertilized soils) to evaluate N deposition dynamics and their effect on ecosystems across China between 1980 and 2010. We find that the average annual bulk deposition of N increased by approximately 8 kilograms of nitrogen per hectare ($P < 0.001$) between the 1980s (13.2 kilograms of nitrogen per hectare) and the 2000s (21.1 kilograms of nitrogen per hectare). Nitrogen deposition rates in the industrialized and agriculturally intensified regions of China are as high as the peak levels of deposition in northwestern Europe in the 1980s[6], before the introduction of mitigation measures[7,8]. Nitrogen from ammonium (NH_4^+) is the dominant form of N in bulk deposition, but the rate of increase is largest for deposition of N from nitrate (NO_3^-), in agreement with decreased ratios of NH_3 to NO_x emissions since 1980. We also find that the impact of N deposition on Chinese ecosystems includes significantly increased plant foliar N concentrations in natural and semi-natural (that is, non-agricultural) ecosystems and increased crop N uptake from long-term-unfertilized croplands. China and other economies are facing a continuing challenge to reduce emissions of reactive nitrogen, N deposition and their negative effects on human health and the environment.

Atmospheric N deposition results from emissions of reactive nitrogen (N_r) species and their atmospheric transport; it expands the footprint of local alterations to the N cycle[3]. Although both natural and anthropogenic sources contribute to atmospheric N deposition, anthropogenic N_r emissions (largely from the agricultural, industrial and transport sectors) have increased substantially since the industrial revolution began[9]; they now make the dominant contribution to N deposition in many regions[3,10]. Increased concentrations of N_r in the atmosphere and, through deposition, in terrestrial or aquatic ecosystems, or both, degrade human health[1] (notably through driving the formation of particulate matter and tropospheric ozone), alter soil and water chemistry[3], influence greenhouse gas balance[4] and reduce biological diversity[5].

The human and environmental costs associated with anthropogenic N_r are well recognized, and active measures in Western Europe and North America have stabilized or reduced N_r deposition in those regions[6,11,12]. Even so, very large costs of excess N_r have been reported in the European Union[8] (€70–320 billion per year) and the United States[13]. In contrast, over the past 30 years China's emissions have increased to the point that it has become by far the largest creator and emitter of N_r globally[2]. However, the rates and trends of N deposition in China since the 1980s are not clear. We would also like to know the consequences of N deposition, for the people and ecosystems of China, its region and the world.

Following rapid economic growth since the early 1980s, China's gross domestic product was estimated at US$5.9 trillion in 2010, making China the world's second largest economy after the United States (http://money.cnn.com/news/economy/world_economies_gdp). In around the year 2000, China surpassed the United States and the European Union (combined) in its production and use of N fertilizers. Moreover, less than half of the fertilizer N applied in China is taken up by crops[14]; the rest is largely lost to the environment in gaseous (NH_3, NO, N_2O and N_2) or dissolved (NH_4^+ and NO_3^-) forms[15,16]. These fluxes—along with N_r emitted during fossil fuel combustion—have resulted in some of the most pronounced air pollution on Earth[1].

Increased N_r emissions must have influenced atmospheric N deposition in and near China, but information on the magnitude, scope and consequences of any change has been lacking. Here we summarize available data nationwide on the bulk deposition of N_r in terrestrial ecosystems. Also, we show that the N cycle has been altered in Chinese ecosystems, both within and outside croplands.

Nitrogen deposition includes wet and dry deposition of both inorganic and organic N forms[2,17], but in most cases only the bulk deposition of inorganic N (NH_4-N and NO_3-N) has been measured systematically[6,18,19]. Bulk N deposition denotes N input from precipitation as measured by an open sampler (Supplementary Methods); it is a relatively simple measure that includes wet deposition and a fraction of the dry deposition, and it is suitable for regional comparisons. We constructed a national data set incorporating all the available bulk N deposition results from monitoring sites throughout China between 1980 and 2010 (Supplementary Fig. 1). This data set was used to test the magnitude and trend of atmospheric N deposition in relation to anthropogenic emissions of reduced and oxidized forms of N.

In spite of site-to-site variability in the data, bulk N deposition increased significantly with time ($P < 0.001$), with an average annual increase of 0.41 kilograms of nitrogen per hectare (kgN ha^{-1}) between 1980 and 2010 (Fig. 1a and Supplementary Table 1). The increase in bulk N deposition was driven mainly by increased volume-weighted N concentrations in rain water (0.063 mgN l^{-1} yr^{-1} on average; Fig. 1b) because annual precipitation in the study area has not changed significantly in the past 30 years (Supplementary Fig. 2 and Supplementary Table 1). NH_4-N was the dominant form in bulk deposition, but the ratio of NH_4-N to NO_3-N in bulk precipitation decreased significantly with time (Fig. 1c, Supplementary Fig. 3 and Supplementary Table 1). Overall, annual bulk N deposition averaged 13.2 and 21.1 kgN ha^{-1} in the 1980s and 2000s, respectively, showing an increase of approximately 8 kgN ha^{-1}, or 60% ($P < 0.001$).

The increase in overall bulk N deposition and the change in the ratio of NH_4-N to NO_3-N in precipitation and deposition (Fig. 1) are similar to the increasing trends of anthropogenic gaseous N_r (NH_3 and NO_x) emissions and changes in their ratio since 1980 (Fig. 2a and Supplementary Fig. 4a). The ratio of NH_4-N to NO_3-N in measured

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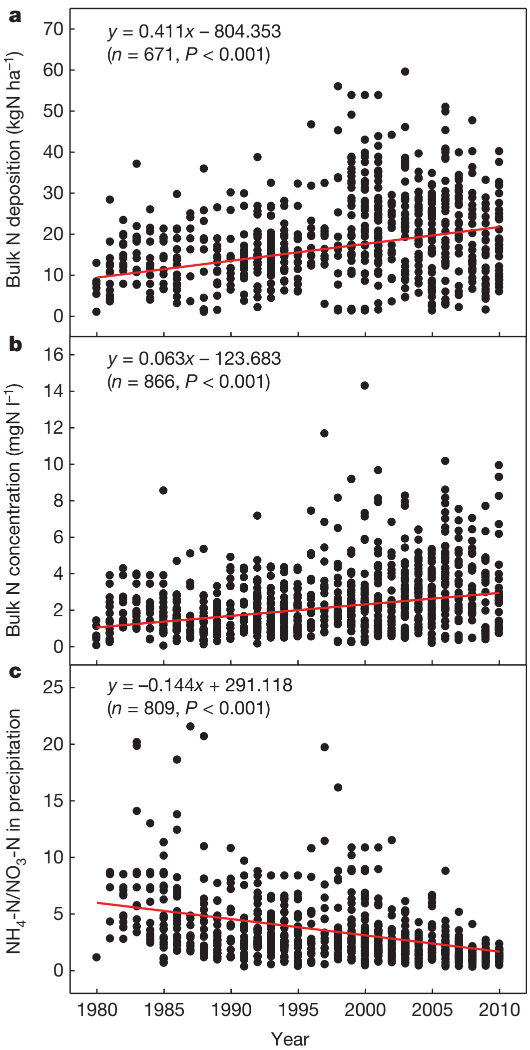

Figure 1 | Trends in N deposition and its components in China between 1980 and 2010. **a**, Bulk N deposition; **b**, bulk N concentration; **c**, ratios of NH_4-N to NO_3-N in bulk precipitation. In spite of large site-to-site variability, both bulk N deposition and N concentration have increased significantly since 1980, and the ratio of NH_4-N to NO_3-N in bulk precipitation has decreased significantly according to linear mixed models (all $P < 0.001$; Supplementary Table 1). Data sources are included in Supplementary Information.

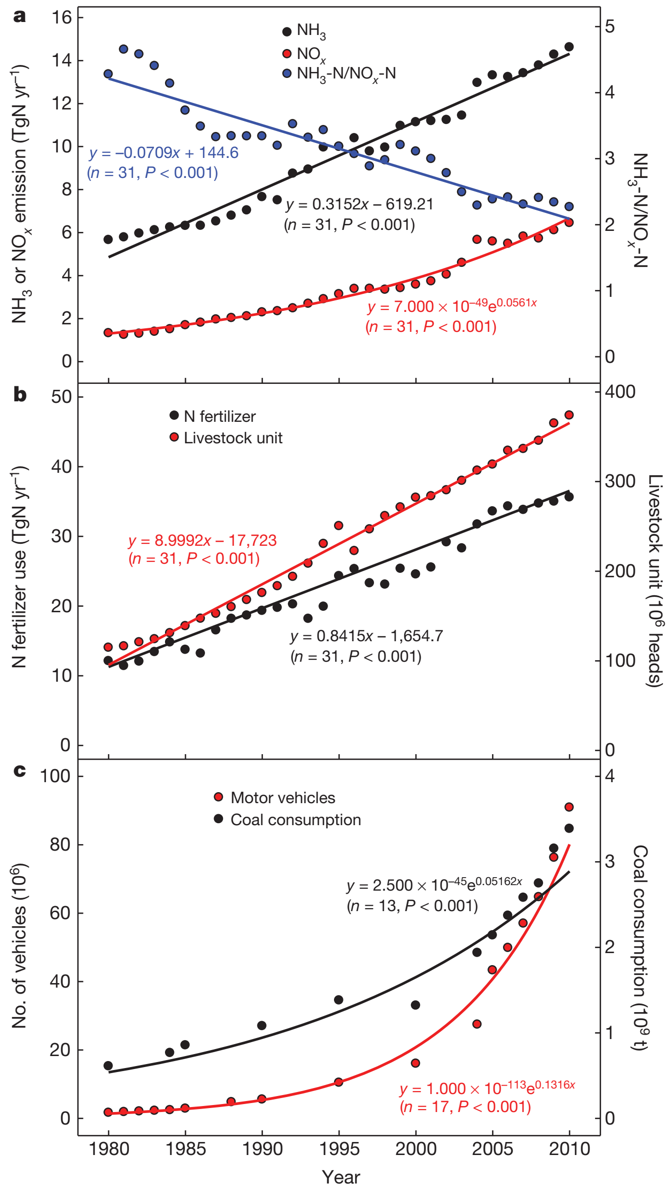

Figure 2 | Trends in NH_3 and NO_x emissions and their main contributors between 1980 and 2010. **a**, NH_3 and NO_x emissions and ratios of NH_3-N to NO_x-N emission; **b**, number of domestic animals (expressed as livestock units) and N fertilizer consumption; **c**, number of motor vehicles and coal consumption. Data sources are cited in Supplementary Information.

bulk deposition decreased from about 5 to 2, and the ratio of NH_3-N to NO_x-N in calculated emissions decreased from about 4 to 2.5; these changes are highly correlated ($P < 0.01$). Emissions of NH_3 doubled (Fig. 2a), reflecting increased agricultural production in that both the use of N fertilizer and the number of domestic animals (expressed as standard livestock units) have also doubled since the 1980s (Fig. 2b and Supplementary Fig. 4b). Fossil fuel power plants, industrial production and motor vehicles are the major sources of NO_x in China and Asia[20]. Coal consumption and the number of motor vehicles increased 3.2- and 20.8-fold, respectively, between the 1980s and the 2000s (Fig. 2c and Supplementary Fig. 4c), driving a more rapid percentage increase in NO_x emission than in NH_3 emission (Fig. 2a), although the net increase in emission was still larger for NH_3 than for NO_x (about 6 TgN versus 4 TgN between the 1980s and the 2000s). The ratio of NH_4-N to NO_3-N in bulk precipitation (Fig. 1c) changed in the same direction and by approximately the same magnitude as the ratio of NH_3-N to NO_x-N emission over the same period (Fig. 2a), despite uncertainties in ammonia emission inventories[2,21].

We analysed the dynamics of bulk N deposition regionally by dividing deposition data into six areas: northern, southeast, southwest, northeast and northwest China and the Tibetan plateau. Human influences on the N cycle differ substantially among these regions. In general, bulk N deposition showed increasing trends and ratios of NH_4-N to NO_3-N showed decreasing trends for all six regions between the 1980s and the 2000s (Supplementary Table 1); highly significant ($P < 0.001$) increases in bulk N deposition were found in northern, southeast and southwest China, significant ($P < 0.05$) increases were found in the Tibetan plateau and northeast China, and no significant ($P = 0.199$) increase was found in northwest China (Supplementary Figs 5 and 6). By comparison with the national average, we found both higher overall rates of deposition and higher annual rates of increase in deposition in the industrialized and agriculturally intensified northern, southeast and southwest China. Annual bulk deposition rates were 22.6, 24.2 and 22.2 kgN ha^{-1} in northern, southeast and southwest China in the 2000s, respectively, with average rates of increase of 0.42, 0.56 and 0.53 kgN ha^{-1} yr^{-1}. A more detailed study[22] of all major deposition pathways shows that total annual N wet and dry deposition on the northern China plain (the central area of northern China) was about 80 kgN ha^{-1}. These levels are much higher than those observed in any region in the United

States[12], and are comparable to the maximum values observed in the United Kingdom[6] and the Netherlands[7] when N deposition was at its peak in the 1980s[8].

Extensive long-term environmental inventories and experiments in China allow us to evaluate some of the consequences of this substantial and continuing increase in N deposition. We have summarized results of an ongoing survey of foliar N concentrations from non-agricultural ecosystems throughout China and from detailed studies of crop N uptake from croplands in long-term trials without N fertilizer (described as zero-N plots hereafter), which are used as reference plots in fertilization experiments. The foliar N data set provides information on how changes in N deposition have influenced plant tissue chemistry in unfertilized, non-agricultural ecosystems. Foliar N increased significantly (all $P < 0.001$) between the 1980s and the 2000s for woody, herbaceous and all plant species (Fig. 3a, Supplementary Fig. 7a and Supplementary Table 1). Foliar N increase for all species averaged 32.8% (24.0 ± 9.2 mg g^{-1} (2000s) versus 18.1 ± 7.2 mg g^{-1} (1980s)), with a higher increase in herbaceous plants than in woody plants (Fig. 3a). In contrast, foliar phosphorus (P) did not change significantly ($P = 0.085$) over the same period (Supplementary Fig. 7b and Supplementary Table 1). Foliar N is largely determined by plant species and plant N nutritional status; foliar N of specific plant species should be stable in natural and semi-natural ecosystems unless some process changes the availability of N relative to other plant resources[23].

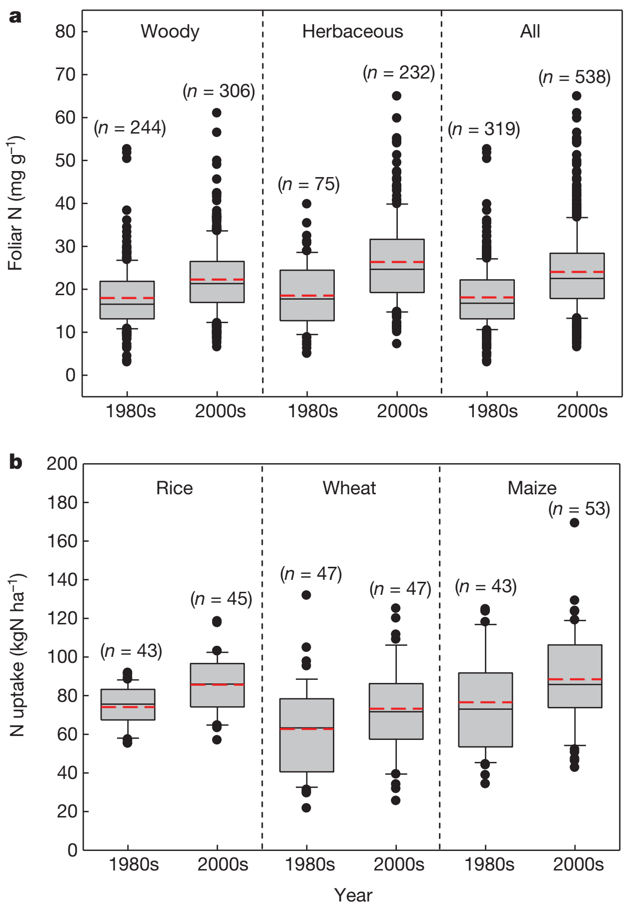

Figure 3 | Comparisons of foliar N concentrations and crop N uptake between the 1980s and 2000s. **a**, Foliar N in woody, herbaceous and all plant species in non-agricultural ecosystems; **b**, N uptake by rice, wheat and maize from zero-N plots in long-term experiments. Both foliar N (all $P < 0.001$) and N uptake (all $P < 0.05$) are significantly higher in the 2000s than in the 1980s. The black and red lines, lower and upper edges, bars and dots in or outside the boxes represent median and mean values, 25th and 75th, 5th and 95th, and $<$5th and $>$95th percentiles of all data, respectively.

Plant species in this study were sampled widely (Supplementary Fig. 1) and analysed by standard procedures (Supplementary Information)—and the evaluation of foliar P should correct for any bias towards sampling high-nutrient plant species late in the record (suggesting no apparent changes in the soil environment)—so the increase in foliar N in unfertilized ecosystems most probably represents a widespread increase in plant N nutritional status caused by the cumulative effects of enhanced N deposition.

In agricultural ecosystems, crop N uptake from zero-N plots in long-term experiments is controlled primarily by N deposition, because soil N pools are relatively stable after 5 to 10 years without N fertilization[22,24]. We summarized the available data on N uptake from zero-N plots in long-term experiments between 1980 and 2010 (Supplementary Information); N uptake by rice, wheat and maize in zero-N plots was significantly higher in the 2000s than in the 1980s (Fig. 3b; all $P < 0.05$). The increase in N uptake averaged 11.3 kgN ha^{-1} across the three major cereals (Fig. 3b and Supplementary Fig. 8).

Overall, the temporal patterns in bulk N deposition, foliar N and N uptake from zero-N plots are consistent with rapidly increased anthropogenic NH_3 and NO_x emissions over the past three decades. The lower ratio of reduced N to oxidized N in measured deposition agrees well with the decrease in the ratio of calculated emissions of NH_3 to NO_x, reflecting a more rapid proportional increase in N_r emissions from industrial and traffic sources than from agricultural sources. All these changes can be linked to a common driving factor, strong economic growth, which has led to continuous increases in agricultural and non-agricultural N_r emissions and, consequently, increased N deposition.

Although we did not measure the impact of atmospheric N_r emissions and deposition from China on the global environment, recent studies indicate that N_r deposited by China may be moving to surrounding marine ecosystems[25] and perhaps to tropical and subtropical forests[26]. Another study[27] reported a strong abnormal spring increase in free tropospheric ozone concentrations in western North America between 1995 and 2008, and suggested that NO_x-induced ozone transport from Asia (mainly from China and India) to North America could be a major source.

Clearly, N deposition has increased significantly in China and has affected both non-agricultural and agricultural ecosystems. So far, China's economic growth model has relied mainly on the consumption of raw materials, and it has caused large anthropogenic N_r emissions in addition to other environmental perturbations[28]. For example, the emitted NH_3 and NO_x gases form secondary aerosols such as NH_4NO_3 in $PM_{2.5}$ (particulate matter with aerodynamic diameter $\leq$2.5 μm) under favourable conditions, decrease visibility and damage human health. The Chinese government has recognized the importance of protecting the environment while developing the economy; recently, it approved the first national environmental standard for limiting the amount of $PM_{2.5}$ (ref. 29).

Our results demonstrate that deposition of reduced forms of N_r continues to be of greatest importance in China (which is responsible for approximately 2/3 of total deposition) but emission and deposition of oxidized N_r are increasing more rapidly. Current environmental policy needs to focus more strongly on reducing present NH_3 emissions from agricultural sources, whereas control of NO_x emissions from industrial and traffic sources will become more important in the near future. It is time for China and other economies to take action to improve N-use efficiency and food production in agriculture and reduce N_r emissions from both agricultural and non-agricultural sectors. These actions are crucial to reducing N deposition and its negative impact locally and globally.

METHODS SUMMARY

Data sets on bulk N deposition, plant foliar N concentration and crop N uptake from non-N-fertilized soils were summarized from published data and measurements across China. Using 315 references and our own deposition monitoring network (CAUDN), we constructed a nationwide data set of the amount of annual precipitation, volume-weighed N concentrations in precipitation and bulk N deposition, as well as the ratio of NH_4-N to NO_3-N deposition. A total of 866 data points of annual volume-weighted N concentrations in precipitation and 671 data points of annual bulk N deposition rates at 270 monitoring sites were summarized for the period 1980–2010. To clarify regional variations, bulk N deposition data from six separate regions were also summarized. Additionally, a total of 981 observations of plant foliar N concentration and 859 observations of foliar P were collected from 666 natural and semi-natural terrestrial plant species or varieties at 245 sites distributed across the whole of China (based mainly on ref. 30; Supplementary Fig. 1), and a total of 278 data points of crop N uptake by rice, wheat and maize were summarized from non-N-fertilized soils in long-term experiments. Emissions of national anthropogenic NH_3 and NO_x were summarized from published data[2] and updated to 2010 (Supplementary information).

Data on N deposition, foliar N and crop N uptake, and other related parameters, were fitted (with year) using linear mixed models or nonlinear regression models for the interval 1980–2010 (SPSS13.0, SPSS Inc.). Differences in these data between the 1980s (1980–1989) and the 2000s (2000–2010) were compared statistically using an unpaired two-tail Student's t-test. A significant difference is assumed when the P value is <0.05 or as otherwise stated. Further details on the data sets and statistical methods are given in Supplementary Methods.

Full Methods and any associated references are available in the online version of the paper.

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Supplementary Information is available in the online version of the paper.

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Author Contributions X.L. and F.Z. designed the research. X.L., Y.Z., W.H., A.T., J.S. and Z.C. conducted the research (collected the data sets and analysed the data). X.L., Y.Z. and P.V. wrote the manuscript. J.W.E., K.G., P.C., A.F. and F.Z. commented on the manuscript.

Author Information Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to F.Z. (zhangfs@cau.edu.cn).

METHODS

Data sources for bulk N deposition. Bulk N deposition data are from two sources: monitoring results from a regional atmospheric deposition monitoring network (that is, the China Agricultural University-organized Deposition Network (CAUDN)); and results published during the period 1980–2010. Only bulk deposition data for inorganic N (NH_4-N and NO_3-N) were summarized in this study because no dry deposition data for N were reported in China in the 1980s and 1990s[2]. Our data sets include year of monitoring at every site; location of every monitoring site; annual amount of precipitation; concentration and deposition of NH_4-N, NO_3-N and total inorganic N (TIN); and ratios of NH_4-N to NO_3-N concentration and deposition in precipitation. Some sites that contained only portions of deposition data (that is, only concentration or deposition of inorganic N) were also included in our data sets. Briefly, bulk N deposition samples from the CAUDN were collected using always-open rain gauges (different from wet-only samplers) on a daily basis and measured by colorimetry (that is, continuous flow analysis) or ion chromatography. For literature deposition data, these are the two most common methods for measuring inorganic N (NH_4-N and NO_3-N) concentrations in precipitation (for details, see references in Supplementary Table 2). Bulk deposition rates of NH_4-N, NO_3-N and TIN were then calculated by multiplying N concentration in precipitation by the amount of precipitation[19].

In this paper, we summarize 866 data points of N concentrations in rainwater and 671 data points of bulk N deposition rates from between 1980 and 2010. All of the data originated from publications and our own results from the CAUDN. In all, we collected 315 references on annual N concentration and deposition results (including 276 journal articles, 19 dissertations and 20 monographs) (Supplementary Table 2) in this data set, covering 270 monitoring sites widely distributed in China (Supplementary Fig. 1). The current bulk N deposition data sets are the most complete deposition data sets in China in spite of some minor weaknesses (that is, relatively fewer monitoring sites and data points in northeast China, northwest China and the Tibetan plateau). Therefore, our meta-analysis based on the data set should be reliable. The same results published in different sources (that is, journals, dissertations or monographs) were cited only once and only one reference source was listed in the following priority: English-language journals, Chinese-language journals, dissertations and monographs.

To clarify regional variations, bulk N deposition data were also summarized on a regional basis (Supplementary Fig. 1): northern China, comprising Beijing, Tianjin, Hebei, Henan, Shandong, Shanxi and Shaanxi provinces; southeast China, comprising Shanghai, Jiangsu, Zhejiang, Anhui, Hubei, Hunan, Jiangxi, Fujian, Guangdong, Hong Kong, Macau, Taiwan and Hainan provinces; southwest China, comprising Sichuan, Chongqing, Guizhou, Yunnan and Guangxi provinces; the Tibetan plateau, comprising Tibet and Qinghai provinces; northeast China, comprising Liaoning, Jilin and Heilongjiang provinces; and northwest China, comprising Xinjiang, Inner Mongolia, Ningxia and Gansu provinces.

Data sources for plant foliar N from non-agricultural vegetation types. A total of 981 observations of plant foliar N content and 859 observations of foliar P were collected from 666 natural and semi-natural terrestrial plant species (including woody and herbaceous species, non-N-fixing and N-fixing species, and evergreen and deciduous species, according to various classification methods) between 1980 and 2010 at 245 sites distributed across the whole of China (Supplementary Fig. 1), on the basis of our field measurements and the literature (for details, see references in Supplementary Table 3). Leaves were sampled mainly during the growing season (July to September). Leaf samples were oven-dried, ground and then measured for N concentrations using the Kjeldahl method. To avoid systematic deviation caused by chemical determination, N samples determined with C and N elemental analysers[30] (after the year 2000) were not included in our analysis. For the few leaf samples lacking detailed time records, the sampling year was assumed to be two years before the associated paper was first submitted (for example, the sampling year was assumed to be 2004 if the paper was submitted in 2006). Mean foliar N was calculated for each species at the same sites within the same sampling year.

Data sources for crop N uptake from zero-N croplands. A total of 278 data points of crop N uptake were collected from non-N-fertilized (zero-N fertilizer input for at least five years) croplands (described as 'zero-N plots' hereafter) during the 1980s and 2000s across China, on the basis of our field experiments and published data[22,31–37]. Nitrogen uptake by rice, wheat and maize includes N accumulation in grain plus straw at harvest (normally from May to October) of the three main cereal crops on zero-N plots. Grain and straw samples were oven-dried, ground and measured for N concentrations using the Kjeldahl method when the harvest process was completed in the field. Nitrogen accumulation in grain or straw was calculated as N concentration multiplied by grain or straw dry matter; crop N uptake was then the sum of grain and straw N accumulation. For a few publications that did not provide crop N uptake data, we used conversion coefficients[38] of grain yield for estimating N uptake by rice, wheat and maize, respectively.

Data sources for anthropogenic NH_3 and NO_x emissions and their main contributors. NH_3 and NO_x (sum of NO and NO_2) emission inventories in China during 1980 and 2010 were obtained from all published data available[2] and updated to 2010 on the basis of data from the National Bureau of Statistics of China (http://www.stats.gov.cn/english/statisticaldata/yearlydata/) and the reported NH_3 and NO_x emission factors[39]; if several emission values were available in one specific year only, an averaged emission value was used. Briefly, China's national emission inventories for NH_3 and NO_x were based on different emission sources and emission factors of specific N_r species. Compared with the NO_x emission inventory (mainly point sources), the NH_3 emission inventory (mainly non-point-source emission) has a relatively large uncertainty[2,21]. The ratios of NH_3-N to NO_x-N emission were then calculated on the basis of averaged annual emission data over the period 1980–2010.

Data on N fertilizer use and domestic animal numbers (expressed as livestock units) were from Chinese Agriculture Statistics (1982–2010). The transformation of domestic animal numbers to livestock units was based on some widely used conversion factors in Europe (http://epp.eurostat.ec.europa.eu/statistics_explained/index.php/Glossary:LSU). Data on coal consumption (as standard coal) and motor vehicle numbers were from the National Bureau of Statistics of China (http://www.stats.gov.cn/english/statisticaldata/yearlydata/).

Statistical analysis. Annual precipitation; N concentration in precipitation; bulk N deposition; ratios of NH_4-N to NO_3-N in bulk precipitation; foliar N, foliar P and crop N uptake from zero-N plots; NH_3 and NO_x emissions and ratios of NH_3-N to NO_x-N emission; N fertilizer use; and numbers of domestic animals, numbers of motor vehicles and coal consumption in China were fitted (with year) by linear mixed models or nonlinear regression models for the interval 1980–2010. We used mixed models[40,41] instead of simple linear regressions because of the large site-to-site variability. The selection of linear versus nonlinear regression depended on the distribution of the 'scatter diagram' (initially judging the temporal variation followed by a linear or nonlinear trend) and on the correlation coefficients (r) and P values in the linear or nonlinear regression equations[41]. Correlation coefficients were tested by a statistical model (SPSS 13.0, SPSS Inc.). Differences between all of the above-mentioned parameters as measured in the 1980s (1980–1989) and the 2000s (2000–2010) were compared statistically using an unpaired two-tail Student's t-test. Significant difference is assumed when the P value is <0.05 or as otherwise stated.

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