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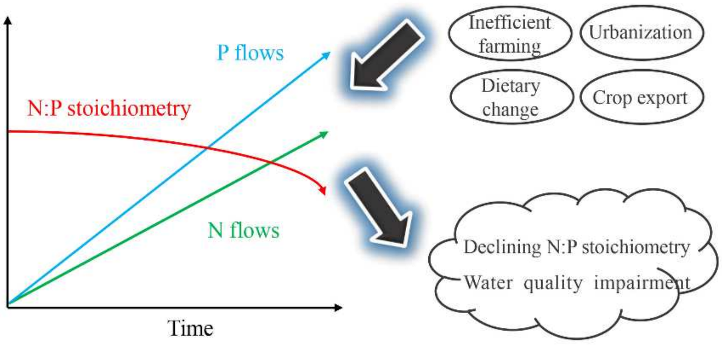
P flows
N:P stoichiometry
N flows
Time
Inefficient farming
Urbanization
Dietary change
Crop export
Declining N:P stoichiometry
Water quality impairment

There are 7114 words in the manuscript.

Enhanced nitrogen and phosphorus flows in a mixed land use basin: Drivers and consequences

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Abstract

Rapid increase in accumulation of phosphorus (P) relative to nitrogen (N) has been observed in human-impacted regions, but the reasons are largely unknown. We developed an Integrated Nutrient Flow Analysis (INFA) model in order to assess the changes in nutrient flows of the Chaohu Lake basin from 1978 to 2015. Results show that the increase in total N input is slower than that of P (3.5-fold versus 4.2-fold) during 1978-2015, while total N loss increases much faster than that of P (3.1-fold versus 2.3-fold). We found a decline trend in the N:P ratio of nutrient input and accumulation since the mid-1990s. The decline in N:P ratio of nutrient loss to waterbodies in the basin is correlated ($p<0.05$) with TN:TP of water concentration in Chaohu Lake, which may be related to the frequent algal blooms in the P-limited lake by supplying more P than N. Using an extended STIRPAT model, we found that nutrient use efficiency, urban rate, diet choice and population are key factors driving the change in nutrient flows, which contribute over 90% to the total change. This study confirms that human activities decrease N:P in regional environment and demonstrates the importance of P management to balance nutrient for improving water quality. The method in this study has a wide application for many other mixed land use regions to address nutrient flows imbalance problems and to explore nutrient management options.

Keywords: nutrient flow; N:P stoichiometry; mixed land use basin; basin nutrient management; substance flow analysis

1 Introduction

Human activities have dramatically accelerated the biogeochemical cycles of nitrogen (N) and phosphorus (P) especially over the last seven decades (Beusen et al., 2016), as a result of agricultural intensification, which relies on heavy inputs of N and P fertilizer (Powers et al., 2016). Increase in N and P availability has brought positive effects by boosting food production to feed more population (Tilman et al., 2011), but result in negative impacts on environment, i.e. excess N and P losses to surface waters, cause nuisance and harmful algal blooms, and thus impair aquatic ecology, biodiversity and water quality (Jarvie et al., 2015; Paerl and Otten, 2013). However, previous studies have shown that rapid increase in anthropogenic N inputs relative to P inputs is a widespread phenomenon at global scale (Peñuelas et al., 2012), as the sources of N are geographically widespread than P, and are also more mobile. As most terrestrial biomes have been proved to be N limitation (Camenzind et al., 2017), more N fertilization may be shifting this limitation, imposing negative effects on ecological diversity and climate change (Dashuan et al., 2016; Wieder et al., 2015). In contrast, in human-impacted areas, faster accumulation rates of P than N were observed in soils, lakes and streams (Peñuelas et al., 2012). For example, a recent study suggested that P accumulates faster than N in human-impacted freshwater ecosystems (Yan et al., 2016). It may exacerbate the impairment of water quality by altering the balance of N and P in receiving waters over long time scales, as P is the limiting nutrient for algal growth in freshwater ecosystems (Schindler et al., 2016). However, how humans influence the N and P flows and N:P stoichiometry is largely unknown in high-human impacted areas. To answer this question, it is necessary to examine historical changes in regional N and P flows from an integrated perspective.

Building a regional nutrient budget is an effective way to assess nutrient flows from different sources. Regional Phosphorus Flow Analysis (RPFA) is a substance flow analysis (SFA) based approach that facilitates quantitative evaluation on anthropogenic P cycles in socio-ecosystems. The PRFA was applied to basin and country scales and has been proved to be robust for assessing historical changes in P flows (Jiang and Yuan, 2015; Liu et al., 2016b). However, none of these studies take N into consideration to provide an integrated understanding of regional N and P flows. Recently, a mass-balance based modeling approach has been developed and applied to estimate N and P inputs to the northeastern United States and the southeastern Canada (Goyette et al., 2016; Hale et al., 2013). However, the studies only consider N and P inputs and do not provide understanding of nutrient flows from loss perspective, which is closely linked to water quality impairment. Hence, our ability to have a comprehensive understanding of the changes in N and P flows from both inputs and outputs perspective is still limited.

This study addresses the question “how human beings reshape regional N flows, P flows and N:P stoichiometry and what the drivers and consequences of the change are” by following steps shown in Figure 1a. We first developed an Integrated Nutrient Flow Analysis (INFA) model and applied it to examine the changes in N and P flows and their stoichiometry in a basin of mixed land use (Chaohu Lake Basin, CLB) over the period of time from 1978 to 2015. Then, we used an extended STIRPAT model to quantitatively assesse the drivers of the changes. At last, we analyzed the relationship between the changes in N:P of nutrient loss and TN:TP concentration in waterbodies to demonstrate the potential consequence of the changes. The contribution of this paper lies in the adaption of the P-specific RPFA model into an integrated version for both N and P. The application of the adapted model in the CLB helps to demonstrate the changes in the N and P flows and their stoichiometry in the mixed land use basin. Furthermore, this paper performed the output of the integrated model in an extended STIRPAT model to analyze the drivers of the human-induced changes in N and P flows. The results are expected to support policy makers to design appropriate nutrient management measures for the CLB.

2 Material and methods

2.1 Study area

The CLB is located in the downstream area of the Yangtze River Basin in eastern China (30°87′-32°13′N, 116°40′-118°37′E, Figure 2), covering a total area of 13959 km^2. The basin falls within the boundaries of eight administrative regions (Hefei, Feidong, Feixi, Chaohu, Wuwei, Luajing, Shucheng and Hanshan). Chaohu Lake lies to the northwest of the Yangtze

River, with an area of 760 km^2 and a storage capacity of 2.1 billion m^3 (at a water level of 8 m). Chaohu Lake is connected to the Yangtze River through the only outlet of the lake, the Yuxi River. Outflow from the lake has been artificial controlled since a dam was constructed in1962. There are 32 rivers flowing into the Lake, and four of them, named Nanfei, Pai, Hangbu-Fengle and Baishitian are the most important, accounting for 90% of total water inflow.

The CLB is of particular interest because this basin is located within a major agricultural area in eastern China and encompasses the capital of Anhui Province (named Hefei), which has undergone a rapid urbanization (Figure S1). As an important agricultural area, the land use of the CLB is dominated by cropland, followed by forest, accounting for 65.9% and 14.3% of the total area, respectively (Figure 2). However, there has been large-scale land use change in the basin: Over the last 30 years, urban land area has increased by 6-fold to 1168 km^2 (Figure S1), with a corresponding rapid increase in population density, reaching 659 capita per km^2. Due to the increasing population, the per-capita water-resources availability in the basin has declined to 784 m^3, which is below the internationally-recognized water storage warning threshold (1000 m^3 capita^{-1}) (Pimentel et al., 1997). Meanwhile, since the 1980s, Chaohu Lake, the important water source for surrounding cities, has changed from oligotrophic to eutrophic, and is now one of the three most eutrophic lakes in China (Duan et al., 2017). Thus, it is a critical to quantify the changes in N and P flows within the CLB as a basis for mitigating eutrophication.

2.2 Estimation of nutrient flows

We developed an Integrated Nutrient Flow Analysis (INFA) model and applied it to quantify the N and P flows in the CLB over the past four decades. The INFA divided the socio-ecosystem into seven compartments: Crop farming, animal breeding, food processing, human consumption, waste disposal, loss to environment, and nutrient exchange through trade (Figure 1b). Compared with the RPFA, the INFA considers some N-specific processes to estimate exchange of N between land and atmosphere. For example, we included biological N fixation by legumes crops and denitrification from fertilizer and manure. Specifically, the INFA considered four N and P inflows to the basin: (1) atmospheric deposition, (2) biological N fixation, (3) chemical fertilizer, (4) food/feed import; 6 outflows: (5) crop food export, (6) animal food export, (7) NH_3, N_2 and N_2O emission from fertilizer applied to the cropland and from manure storage; wind erosion for P, (8) erosion and runoff from cropland, (9) discharge to water via wastewater, (10) waste accumulation, and (11) soil accumulation; and 10 nutrient flows between compartment: (12) local crop animal feed (13) crop products, (14) animal products, (15) food consumption, (16) non-recycled crop straw, (17) non-recycled animal manure, (18) non-edible part of food, (19) human wastes, (20) straw and manure recycled to cropland and (21) straw recycled as feed. Here, we defined the flow (7), (8) and (9) as loss from the basin. All the calculations are based on the mass balance principle:

$$\sum_{1}^{n} In_i = \sum_{1}^{m} Out_j + \sum Stock$$

Detailed calculation methods of the flows can be found in the Supplementary Material (SM).

2.3 Assessment on drivers of changes

In this study, the STIRPAT model was used to assess the contributions of the factors to nutrient inputs. STIRPAT is a stochastic model for assessing the effect of humans on the environment, derived from IPAT (Impact=Population×Affluent×Technology) model (Dietz and Rosa, 1994; York et al., 2003). STIRPAT has been widely used to assess the anthropogenic driving forces on greenhouse gas emission and nutrient inputs (Cui et al., 2013; Wang et al., 2013). The standard STIRPAT model is:

$$I = aP^b A^c T^d e \qquad (1)$$

where a is the constant, e is the error term, and b, c, and d are the exponents of P, A and T, respectively.

As the standard STIRPAT model is a nonlinear multivariate equation, it is difficult to calculate the coefficients of a, b, c, d, and e. In the typical application, all the variables in Eq. (1) are often converted to logarithmic form to facilitate the calculation (York et al., 2003). We then obtained the Eq. (2):

$$\ln I = a + b\ln P + c\ln A + d\ln T + e \tag{2}$$

where, a and e are ln of a and e in Eq. (1).

Here, we selected seven factors population, diet choice, urban rate, crop-food export/import, animal-food export/import, nutrient use efficiency of crop farming (NUE_c) and animal farming (NUE_a). Diet choice was defined as the ratio of animal-food to total. Crop- and animal-food export/import was defined as the ratio of exported/imported crop- and animal-food to total. NUE_c and NUE_a were defined as the ratio of nutrient converted to animal- and crop-food to total. Diet choice (A_1), urban rate (A_2) and crop- and animal-food export/import (A_3 and A_4) were used as proxies for societal affluent. NUE_c (T_1) and NUE_a (T_2) were used as proxies for technology.

By adding these factors in equation (2), we obtained equation (3):

$$\ln I = a + b\ln P + c_1\ln A_1 + c_2\ln A_2 + c_3\ln A_3 + c_4\ln A_4 + d_1\ln T_1 + d_2\ln T_2 + e \tag{3}$$

In this study, we used the ordinary least squares (OLS) regression to evaluate the the variance inflation factors (VIFs) of the variables. Based on previous studies, a VIF exceeding 10 usually means that there is an obvious multicollinearity among the variables (Marquaridt, 1970). In such situation, the OLS regression analysis is not suitable for the calculation of coefficients, and the ridge regression analysis is a better choice to overcome the risk of multicollinearity (Hoerl and Kennard, 1970). The ridge regression is an improved OLS regression methods (Eq. 4), which uses a variable coefficient (λ) to improve the stability of regression coefficient estimations.

$$y = \boldsymbol{X}\boldsymbol{\beta} \rightarrow \boldsymbol{\beta}(\lambda) = (\boldsymbol{X}^T\boldsymbol{X} + \lambda\boldsymbol{I})\boldsymbol{X}^T y \qquad (4)$$

Ridge regression in this study was performed in R language using “glmnet” package, which provides automatic screening of the best estimated λ (Friedman et al., 2010).

2.4 Data sources

The N and P flows for the basin from 1978 to 2015 were calculated year-by-year based on 8 administrative division survey data (Table 1). Basic data, including population, fertilizer use, crop yield, sown areas, number of animal sales, year-end animal and food consumption, were derived from local governmental yearbooks and bulletins (APBS, 1989-2016; CMBS, 1979-2016; HMBS, 1979-2016; LMBS, 1979-2016; MMBS, 2012-2016; WMBS, 2012-2016). Variables like nutrient contents of harvested crops, straws and animal products; nutrient excretion for each animal category; and N and P loss factors in crop farming, animal breeding and waste disposal were sourced from literature data and field investigation.

3 Results and discussion

3.1 Changes in input and loss

3.1.1 Patterns of N input and loss

At the basin scale, total N input (TNI) to the CLB increased 3.5-fold, from 62 Gg N (equivalent to 133 kg-N ha^{-1} yr^{-1}) in 1978 to 214 Gg N (equivalent to 503 kg-N ha^{-1} yr^{-1}) in 2015 (Figure 3a). We found a strong linear fit function of TNI by year (R^2=0.89, p<0.001), indicating a continuously increasing trend of TNI. N fertilizer application was the major contributor to TNI, which increased significantly with an annual growth rate of 4.2% during 1978-2015 (Table 2). N fixation within the basin was the second largest source of TNI, increasing slightly from 41 to 52 kg N ha^{-1} yr^{-1} during the study period, attributed to the increase in bean and peanut cultivation for farm profit. These two largest sources comprised ~80% of TNI throughout the study period. N input in imported feed climbed to a peak of 29 Gg N in 2003 and experienced a dramatic decrease in the mid-2000s due to animal diseases during 2007-2008. Although N deposition was of minor importance, it increased by 2-fold over the study period due to increasing NO_x emission (Liu et al., 2013).

Total N loss increased by 3.1-fold, from 92 to 280 kg ha^{-1} during the study period (Figure 3c). As total N loss increased at a lower rate than that of TNI, the N loss-to-input ratio showed a decreasing trend, from 69% to 56%. N loss via NH_3, N_2O and N_2 to the atmosphere were greatest, accounting for 76% (70-77%) of total during 1978-2015, followed by N losses via runoff and erosion to waters, accounting for 19% (18-20%) of total. The N discharge to surface water increased from 9 to 16 kg N ha^{-1} between 1978 and 2003, and then decreased to 12 kg N ha^{-1} in 2015. The decrease in N discharge to waters is attributed to the construction of wastewater treatment plants in the urban areas.

3.1.2 Pattern of P input and loss

Overall, total P input (TPI) increased by 4.2-fold from 11.8 Gg (25.3 kg ha^{-1}) in 1978 to 49.2 Gg (115.2 kg ha^{-1}) in 2015 (Figure 3b), and also had a strong linear fit function by year (R^2=0.97, p<0.001). Fertilizer was the single dominant source of TPI, accounting for 74-87% of TPI during 1978-2015 (Table 2). P input through the imported feed increased significantly by 3-fold throughout the study period. The atmospheric P deposition was very small, comprising less than 4% of TPI.

Total P loss increased by 2.3-fold from 5.2 to 12.1 kg ha^{-1} during 1978-2015 (Figure 3d). The increase is much lower than that of TPI, leading to a significant decrease of loss-to-input ratio, from 21% to 11%. P loss via erosion and runoff was the dominant P fate since the early 1980s and contributed to over 60% of total loss in 2015. Change in P discharge to surface water was similar to N, but the contribution to total P loss decreased from 49% to 35% during 1978-2015.

3.1.3 Spatial pattern of nutrient input and loss

At the administrative region scale, total N and P input increased by quite different degrees across different counties, ranging from 127 to 1373 kg ha^{-1} for N and 15 to 341 kg ha^{-1} for P (Figure S2a). The hotspots were two counties located in the northwestern basin (Hefei and Feidong). Hefei, in which the capital city of Anhui Province is located, has the most pronounced increase in TNI and TPI by 6-fold and 9-fold, respectively. It is not accident because Hefei has undergone a rapid urbanization in the past three decades (Figure S1), leading to a large increase in food imported to support the population and a transfer of cropland into urban areas. As for Feidong, the major cause of the striking increase in TPI and TNI was the intensification of crop faming and animal breeding, which increased the nutrient input via fertilizer and imported animal feed.

Spatially, the changes in nutrient loss ranged from 62 to 373 kg ha^{-1} for N and from 7.9 to 18.0 kg-P ha^{-1} for P. The most significant increase areas were also in Hefei and Feidong (Figure S2b). For Hefei, the increase in N loss was driven by discharge of sewage and emission of NO_2, NH_3 and N_2O from anthropogenic bio-solids. As for Feidong, the major reason for the significant increase in loss was agricultural runoff, and emission of NO_2, NH_3 and N_2O from fertilizer application and animal manure.

3.2 Change in N:P stoichiometry

The N:P molar ratio in total nutrient input increased slightly from 11.6 in 1978 to 13.6 in 1993 (Figure 4a), due to rapid increase in atmospheric N input via N fixation and atmospheric N deposition (Table 2). However, the trend had reversed since 1994, when increase in N input was overtaken by a more rapid increase in P input from chemical fertilizer application. Across the study period, the N:P in total nutrient input was bounded between that of cropland input and that of livestock requirement, indicating that N and P input were well matched with demand.

Besides, the N:P in total nutrient input converged toward that of crop uptake over time, i.e., nutrient input to cropland became more closely match to crop needs.

In contrast to input, the N:P molar ratio in total nutrient loss from the CLB increased significantly over the study period, from 39.0 to 51.3 (Figure 4b). Before the mid-1990s, the increase was attributed to the combined effects of the increase in N:P in loss from cropland and human waste. The continuously increasing N:P of loss from human waste reflects the increasing consumption of animal protein, which contains more N than P. Since the mid-1990s, however, there was a period with little net change in the N:P in total nutrient loss from the CLB. This is the combined effects of the continued increase in N:P in human food consumption and the decrease in N:P in nutrient loss from crop farming and livestock breeding, with the latter reflecting the enrichment of N relative to P in livestock diet and feed.

3.3 Sources and drivers of the changes

The variables' VIFs employed by OLS were far larger than 10 (Table S18), indicating the obvious multicollinearity among the variables. We thus applied ridge regression for Eq. (3) to deal with the multicollinearity. Through the four-fold cross validation using "glmnet" package in R language, we found minimum mean square error estimations existed when λ were 0.039 and 0.047 for N and P respectively (Figure S4). The regression coefficients of all explanatory variables under such conditions were significant at the level of 0.0001 and the R square was 0.97, indicating a good reliability of goodness of fit (Table 3, Figure 3a and 3b).

The results show that the key factors increasing the nutrient input were NUE_c, urban rate, diet choice, population and NUE_a, which together contributed to ~90% of the changes in N and P input in the CLB (Table 3). Among the factors, decreasing NUE_c accounted for the largest contributions to the changes in N input (30%) and P input (25%). This is reasonable as the nutrient input to crop farming was even larger than the total nutrient input to the whole basin, when the internal nutrient recycling from wastes was included (Table 1). The increase in nutrient input was sourced from the large application of chemical fertilizer, accounting for 61% and 75% of N and P input to crop farming in 2015 (Figure S3a, S3b), as the over-fertilization is a common phenomenon arising from the availability of cheaper fertilizer and a lack of guiding appropriate fertilization to the farmers in the CLB.

The urbanization and diet choice contributed 32% and 37% to the changes in N and P input (Table 3). In the CLB, the diet had changed toward more animal protein, while the N intake from animal food increased from 7% in 1978 to 47% in 2015; the corresponding increase for P was 1% to 16% (Figure S3e, S3f). This change resulted in greater nutrient input in the food production process, as the NUE_a was lower than NUE_c. The rapid urbanization, increasing from 13% in 1978 to 33% in 2015 (Table S17), also accelerated the change toward high animal protein diet, as urban residents consumed more animal food than rural residents (Table S7). The population was also an important deriving factor, with contribution of 13% and 17% to N and P inputs, respectively. Although the daily nutrient intake from food decreased from 10.0 to 8.2 g capita^{-1} for N and from 2.8 to 1.5 g capita^{-1} for P, the increasing population made the total N intake stable and only a slight decrease for P (Table 2).

The dramatic expansion of animal breeding, with a 7.2-fold increase in feed input (Figure S3b, S3c), caused NUE_a to have a contribution of 10% and 11% for the changes in N and P input, respectively. As intensive feedlots became more common, the nutrient converted to manure became more spatially concentrated and led to an excess of nutrients, relative to the capacity of cropland around the feedlots. Between 1978 and 2015, an increasing proportion of N and P in manure either accumulated within the soil or lost to surface water, making animal breeding an important source of pollution.

Overall, NUE_c, urban rate, diet choice, population and NUE_a were key factors driving the changes in nutrient input to the CLB. However, while the urbanization trend is the inevitable result of economic development, the practical strategies to regulate nutrient flows are focusing on increasing NUE_c and NUE_a, guiding a reasonable diet choice and adjusting industrial structure by reducing reliance on the agricultural economy.

3.4 Consequences of the changes

The N:P stoichiometry of loss was, on average, 4.2 times higher than that of input (Figure 4a, 4b). This indicates that in the CLB, N is more likely to be lost, whereas P is more likely to accumulate. When nutrient accumulation is considered as an indicator of regional N:P stoichiometry, a decreasing trend of regional N:P was observed since the middle 1990s (Figure 5a), reflecting greater rate of increase in P input relative to N input to the CLB. This finding is the opposite of the global trend (Peñuelas et al., 2013), but is consistent with the finding for three subtropical catchments in China where decreasing N:P molar ratio was observed in agricultural soils (Liu et al., 2016a). The biogeochemical cycles of C are also highly impacted by changes in regional N:P inputs; for example, it has been widely observed that P additions alone can increase terrestrial C pools and fluxes by 10-23%, aboveground production by 34% and belowground biomass production by 13% (Li et al., 2016; Peng et al., 2017)). In this case, the greater enrichment of P could have further effects on bio-productivity by interactions with the C cycle.

We examined the change in N:P stoichiometry of nutrient loss to surface waters and the relationship with TN:TP concentration in the Chaohu Lake, the receiver of all waters from the drainage areas of the CLB. We found that N:P stoichiometry in nutrient loss to waters increased to 14 in the mid-1990s, and then decreased steadily to 12 at the end of study period (Figure 4c). There was significant correlation between N:P of in nutrient loss to water and lake-water TN:TP concentration ($p<0.05$), indicating the obvious impact of human activities on decreasing TN:TP concentration in Chaohu Lake. Although an insight assessment of environmental consequences from changing nutrient stoichiometry is beyond the scope of this work, the decrease in N:P ratios could be a potential cause of the frequent algae blooms in

Chaohu Lake since 1980s (Zhang et al., 2014), as Chaohu Lake is P-limited, with typically much higher TN:TP ratio in the lake wasters (16-45, Figure 4d) than the Redfield ratio of 16 (Huang et al., 2015; Redfield, 1958). Our results indicate that, in addition to controlling the amount of nutrient losses, balancing N:P stoichiometry may also important to achieve improvements in water quality and to reduce the incidence of nuisance and harmful algal blooms.

3.5 Uncertainties

There are two main sources of uncertainties in this study: structural uncertainties associated with model construction, and uncertainties from basis dataset. Structural uncertainties refer to assumptions for calculating nutrient flows. For example, the assumptions used to allocate crop products to meet local demand is a source of uncertainty in assessing exchange between the basin and outside, as a larger proportion of crops produced in the CLB could be exported and crops consumed locally could be imported. This may lead to over- or under-estimations of the total N and P input. We treated nutrient in solid wastes and the underused manure as water accumulation, which could be washed out into waters. The simplified assumption may cause an evident impact on the estimation of nutrient losses in northwestern CLB, where population and animal industries are concentrated. Nevertheless, this kind of structure uncertainty is difficult to be assessed quantitatively, which is also beyond the scope of this study.

Uncertainties also come from basis dataset. The activity dataset was self-reported from Hefei, Lu’an, Ma’an’shan and Wuhu Statistics Office, including a certain level of uncertainty. We considered these basic data are with a confidence interval of 5%. The parameters were obtained from empirical investigation and literature. The confidence intervals of parameters from empirical investigation, such as treatment ratio of waste water and recycling ratio of straw and manure, were estimated at 10%. To minimize the uncertainty of parameters from literature, those from studies conducted in neighboring regions were the primary choice. We also collected as much literature as possible to derive confidence intervals for each parameters. A Monte Carlo simulation was then performed with 10 000 iterations to quantitatively assess the uncertainties. The 95% of confidence intervals of the distribution of outputs are shown in Table 2. In general, the aggregated uncertainties are within a relative narrow range of 11-16% for TNI and 16-23% for TPI during the study period, indicating that our model is robust relative to uncertainties from data and parameters. The sensitivity analysis shows that the variance of total nutrient inputs and losses are sourced from several parameters (Figure 5). For example, the nutrient content in fertilizer has the greatest role in the uncertainties, contributing to 61-84% of variance of total N and P inputs in 2015. Thus, special attention should be paid for these parameters to reduce uncertainties in assessing nutrient flows.

4 Conclusion

This study developed an Integrated Nutrient Flow Analysis (INFA) model to assess historical N and P flows in a mixed land use lake basin. This allowed a comprehensive evaluation of the changes in N and P flows, N:P stoichiometry and a thorough understanding of their links with human activities over a 38-year period in the CLB.

Our results indicate that the increase of total N input was slower than that of P (3.5- fold versus 4.2-fold) during 1978-2015, while total N losses increased much faster than that of P (3.1-fold versus 2.3 fold), with a decline in N:P in nutrient inputs and accumulation within the basin since the mid-1990s. We also found that a decline in N:P loss to waterbodies in the basin was correlated ($p<0.05$) with TN:TP concentration in Chaohu Lake. The results confirm a fact of human-induced decrease in N:P in regional environment, which may be related to the frequent algal blooms in the P-limited lake by supplying more P than N. The changes in N and P inputs were driven by nutrient use efficiency, urban rate, diet choice and population, which had a combined contribution over 90%. Mitigating the intensification and imbalance in N and P flows requires a focus on improving the efficiency of nutrient use in agriculture, as well as exploring the potential of wider interventions such as lowering animal protein in diets and changing regional industry structure.

The method applied in this study provides a holistic picture of N and P flows and stoichiometry in human-impacted areas, and a basis for managing nutrients balance. Moreover, the method has a wide application for many mixed land use regions to explore nutrient management options.

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Appendix A. Supplementary data

Appendix B. Notation list

1. P = Phosphorus
2. N = Nitrogen
3. kg = 10^3 grams
4. Gg = 10^9 grams
5. ha = hectare
6. INFA = Integrated Nutrient Flow Analysis Approach
7. CLB = Chaohu Lake basin
8. $NUE_{c/a}$ = Nutrient use efficiency of crop farming / animal breeding
9. STIRPAT = The Stochastic Impacts by Regression on Population, Affluence, and Technology
10. TNI = Total nitrogen input
11. TPI = Total phosphorus input

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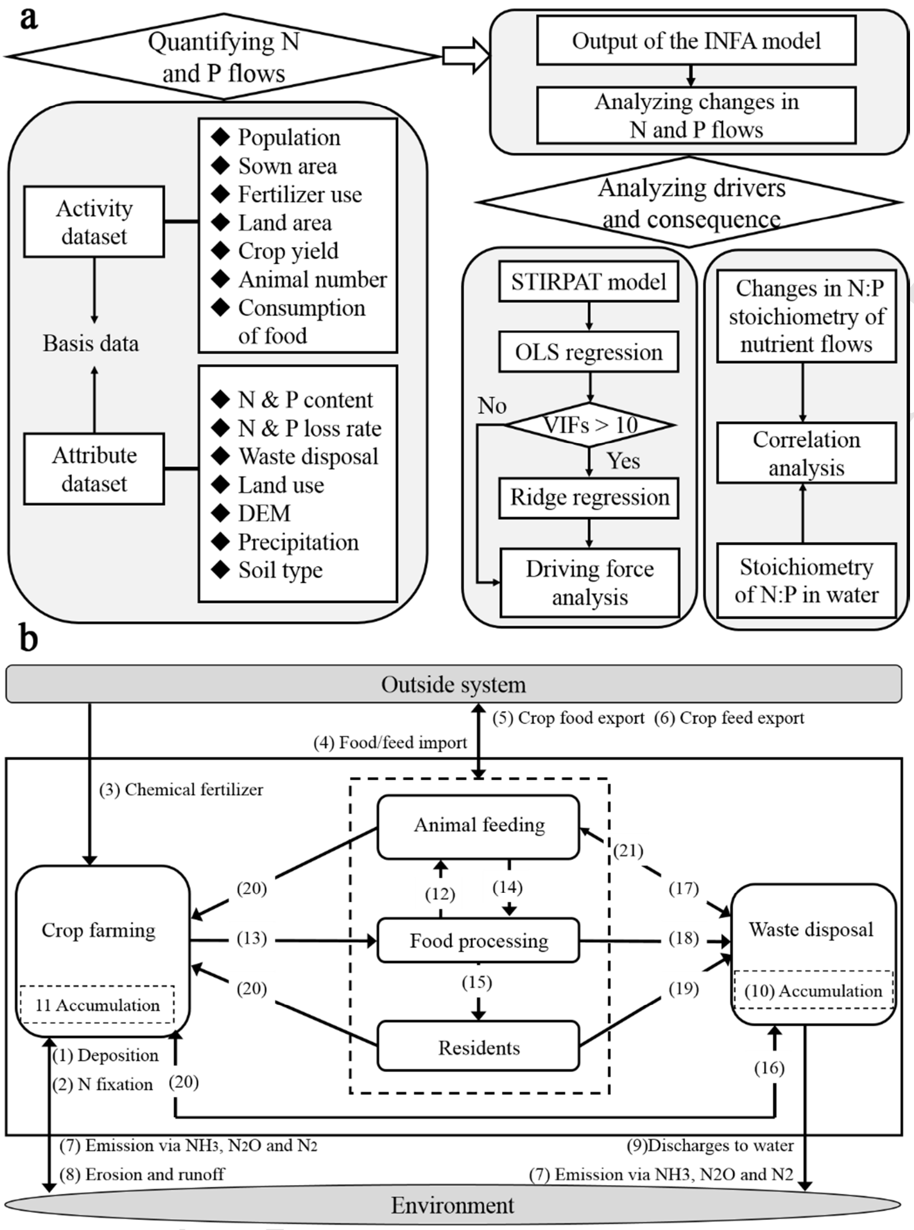

Figure 1 (a) The flow chart of this study. (b) Schematic diagram of the INFA.

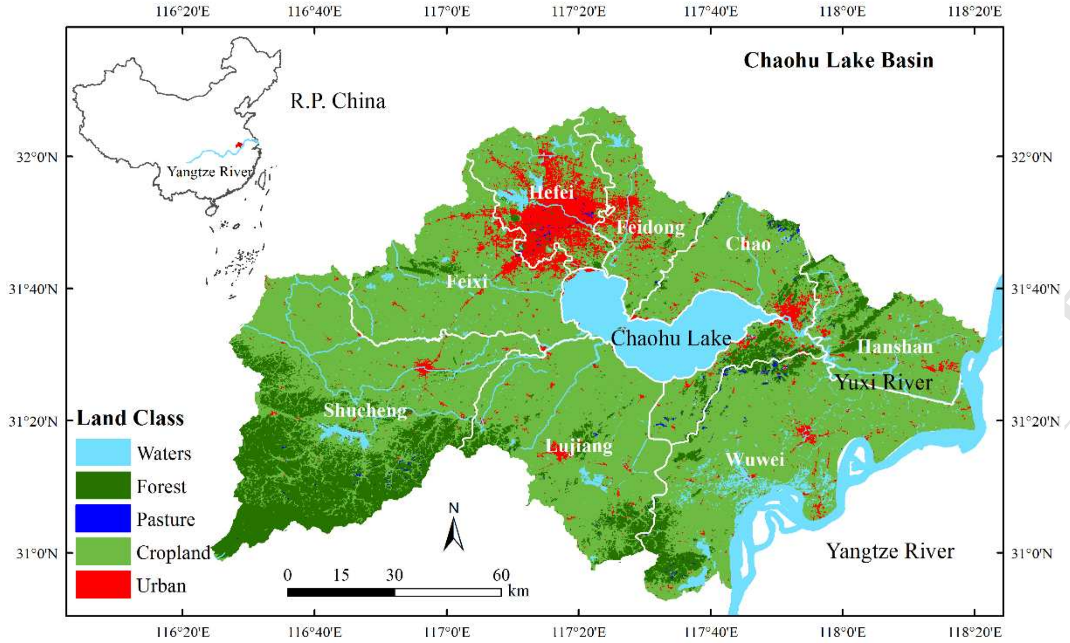

Figure 2 Location and land use of the Chaohu Lake Basin.

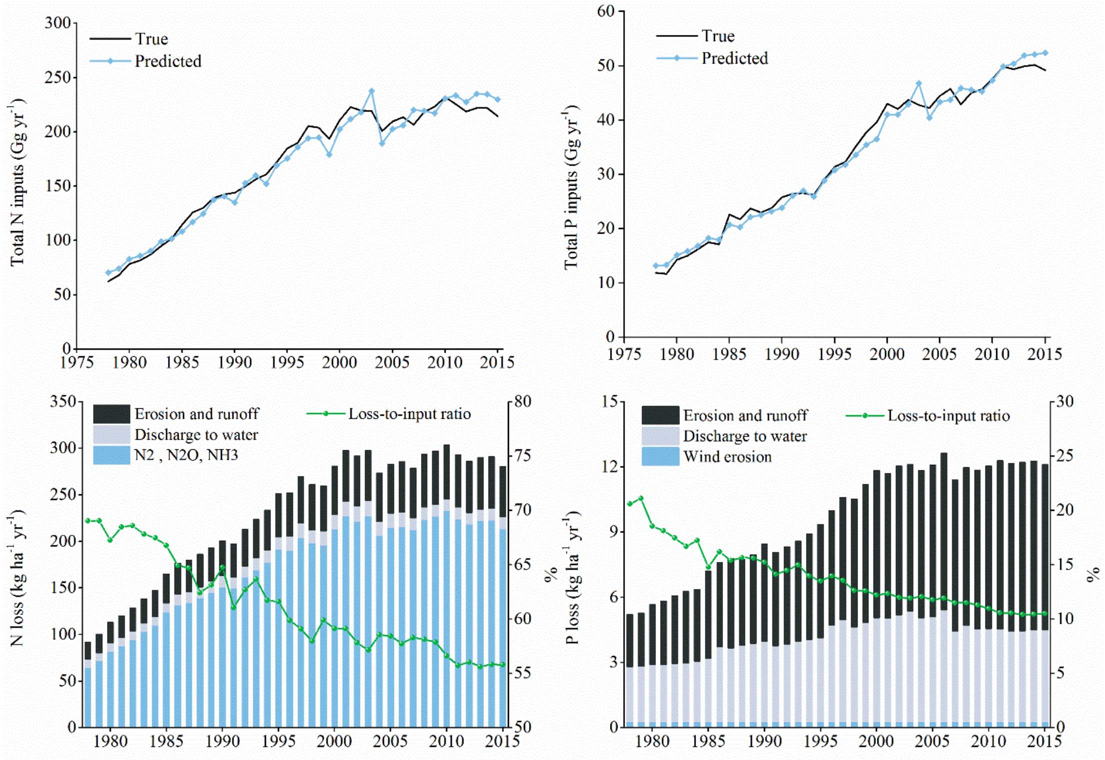

Figure 3 Temporal trends in the Chaohu Lake Basin for (a) total nitrogen inputs (b) total phosphorus inputs, (c) nitrogen losses per cropland area (ha^{-1}) and (d) phosphorus losses per cropland area (ha^{-1}).

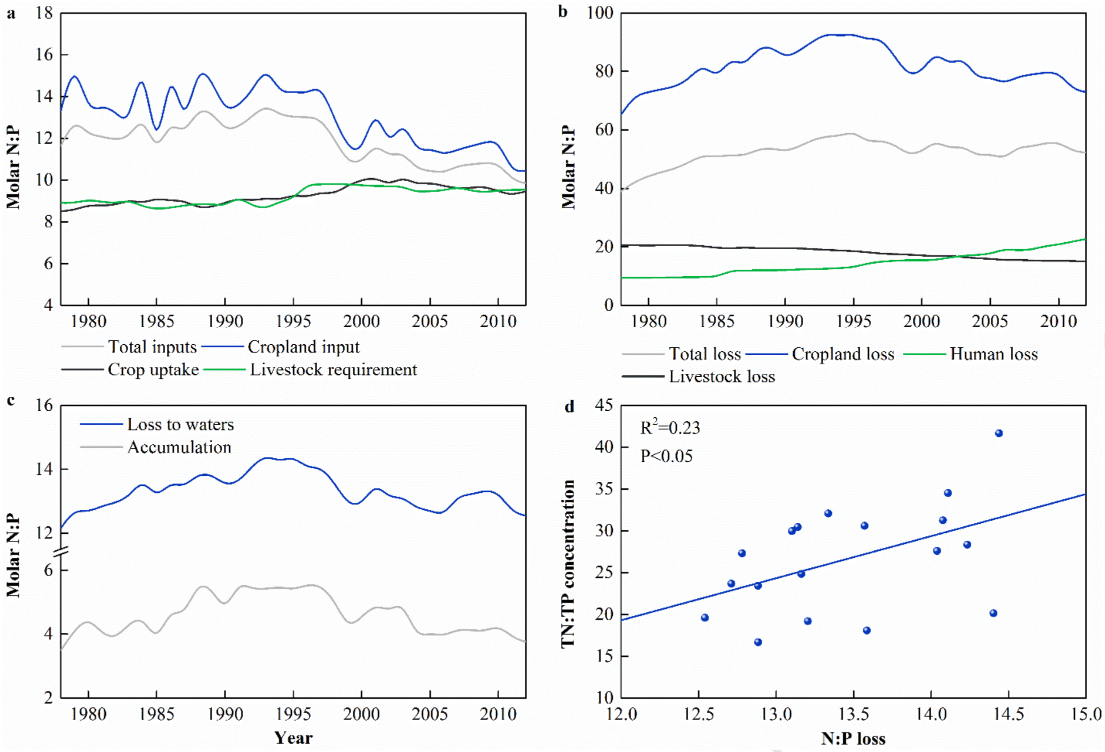

Figure 4 Changes of molar stoichiometry. (a) Stoichiometry in nutrient input. (b) Stoichiometry in nutrient loss. (c) Stoichiometry of N:P in emission to waters and accumulation. (d) Relationship between N:P loss to surface waters in the Chaohu Lake Basin and TN:TP concentration ratios in Chaohu Lake. TN and TP concentration data were collected from the study by Huang et al. (2015) from 1992 to 2009.[39]

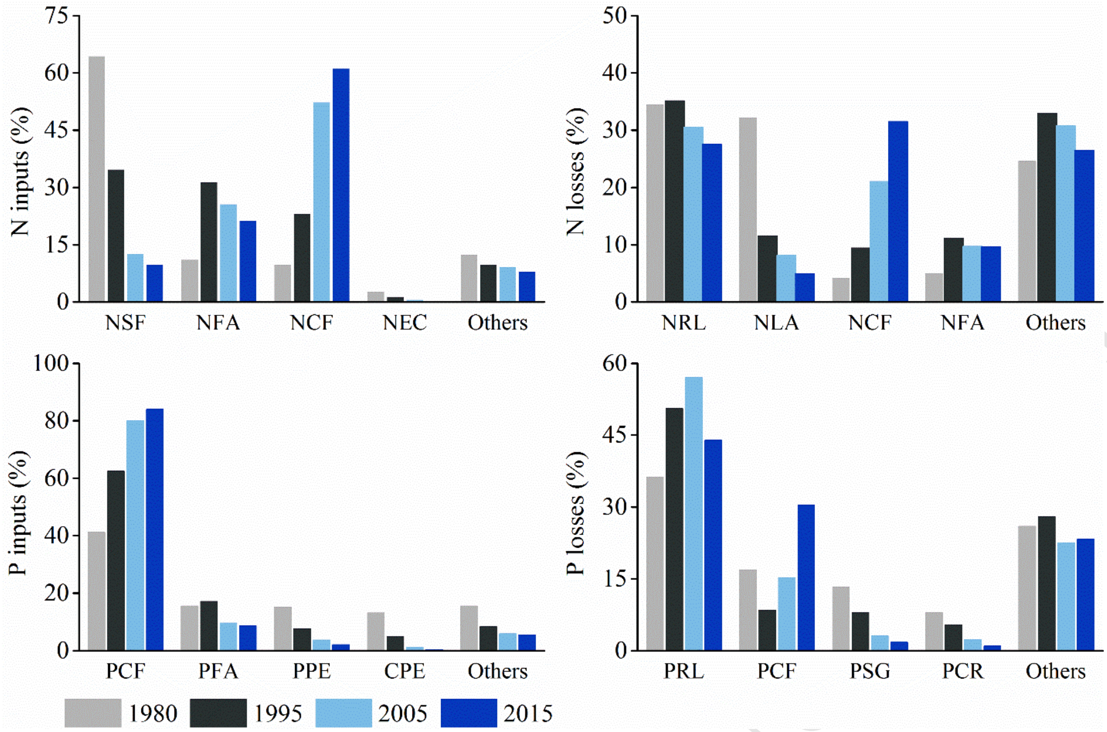

Figure 5 Contribution to variance of (a) total N inputs and (b) total N losses (c) total P inputs and (d) total P losses. NSF, NFA, NCF, NEC, NRL and NLA represent non-symbiotic fixation rate, amout of N fertilizer application, N content in fertilizer, N excretion rate of cattle, runoff and leaching rate of N, manure loss via N_2. PCF, PFA, PPE, CPF, PRL, PSG and PCR are P content in fertilizer, amout of P fertilizer application, P excretion rate of pig, P excretion rate of cattle, runoff and leaching rate of P, sewage generation rate and P content in rice.

Table 1 Data list of basis dataset used in this study

Category	Sub-category	Unit	Note	Source
Activity dataset				
Cropland areas	Paddy land and upland	1000 ha	1978-2015 (Table S1)	APBS, 1989-2016 CMBS, 1979-2016 HMBS, 1979-2016 LMBS, 1979-2016 MMBS, 2012-2016 WMBS, 2012-2016
Snow areas	Rice, wheat, maize, bean, tuber, peanut, rape, sesame and cotton	ha	1978-2015 (Table S2)	
Fertilizer use	Compound fertilizer, N fertilizer and P fertilizer	1000 t	1978-2015 (Table S3)	
Crop yield	Rice, wheat, maize, other cereals, bean, tuber, peanut, rape, sesame, cotton, sugarcane and vegetable	1000 t	1978-2015 (Table S4)	
Animal number	Pig, cattle, goat, poultry, fish, shrimp, shell and other aquatic products	1000 unit livestock; 1000 t for aquatic product	1978-2015 (Table S5)	
Population	Urban and rural	1000 unit	1978-2015 (Table S6)	
Food consumption	Cereal, vegetable, oil, meat, chicken, fish, egg, milk	kg $capita^{-1}$ yr^{-1}	1978-2015 (Table S7)	
Attribute dataset				
Nutrient content in	Rice, wheat, maize, other cereals, bean, tuber, peanut, rape, sesame,	%	Fixed value	Literature data

crops	cotton, sugarcane and vegetable		(Table S9)	
Loss rate of nutrient	Chemical fertilizer for upland and paddy land; Manure for application and storage.	%	Fixed value (Table S10)	Literature data
Nutrient content in animal	Pig, cattle, goat, poultry, fish, shrimp, shell and other aquatic products	%	Fixed value (Table S11)	Literature data
Nutrient content in food	Cereal, vegetable, oil, meat, chicken, fish, egg, milk	%	Fixed value (Table S12)	Literature data
Straw utilization	Recycling, livestock feed and biofuel	%	1978-2015 (Table S13)	Literature data (1978-2005); Field investigation (2006-2015).
Manure utilization	Recycling	%	1978-2015 (Table S14)	Literature data (1978-2005); Field investigation (2006-2015).
Treatment rate of sewage	Urban	%	1978-2015 (Table S15)	Field investigation
Animal extraction rate	Pig, cattle, goat and poultry	g capita^{-1} day^{-1}	Fixed value Table S16	Literature data
Precipitation	Daily precipitation	mm	1978-2015	http://data.cma.cn/
Land use	Urban, cropland, forest,	-	1988, 1998,	Interpreted from SPOT data

	pasture and water		2008, 2012	with pixels of 10m
DEM	Slope	m	2010	https://search.earthdata.nasa.gov/
Soil proprieties	Depth	-	2010	China Soil Map (v1.1) http://westdc.westgis.ac.cn.
	Type	-		

Table 2 Changes in N and P flows in the Chaohu Lake Basin in 1980, 1995, 2005 and 2015. The values presented are five-year annual averages.

	N								P							
	1980		1995		2005		2015		1980		1995		2005		2015	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Total input	75	64-86	165	146-183	214	188-241	220	190-251	14.0	11.7-16.4	28.1	23.0-33.2	43.0	33.8-52.3	49.8	38.1-61.6
Deposition	4	4-5	7	6-8	8	7-9	10	9-11	0.8	0.3-1.5	0.8	0.3-1.5	0.7	0.3-1.4	0.7	0.3-1.4
Fertilizer	47	42-52	130	116-145	160	138-184	163	136-192	11.0	9.3-12.8	24.0	19.4-28.8	34.6	26.0-43.5	41.3	30.1-52.8
N fixation	19	10-28	20	11-30	22	13-32	22	13-32	0.0	0-0	0.0	0-0	0.0	0-0	0.0	0-0
Net feed import	5	1-9	7	2-12	23	16-31	25	18-32	2.2	0.8-3.6	3.3	1.5-5.1	7.6	4.9-10.4	7.7	5.1-10.4
Total output	75	64-87	165	146-183	214	188-241	220	190-251	14.0	11.7-16.4	28.1	23.0-33.3	43.0	33.8-52.3	49.8	38.1-61.6
Crop food export	6	3-9	9	6-12	13	10-17	30	26-34	1.9	0.9-2.9	2.6	1.6-3.7	3.9	2.8-5.0	8.7	7.0-10.5
Animal food export	1	0-1	1	0-1	8	6-9	7	5-9	0.1	0-0.1	0.1	0-0.1	0.5	0.4-0.7	0.5	0.3-0.7
Total loss	37	30-45	78	65-91	95	78-112	94	76-114	0.1	0.1-0.1	0.1	0.1-0.1	0.1	0.1-0.1	0.1	0.1-0.1
Atmospheric loss	51	42-61	102	85-120	125	103-148	123	99-148	2.6	2.1-3.3	4.0	3.2-4.9	5.3	4.1-6.6	5.2	4.1-6.4
Erosion and runoff	4	2-6	6	3-9	7	4-11	5	3-8	1.2	0.7-1.9	1.7	1.0-2.6	2.2	1.2-3.4	1.7	0.9-2.7
Discharge to water	10	4-16	19	8-29	23	10-36	24	10-37	1.3	1.1-1.5	2.2	1.9-2.5	3.0	2.4-3.5	3.3	2.6-4.0
Waste accumulation	17	14-20	20	15-24	30	24-37	31	24-39	3.4	2.6-4.3	4.6	3.5-5.8	7.7	6.2-9.3	7.7	6.2-9.3
Soil accumulation	0	-13-14	33	14-53	38	15-63	29	4-55	6.1	3.5-8.6	16.8	11.9-21.9	25.7	17-34.4	27.7	16.6-38.9
Animal feed	12	10-13	18	15-20	30	25-34	29	26-33	1.9	1.6-2.3	2.9	2.4-3.6	4.6	3.7-5.9	4.8	3.9-5.8
Crop products	29	26-31	32	29-36	34	31-38	45	41-50	8.5	6.9-10.2	9.6	7.8-11.4	10.1	8.3-11.9	13.2	11.0-15.6
Animal products	4	3-4	8	7-9	24	21-26	27	24-31	0.6	0.5-0.7	1.3	1.1-1.5	3.5	3.0-4.0	4.0	3.4-4.5
Food consumption	24	22-26	28	25-31	29	26-32	27	24-29	6.7	5.5-8.1	7.3	5.9-8.6	6.7	5.6-8.0	5.2	4.4-6.1
Crop straw	27	23-30	32	28-37	41	34-48	47	40-54	3.7	3.2-4.2	4.5	3.9-5.3	5.8	4.8-6.8	6.5	5.5-7.5
Animal manure	20	17-23	25	21-29	35	29-40	30	25-35	4.5	3.2-5.8	6.1	4.4-7.7	9.4	7.0-11.9	9.0	6.7-11.4
Processing waste	1	1-2	3	2-4	8	6-10	9	7-11	0.4	0.4-0.5	0.9	0.8-1.1	2.5	2.1-2.9	2.8	2.3-3.2
Human wastes	24	21-27	28	25-31	29	26-33	27	24-30	6.7	5.4-8.1	7.3	5.9-8.7	6.7	5.5-8.0	5.2	4.3-6.2
Straw/manure recycled	42	37-47	51	45-57	64	56-73	67	58-77	9.8	8.1-11.6	11.4	9.6-13.4	13.8	11.6-16.2	13.6	11.3-15.9
Straw feed	7	6-9	8	6-10	5	4-7	4	3-5	1.0	0.8-1.2	1.1	0.9-1.4	0.8	0.6-0.9	0.5	0.4-0.6

494 Table 3 Contributions of factors to the changes of total nutrient input.

Factors	Annual grow rate (%)	Regression coefficient	Effect on change of inputs [a]	Contribution to changes [b]	*t*-Statistic
N inputs	3.313				
Urban rate	2.609	0.210***	0.548	17	5.799
Population	0.915	0.475***	0.435	13	2.897
Dietary	5.286	0.093***	0.492	15	3.347
Crop export	2.837	0.101***	0.287	9	6.471
Animal export	-0.619	-0.041***	0.025	1	2.916
NUEc	-1.064	-0.952***	1.013	30	15.14
NUEa	2.901	0.118***	0.342	10	4.558
Others		5.869***	0.172	5	
λ	0.039				
R-square	0.97				
P inputs	3.820				
Urban rate	2.609	0.304***	0.793	21	9.116
Population	0.915	0.713***	0.652	17	5.447
Dietary	6.530	0.091***	0.594	16	3.467
Crop export	2.787	0.074***	0.206	5	4.924
Animal export	-0.668	-0.068***	0.045	1	3.417
NUEc	-1.375	-0.687***	0.945	25	10.951
NUEa	2.737	0.159***	0.435	11	5.361
Others		3.056***	0.149	4	
λ	0.047				
R-square	0.97				

495 [a] Effect on changes of input=average annual growth rate × regression coefficient

496 [b] Contribution to changes =effect on changes of input × average annual growth rate

497 *** Significance is at the 0.001 level

Supplementary Material (SM) to the full paper:

Enhanced nitrogen and phosphorus flows in a mixed land use basin:

Drivers and consequences

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Contents of this file: 29 pages, 16 SI Tables and 3 SI Figures.

Further information is provided on description of method (Text S1, Pages S2-S5), basic data (Table S1-S7, Pages S6-S17), parameters (Table S8-S15, Pages S18-S21), data used in assessing driving factors (Table S16, Pages S22-S23), land use change in the CLB (Figure S1, Pages S27), spatial patterns in nutrient inputs and losses (Figure S2, Pages S28), nutrient inputs and outputs for crop farming, animal breeding and human consumption (Figure S3, Pages S29).

Text S1 details of the calculation of N and P flows

1 Atmospheric inputs

For N, atmospheric inputs involves atmospheric deposition and biological N fixation. Atmospheric N deposition was estimated from cropland area (Table S1) and annual N deposition rate, which was based on a linear regression function of deposition rate with year by Liu (2013):[1]

$$f(t) = 0.411t - 804.353$$

Here, $f(t)$ is annual N deposition rate (kg N ha^{-1}) in the year t. This equation was fitted by 671 observed data in China from 1980 to 2010, which just covers our study period. Biological N fixation can be estimated by area-based approach and yield-based approach.[2] Goyette et al. (2016) compared the two approach and found that the latter was more relevant in longer-term assessments of leguminous crops.[3] Thus, we followed Le Noë et al. (2017) using a simplified yield-based equation to estimate N fixation caused by leguminous crops:[4]

$$f(\eta) = 1.23\eta$$

Here, $f(\eta)$ is N fixation and η is yield of leguminous crops. In the Chaohu Lake Basin (CLB), leguminous crops includes beans and peanut. The leguminous crops were grouped as beans and peanuts. As for non-leguminous crops, we used area-based approach followed by a parameter of 15-30 kg N ha^{-1} with a mean value of 22.5 kg N ha^{-1} and the sown area of non-leguminous crops (Table S2).[5]

For P, the atmospheric inputs involves atmospheric deposition, which is mainly particulate dust driven by natural force.[6] The temporal change was assumed to be negligible, thus we estimated atmospheric P inputs using cropland area (Table S2) and fixed observation date of atmospheric deposition rate in this area with a range of 0.37 and 3.95 kg N ha^{-1} and mean value of 0.81 kg N ha^{-1}.[7]

2 Crop farming

N and P inputs to crop farming refer here to atmospheric deposition, N fixation, chemical fertilizer, and recycled organic fertilizer (crop straw, animal manure and human manure). Estimates of N and P inputs in chemical fertilizer were based on county-level fertilizer use data (Table S3) and nutrient content in fertilizer. In this study, chemical fertilizers were aggregated into 3 main type, including N fertilizer, P fertilizer and compound fertilizer. N and P content in compound fertilizer is assumed at 33% and 33% based on local investigation with a confidence interval of ±12%.[8] The estimates of N and P inputs in recycled organic wastes are shown in following sections.

N and P outputs from crop farming include harvested crops, crop straws, nutrient loss to atmosphere and surface water. N and P in harvested crops were taken as dry weight (Table S4) and were converted from dry weight to kg N yr^{-1} based on the N and P contents of each crop types (Table S9). N and P in crops straws were estimated from weight of harvested crops, harvest index and the N and P contents of each crop types (Table S9). N losses to atmosphere via N_2, N_2O and NH_3 were estimated based on total N inputs in chemical and organic fertilizer and N emission rate of N_2, N_2O and NH_3 for each fertilizer (Table S10). N and P losses to surface waters through surface erosion and runoff were calculated by an adopted method developed by Velthof et al. (2009):[9]

$$L = A * Sur * f_{lu} * \mathrm{minimum}(f_p, f_{rc}, f_s)$$

where, L is N or P losses through surface erosion and runoff; A is N or P applied to croplands via inorganic and organic fertilizer; Sur is a maximal surface runoff fraction for applied N and P (Table S10); f_{lu} is a reduction factor related to slope classes; f_p, f_{rc} and f_s are set of reduction factors for precipitation, soil depth and soil type. N and P accumulation in crop soil could be established as the difference between N and P inputs and their losses.

3 Animal breeding

N or P in animal products were calculated based on county-level sales number of animals (Table S5) and N and P content of each animal types (Table S11). N and P in live animals were broken down into edible and inedible production. The former represents meat products that is prepared for human consumption, and the latter includes bone, blood and other unavoidable losses in food processing stage.

Animal excretion has often been estimated by annual sale number and average N and P excretion amount of each animal types.[5, 10] For cattle, which reside on a farm more than one year, the replacement to offset sale number is continuous, thus it is reasonable to assume annual sale number as the population throughout the year. However, for pig, goat and poultry which reside on a farm less than whole year, annual sale number may over-estimate the N and P excretion.[2] We thus adjusted animal population by considering life cycle of pig, goat and poultry followed.[11]

$$P = \frac{(I_{sales} + I_{end})}{1 + cycles}$$

Here, *P* is defined as a daily average breeding amount. I_{sales} is the sale number of animal (Table S5). I_{end} is the year-end number of animal (Table S5). Cycles is related to life cycle of animal, which is defined as 365 days divided by the days from birth to sales.

As it is difficult to get values of per capita feed requirement of each year during study period, animal feed requirements were calculated by N and P in animal products, added by and N and P in excretion based on mass balance principle.

4 Human consumption

N and P consumptions were estimated by annual county-level population (Table S6), annual food consumption per capita (Table S7), and N and P content in food (Table S12). Annual food consumption per capita were obtained from censuses for the whole basin. N and P in human excretion, kitchen residues and sewage were calculated by 88%, 4% and 8% of food intake based on empirical investigation and previous study. [8, 12]

5 Waste disposal

N and P in wastes include animal and human excretion, inedible part of animal products, crop residues, kitchen residues and sewage. N and P in straw and excreta recycled to crop farming were calculated by multiplying the total N or P in them with recycling ratios of straw (Table S13), human excreta (Table S14) and animal manure (set as 70%, 70%, 40% and 55% for pig, cattle, goat and poultry based on our empirical investigation). N losses from excretion to atmosphere via NH_3 were 15-35% of N in total excretion corrected for recycled proportion (Table S10).[4, 13] N and P in sewage discharged to surface water was corrected for N and P in sewage and the proportion disposed by sewage treatment plants (Table S15). Thus, N and P accumulation in crop soil could be established as the difference between N and P inputs and their losses.

6 Food and feed imports and exports

As the essential function of local agricultural production, we assumed that crop and animal production meets local human demand, following Yuan et al. (2014).[8] Thus, the N and P in exchanged food could be obtained as the difference between local food production and human consumption. A positive and negative value would indicate net export and import, respective. By analogy, the annual N and P imports and exports in feed could obtained as the difference between local feed production and animal feed requirements.

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Table S1 Cropland areas in the CLB from 1978 to 2015 (unit: thousand ha). PL and UL represent paddy land and upland, respectively.[1-9]

Year	Hefei		Feidong		Feixi		Chaohu		Wuwei		Lujiang		Hanshan		Shucheng	
	PL	UL	PL	UL	PL	UL	PL	UL	PL	UL	PL	UL	PL	UL	PL	UL
1978	12	3	61	35	58	16	39	14	77	15	64	6	20	3	38	7
1979	12	2	62	33	58	16	39	14	78	15	64	6	20	3	38	7
1980	12	2	61	33	58	16	38	15	78	15	64	6	20	3	38	7
1981	11	3	62	32	58	16	38	15	77	15	64	6	20	3	38	7
1982	12	2	64	31	58	16	38	14	77	15	64	6	20	3	38	7
1983	11	2	65	30	58	16	38	14	77	15	64	5	20	3	38	7
1984	11	3	67	28	58	16	38	14	78	14	64	5	20	3	38	7
1985	11	3	66	28	57	16	38	14	77	14	64	6	20	3	38	7
1986	11	2	57	37	57	16	38	15	77	14	64	6	20	3	38	7
1987	11	3	56	37	55	18	37	15	76	15	72	6	20	3	38	7
1988	10	3	56	37	57	15	37	15	75	15	73	6	20	3	38	7
1989	10	2	56	37	57	15	37	15	75	15	73	6	20	3	38	7
1990	10	3	56	37	56	16	37	15	75	15	72	6	20	3	38	7
1991	10	3	56	37	56	16	37	15	73	17	72	6	19	3	39	6
1992	10	2	55	37	56	16	37	15	72	17	71	6	19	3	38	7
1993	8	2	55	37	55	15	36	15	69	21	71	6	19	3	38	6
1994	8	2	55	37	55	15	36	15	68	21	71	6	19	4	38	7
1995	8	2	55	37	55	16	36	15	67	22	70	6	19	4	37	7
1996	8	2	55	37	55	15	35	14	66	23	70	6	18	4	39	5
1997	8	2	55	37	53	17	35	14	65	23	69	6	18	4	38	7
1998	9	2	55	37	54	17	35	15	62	27	69	7	18	4	37	6
1999	9	2	54	37	54	15	35	14	58	29	68	6	18	4	37	6

2000	8	2	57	34	54	15	35	13	58	29	65	8	18	4	37	6
2001	7	2	56	35	54	15	35	13	58	29	67	6	18	4	36	6
2002	7	3	53	32	54	16	35	13	58	28	66	6	18	3	35	8
2003	7	2	52	24	66	13	35	13	57	29	66	6	18	3	33	9
2004	7	2	54	22	54	13	35	13	56	29	66	6	28	3	27	14
2005	7	2	54	22	53	13	35	13	56	29	66	6	32	3	36	5
2006	9	4	57	20	49	11	35	13	56	29	66	6	32	3	36	5
2007	9	3	58	20	49	11	34	14	56	29	66	6	32	3	27	15
2008	8	3	59	20	49	11	34	14	56	29	66	6	32	3	27	15
2009	9	3	61	19	49	12	34	14	56	29	66	6	32	3	27	15
2010	6	2	62	19	49	12	34	14	56	29	66	6	32	3	27	15
2011	6	2	63	18	50	12	34	13	56	29	66	6	32	3	35	7
2012	6	2	63	18	50	12	34	13	56	29	66	6	32	3	37	4
2013	6	2	63	17	50	12	33	13	55	29	66	6	32	3	37	4
2014	6	2	63	17	50	12	33	13	55	31	66	6	32	3	37	4
2015	6	2	63	17	50	12	33	13	56	29	66	6	32	3	37	4

Table S2 Sown areas in the CLB from 1978 to 2015 (unit: ha). County-scale sown areas is not provided here limited by space.[1-9]

Year	Rice	Wheat	Maize	Bean	Tuber	Peanut	Rape	Sesame	Cotton
1978	514222	82927	2612	8235	25823	21891	83319	1584	33956
1979	513067	83303	2613	8847	26207	21969	83107	1584	33964
1980	506761	79680	1625	7292	20022	21060	95940	1078	34182
1981	486738	83465	1658	8147	19858	19791	100845	1479	34452
1982	487249	86732	1690	8926	19622	18701	113126	1431	33013
1983	468814	90828	1722	9720	19428	17598	118718	1378	32344
1984	493641	98596	1835	8684	10231	19183	158619	1324	31045
1985	488046	68183	1359	10327	19153	24740	173975	2337	24773
1986	491607	58593	1494	10485	18573	26484	186738	2719	23675
1987	501461	61687	1296	9850	18050	24579	213626	1596	24421
1988	493341	72837	1641	9418	18046	25396	167319	1875	24920
1989	495687	68567	1796	9111	18450	25241	161365	2072	24518
1990	490809	83103	1916	8310	17763	23655	180258	1687	28677
1991	447071	80821	3544	7227	15623	20815	192767	2010	39880
1992	453457	68317	3792	7676	16151	20965	190906	2179	43250
1993	448084	85371	6310	13360	13680	24444	163630	2204	42492
1994	436508	78549	5885	12616	12251	24983	169180	2228	50290
1995	431574	77325	7902	12524	12554	22077	184794	2255	54642
1996	436723	87137	7286	14993	12474	22369	164199	2137	58924
1997	419442	94728	10413	16110	13995	24072	175498	2221	61944
1998	410256	91196	13539	16299	13608	24802	194357	2613	54777
1999	397042	81192	19488	17238	15747	28296	203565	3052	45273
2000	371856	68763	18395	18115	16826	29933	209305	4341	53090
2001	337570	56382	21778	20513	15975	24521	216158	5294	61444
2002	336681	53470	21669	20745	14410	23407	218279	5462	58823
2003	326300	48353	22838	20563	14664	22367	217733	5288	70265
2004	400959	74348	17818	19295	10550	18899	219401	4280	64452
2005	405449	42753	18712	18763	10358	18557	221446	4415	64245
2006	417607	48834	15362	18991	11423	19533	211218	3696	68311
2007	406222	72526	14586	22515	9480	16172	163032	3354	69362
2008	418399	76153	14615	17534	9823	17379	173371	3761	76668
2009	416788	77818	16820	18033	11847	17303	193987	3849	71701
2010	410511	85156	19122	15022	11476	18874	182754	4091	70719
2011	401174	91329	18687	14859	11494	18594	163722	3993	74958
2012	407778	93603	19706	15609	11881	18654	154848	3929	73136
2013	409204	97812	21639	15726	10993	19801	131290	4061	66008
2014	416404	97854	22562	15816	11246	20225	122520	4135	54083
2015	416676	99993	25676	16312	11021	20265	115388	4195	50840

Table S3 Fertilizer use in the CLB from 1978 to 2015 (unit: kt).[1-9] C represents compound fertilizer. N represents nitrogen fertilizer. P represents phosphorus fertilizer.

Year	Hefei			Feidong			Feixi			Chaohu			Wuwei			Lujiang			Hanshan			Shucheng
	C	N	P	C	N	P	C	N	P	C	N	P	C	N	P	C	N	P	C	N	P	C
1978	0.0	1.3	0.2	1.1	3.4	1.1	0.9	12.1	7.9	0.2	2.6	0.9	0.0	2.7	1.4	0.1	3.6	2.5	0.0	0.6	0.2	15.7
1979	0.0	1.2	0.3	1.1	3.9	1.1	0.9	12.1	7.9	0.2	4.0	0.9	0.0	4.7	1.4	0.1	5.4	2.5	0.0	1.4	0.2	15.7
1980	0.3	1.8	1.0	2.4	7.2	2.4	0.9	12.1	7.9	0.1	4.1	1.8	0.4	7.5	2.0	0.1	5.2	3.2	0.0	3.9	0.8	15.7
1981	0.2	1.8	0.7	4.7	9.3	4.4	0.9	12.1	7.9	0.4	7.7	1.9	0.5	7.2	2.3	0.1	6.4	3.0	0.3	2.4	0.7	15.7
1982	0.1	2.1	0.9	6.9	11.7	6.4	0.9	12.1	7.9	0.6	9.0	1.9	0.7	7.6	2.7	0.2	7.0	2.7	0.6	2.4	0.5	15.7
1983	0.2	1.7	1.0	9.1	15.1	8.3	0.9	12.1	7.9	0.9	7.3	2.0	0.9	10.2	3.0	0.2	8.8	2.5	0.8	3.8	0.3	15.7
1984	0.3	1.7	0.9	8.7	16.2	7.4	2.0	12.3	7.4	1.1	6.6	2.1	1.1	12.5	3.4	0.2	9.8	2.3	1.1	4.6	0.1	15.7
1985	0.5	1.8	1.5	8.3	14.2	7.1	2.3	16.3	7.8	2.8	11.8	5.1	1.8	15.8	8.3	0.7	9.8	3.0	1.6	5.9	1.2	15.7
1986	0.5	1.4	0.8	10.7	17.9	9.2	1.2	18.2	9.0	2.7	11.7	3.3	1.4	16.4	4.1	0.5	10.9	2.2	1.5	6.1	1.3	17.1
1987	0.4	2.0	0.9	12.7	17.9	11.5	1.0	18.2	11.2	3.1	11.9	2.3	1.9	15.5	4.8	1.8	12.9	1.3	2.1	6.8	1.2	18.5
1988	0.7	2.9	0.8	9.8	18.4	8.4	1.1	18.7	8.2	3.8	13.1	2.3	2.5	16.9	4.6	0.6	13.1	4.7	3.1	6.5	1.5	17.2
1989	0.6	2.3	0.8	11.3	19.1	9.6	1.4	19.4	9.4	4.4	11.3	2.7	2.5	17.4	5.0	0.5	16.2	1.6	3.5	6.5	1.6	20.3
1990	0.5	2.6	0.8	11.8	19.8	10.1	1.3	20.1	9.9	4.4	11.4	3.2	3.0	17.4	5.2	0.7	15.7	4.8	3.1	9.2	1.7	21.2
1991	0.5	2.1	0.8	11.9	19.6	10.0	1.5	19.8	9.8	5.1	15.0	2.8	3.1	16.7	5.0	2.1	14.5	3.7	4.5	8.0	1.6	21.7
1992	0.7	4.1	0.9	12.0	21.8	9.6	4.1	27.7	9.2	5.2	14.9	2.7	3.4	16.6	5.5	1.8	13.6	4.9	5.1	7.2	1.8	20.2
1993	0.8	4.3	0.9	11.9	24.1	9.1	6.6	26.9	8.5	6.5	14.7	2.8	4.3	17.9	5.2	2.6	15.4	5.1	6.6	6.9	1.6	18.0
1994	0.7	4.6	0.8	12.4	26.4	9.3	8.4	26.1	9.3	7.2	14.9	2.8	5.2	18.8	6.2	4.3	17.4	5.4	7.7	7.2	1.7	17.9
1995	0.7	4.1	1.1	16.5	28.8	9.5	10.4	30.1	9.2	8.0	15.0	2.9	6.0	19.6	7.3	5.9	19.4	5.6	8.9	7.5	1.8	19.2
1996	0.2	5.1	0.7	13.6	23.6	7.9	10.8	26.7	8.5	8.7	15.2	2.9	6.9	20.5	8.3	7.6	21.3	5.9	10.0	7.8	1.8	17.8
1997	0.8	3.5	0.9	17.8	21.4	7.7	11.5	33.3	7.0	8.3	15.3	2.8	7.8	21.3	9.4	9.2	23.3	6.1	11.2	8.1	1.9	22.6
1998	1.1	3.7	1.0	19.2	19.1	7.0	10.0	32.6	6.8	14.3	13.3	4.3	5.9	21.2	11.6	11.2	23.0	6.3	11.0	7.7	1.9	23.0

1999	1.5	6.1	1.1	21.8	22.0	12.8	10.6	20.8	3.7	20.2	11.2	4.3	4.0	21.1	11.2	13.2	22.7	5.8	10.8	7.4	1.8	23.0
2000	1.8	5.0	1.2	32.6	30.9	18.6	9.0	17.5	0.5	22.2	11.6	4.3	6.1	22.7	10.9	13.3	26.1	5.2	11.4	7.4	1.7	22.8
2001	1.8	5.5	1.7	27.6	36.7	7.6	12.2	23.0	7.1	21.2	14.0	8.4	6.4	22.7	9.2	16.1	26.1	4.6	10.8	7.2	1.6	22.6
2002	12.2	11.3	5.7	23.0	29.9	6.7	11.4	20.6	9.9	15.2	10.5	4.4	6.4	25.0	9.2	14.1	21.5	4.1	11.3	6.4	1.8	23.3
2003	3.7	8.3	4.7	29.0	24.3	6.3	11.5	19.2	9.0	8.0	16.0	3.1	7.4	25.1	9.3	18.7	23.3	4.6	11.8	6.7	1.7	23.8
2004	3.0	6.3	3.3	26.2	15.6	6.6	13.6	16.8	9.8	8.2	15.0	3.4	8.7	26.2	9.8	19.2	23.4	4.9	12.6	6.7	2.0	21.1
2005	4.2	7.2	3.9	27.3	15.6	6.7	15.6	16.9	9.6	9.6	15.9	3.9	8.8	28.4	10.0	20.0	23.9	6.0	12.4	7.8	2.2	20.6
2006	4.6	12.1	6.8	28.0	15.6	6.9	16.5	15.4	7.0	9.6	17.2	3.7	9.1	28.9	10.3	21.3	21.1	6.4	13.4	6.8	2.4	20.9
2007	4.8	10.5	5.7	30.8	15.3	6.5	17.5	18.7	7.7	10.8	18.0	3.7	9.1	29.5	10.4	21.3	22.4	6.8	11.9	7.2	2.4	21.3
2008	5.1	10.9	5.8	32.5	15.4	6.6	9.7	26.0	11.3	11.5	18.6	3.7	9.3	29.4	10.4	22.4	22.7	6.9	11.7	7.9	2.5	21.9
2009	4.8	8.5	5.4	34.2	18.2	6.6	12.0	27.3	11.1	10.4	18.0	3.2	9.8	29.4	10.6	23.5	23.0	6.2	12.2	8.2	2.9	22.5
2010	5.5	6.7	3.1	37.2	17.6	6.7	12.3	27.4	11.7	10.6	18.5	3.3	9.8	29.5	10.6	25.2	24.7	7.2	26.0	8.8	3.2	23.0
2011	2.0	2.4	0.8	38.9	17.4	6.5	11.4	26.6	11.7	12.5	18.2	4.9	9.9	29.6	10.6	39.6	17.7	7.2	14.6	6.0	2.9	23.2
2012	2.4	1.8	0.7	39.2	17.6	6.4	11.6	25.6	11.8	15.7	14.6	4.1	10.0	29.6	10.7	33.7	17.7	7.2	23.2	5.9	2.9	24.1
2013	2.6	1.9	0.7	33.2	17.8	6.7	12.2	27.8	12.0	15.9	13.8	4.1	8.5	25.2	9.2	33.7	17.7	7.2	14.7	5.7	3.1	24.9
2014	2.8	1.9	0.6	33.1	16.5	6.3	13.1	27.7	10.0	15.6	13.6	4.0	7.4	22.5	8.2	36.4	17.9	7.3	13.8	6.0	2.8	25.6
2015	2.7	1.7	0.6	32.4	14.9	5.8	12.6	25.1	9.8	13.4	12.1	3.5	7.4	22.4	8.2	34.6	16.9	6.7	14.0	5.9	2.7	25.0

Table S4 Crop yield in the CLB from 1978 to 2015 (unit: kt).[1-9] County-scale sown areas is not provided here limited by space.

Year	Rice	Wheat	Maize	Other cereals	Bean	Tuber	Peanut	Rape	Sesame	Cotton	Sugarcane	Vegetable
1978	2230	160	6	9	14	75	35	100	1	17	14	286
1979	2277	194	7	7	14	76	38	109	2	14	14	284
1980	1968	187	4	6	15	56	37	128	2	13	14	343
1981	2262	199	4	7	16	60	35	149	2	15	14	339
1982	2369	212	4	8	17	64	34	170	2	15	14	350
1983	2076	226	5	8	19	68	32	185	1	16	16	347
1984	2490	252	5	8	18	69	35	193	1	20	20	348
1985	2516	172	4	11	18	72	52	255	2	18	25	610
1986	2674	154	5	8	22	75	58	233	3	20	31	722
1987	2658	169	5	9	20	72	54	237	1	20	33	695
1988	2648	203	5	9	20	77	61	134	2	19	23	663
1989	2716	198	6	9	19	77	54	157	2	17	17	719
1990	2796	220	6	7	14	71	49	237	2	26	16	734
1991	1758	136	5	8	9	51	30	191	1	24	13	778
1992	2482	166	14	11	12	65	45	239	2	31	20	804
1993	2686	253	31	46	27	55	61	242	2	35	24	950
1994	2496	236	18	27	25	50	31	230	2	42	22	1049
1995	2486	237	29	27	25	52	50	284	3	48	19	1070
1996	2672	301	27	36	32	55	50	257	3	49	20	1153
1997	2545	371	55	34	35	66	75	273	3	55	21	1283
1998	2657	293	70	22	38	73	71	270	3	52	47	1495
1999	2614	280	96	17	38	94	77	392	4	43	71	1491
2000	2172	235	85	8	42	112	77	379	6	54	86	1291

2001	2161	196	108	8	47	91	63	434	8	69	59	1361
2002	2389	166	118	6	49	101	66	351	8	63	70	1405
2003	1788	125	100	6	47	93	57	355	7	68	69	1564
2004	2865	141	99	4	50	77	56	491	7	78	47	1467
2005	2670	164	91	3	48	74	53	481	7	68	40	1586
2006	3007	209	83	8	55	76	60	472	5	88	70	1650
2007	2812	326	68	4	53	63	50	380	5	75	77	1850
2008	2982	370	76	2	55	73	60	415	6	83	68	1827
2009	3062	378	92	2	54	84	60	462	6	79	51	1965
2010	3051	419	101	2	45	87	71	368	6	79	52	2109
2011	3087	419	95	0	43	100	68	298	6	88	53	2214
2012	2939	402	89	3	40	85	70	373	6	85	52	2340

Table S5 Annual sale and year-end number of animal in the CLB from 1978-2015.[1-9] S and Y represent sale number and year-end number. Unit of pig, cattle and goat is thousand capita. Unit of poultry is million capita. Unit of fish, shrimp, shell and other aquatic product is kt. County-scale sown areas is not provided here limited by space.

Year	Pig		Cattle		Goat		Poultry		Fish	Shrimp	Mollusc	Others
	S	Y	S	Y	S	Y	S	Y	S	S	S	S
1978	790	1382	5	228	2	12	2	1	20	1	0	0
1979	776	1374	5	231	3	12	2	1	21	1	0	0
1980	750	1508	5	237	4	13	2	1	22	1	0	0
1981	717	1322	5	243	3	12	2	1	23	1	0	0
1982	741	1323	5	249	3	10	2	1	24	1	0	0
1983	740	1306	5	253	3	9	2	2	25	2	0	0
1984	826	1395	5	254	3	8	2	2	26	2	0	0
1985	952	1501	4	255	3	9	3	2	27	2	0	0
1986	1062	1582	4	257	5	13	3	2	30	2	1	0
1987	1038	1464	4	261	7	21	3	2	27	2	0	0
1988	1119	1483	5	257	10	20	3	2	33	3	1	0
1989	1124	1534	4	261	11	21	3	2	30	3	1	0
1990	1191	1579	5	262	10	19	3	2	35	3	1	0
1991	1193	1468	8	257	12	15	3	2	35	3	1	0
1992	1251	1521	9	247	12	21	3	3	38	3	1	0
1993	1351	1496	12	245	25	27	4	3	47	3	2	0
1994	1466	1473	14	251	33	27	5	3	71	5	4	1
1995	1532	1466	16	257	37	44	5	3	98	6	5	1
1996	1717	1576	22	263	73	66	6	3	135	11	7	2
1997	2067	1601	28	271	85	58	8	4	167	14	8	2
1998	2043	1578	32	264	72	56	9	4	178	19	9	4
1999	2229	1674	35	257	80	60	10	4	190	25	12	6
2000	2238	1702	39	244	103	84	11	4	198	26	13	7
2001	2384	1435	43	239	132	107	12	4	195	34	15	18
2002	2454	1701	44	260	180	172	12	4	190	35	17	17
2003	2467	1605	44	242	227	177	14	4	201	39	14	13
2004	2539	1545	44	216	229	147	16	5	197	41	12	20
2005	2546	1457	45	213	226	123	17	4	207	42	11	12
2006	2514	1303	45	173	215	119	18	4	212	49	10	19
2007	1915	950	72	124	90	54	12	4	175	47	8	9
2008	2149	1172	64	128	96	78	14	5	179	49	9	11
2009	2251	1226	66	130	99	73	15	6	191	50	10	13
2010	2294	1228	63	139	102	73	15	6	201	53	8	13
2011	2341	1239	59	134	94	64	16	6	206	61	7	11
2012	2452	1265	60	128	103	77	16	7	219	64	7	12
2013	2562	1291	60	122	111	89	16	7	231	67	8	13
2014	2673	1317	60	116	119	101	17	8	244	70	8	14
2015	2784	1343	61	110	128	113	17	8	257	73	9	15

Table S6 Population in the CLB from 1978 to 2015 (unit: thousand).[1-9]

Year	Hefei		Feidong		Feixi		Chaohu		Wuwei		Lujiang		Hanshan		Shucheng	
	Urban	Rural	Urban	Rural	Urban	Rural	Urban	Rural	Urban	Rural	Urban	Rural	Urban	Rural	Urban	Rural
1978	465	245	33	870	39	731	82	600	73	1129	45	920	31	324	90	741
1979	491	248	34	886	43	739	88	606	74	1144	47	930	33	328	90	748
1980	518	252	39	897	47	746	94	612	79	1155	52	941	36	331	90	756
1981	539	256	42	903	51	754	98	616	85	1153	67	953	37	334	90	763
1982	555	260	46	908	54	762	102	621	91	1151	82	966	38	337	90	771
1983	571	259	48	912	50	780	105	625	92	1158	65	958	39	339	90	778
1984	594	259	57	905	52	785	111	625	98	1155	88	985	48	331	90	786
1985	625	257	104	868	64	781	117	623	101	1156	95	978	54	327	77	796
1986	645	257	104	872	77	779	112	629	97	1159	95	981	53	331	87	795
1987	669	258	104	877	81	784	115	628	98	1165	97	982	54	333	89	801
1988	692	261	112	880	84	802	120	633	98	1177	96	998	53	342	91	811
1989	712	265	112	891	86	816	122	637	98	1192	96	1015	54	344	91	823
1990	733	269	114	906	86	830	124	647	99	1208	95	1047	53	351	91	835
1991	753	271	115	913	87	839	127	652	100	1216	96	1055	54	354	92	848
1992	790	270	125	906	95	843	132	653	104	1210	106	1052	55	354	98	852
1993	820	269	126	909	96	853	138	656	104	1214	108	1052	59	353	93	857
1994	867	260	132	910	98	856	182	619	110	1214	110	1054	61	357	90	861
1995	904	255	137	925	101	858	227	582	116	1215	113	1056	62	360	93	868
1996	930	260	140	930	102	863	271	545	122	1216	115	1058	64	363	95	869
1997	972	257	139	944	104	866	277	546	127	1222	118	1052	69	363	98	877
1998	1001	279	140	946	104	841	280	547	130	1223	121	1052	71	363	133	844
1999	1028	275	144	945	110	842	290	547	132	1232	131	1045	72	364	153	825

2000	1075	270	152	949	112	845	299	546	134	1240	135	1050	74	363	156	827
2001	1107	272	152	950	115	844	306	543	137	1251	138	1049	76	363	158	828
2002	1170	295	150	925	120	842	313	540	147	1245	141	1049	77	362	162	827
2003	1250	309	126	940	125	840	321	538	173	1221	145	1044	79	362	161	828
2004	1358	277	125	943	127	842	323	543	178	1216	148	1037	82	360	161	830
2005	1503	250	125	938	131	840	210	651	161	1234	150	1032	83	360	163	829
2006	1605	326	131	957	132	766	221	646	165	1137	151	1016	86	353	164	828
2007	1664	320	137	965	138	773	226	648	171	1242	155	1014	86	356	129	866
2008	1716	319	143	966	140	785	229	649	175	1245	157	1014	86	356	130	870
2009	1763	323	131	961	140	790	235	652	178	1247	158	1015	86	357	130	869
2010	1783	372	134	958	139	794	237	654	179	1245	161	1019	84	361	127	868
2011	1818	365	138	946	139	800	239	657	180	1249	162	1024	84	363	126	874
2012	1857	365	138	948	140	804	239	650	180	1243	165	1026	84	360	124	872
2013	1896	364	139	950	141	809	240	643	179	1238	168	1028	85	357	122	869
2014	1934	364	140	952	142	814	241	636	178	1233	171	1030	85	355	120	867
2015	1973	363	140	955	142	818	241	629	177	1227	174	1032	85	352	117	864

Table S7 Food consumption in the CLB from 1978 to 2015 (unit: kg $capita^{-1}$).[1-9]

Year	Urban								Rural							
	Cereal	Vegetable	Oil	Meat	Chicken	Fish	Egg	Milk	Cereal	Vegetable	Oil	Meat	Chicken	Fish	Egg	Milk
1978	148	121	6	17	3	5	7	4	279	110	1	2	1	1	2	0
1979	148	121	6	17	3	5	7	4	279	110	1	2	1	1	2	0
1980	148	121	6	17	3	5	7	4	279	110	1	2	1	1	2	0
1981	148	121	6	17	3	5	7	5	278	108	1	3	1	1	2	0
1982	145	119	6	17	4	5	7	5	278	106	2	3	1	1	2	0
1983	141	118	6	17	4	6	7	5	278	104	2	4	1	1	2	0
1984	137	116	7	18	5	7	7	6	277	102	3	4	2	1	2	0
1985	134	115	7	18	6	7	7	5	277	101	3	5	2	1	2	0
1986	135	116	7	19	6	7	7	5	281	100	4	6	2	1	2	0
1987	137	116	7	19	5	7	7	4	284	100	4	6	2	1	2	0
1988	138	117	7	20	5	7	7	4	288	100	5	7	2	2	2	0
1989	140	118	7	21	5	7	8	5	293	104	5	7	2	2	2	0
1990	142	119	8	22	5	7	8	5	293	103	5	8	2	2	2	0
1991	136	116	8	22	5	8	8	6	264	83	6	8	2	2	2	0
1992	129	114	8	23	5	9	9	5	264	93	6	7	2	2	3	0
1993	123	111	8	23	5	10	9	5	273	93	6	8	2	2	3	0
1994	117	109	8	23	6	11	9	5	265	86	6	8	2	2	3	0
1995	111	107	8	24	6	12	10	5	257	79	6	8	2	3	4	0
1996	109	119	9	25	5	13	11	5	255	81	7	9	2	4	4	0
1997	98	105	11	26	10	12	18	6	250	80	7	7	3	3	4	0
1998	88	103	10	25	10	13	15	6	245	79	7	8	3	3	4	0
1999	88	110	10	24	12	14	17	8	251	79	7	8	3	4	4	0

2000	83	102	10	24	12	13	15	13	270	80	8	10	4	4	6	0
2001	80	106	10	23	11	13	15	13	259	84	8	11	4	4	5	0
2002	79	104	10	26	13	16	16	13	244	82	8	9	4	5	5	0
2003	80	107	10	27	13	16	17	16	230	81	7	9	4	5	5	0
2004	78	108	11	26	12	10	16	19	210	81	5	8	4	5	5	0
2005	83	109	12	28	12	12	16	16	215	79	6	11	5	5	5	0
2006	82	117	12	32	12	13	17	17	211	76	6	11	4	5	5	1
2007	77	107	11	24	10	11	14	17	195	76	6	9	5	6	5	1
2008	73	117	12	22	10	10	15	18	191	77	7	9	5	6	6	1
2009	64	113	11	24	10	10	15	17	182	77	6	10	5	5	6	1
2010	72	92	10	24	10	12	14	18	175	77	7	10	5	6	6	2
2011	73	102	9	25	11	10	14	19	164	73	8	11	5	6	6	3
2012	71	97	10	25	10	11	14	16	150	69	8	12	6	6	7	3
2013	69	124	16	24	12	16	18	19	149	61	8	13	7	7	7	3
2014	71	101	12	26	9	11	13	19	145	88	14	18	10	8	9	3
2015	70	92	10	26	10	12	13	19	142	91	10	19	11	9	10	4

Table S8 Population density of eight counties in the CLB (capita km^{-2}).

Year	Hefei	Feidong	Feixi	Chaohu	Wuwei	Lujiang	Hanshan	Shucheng
1980	913	422	381	347	504	427	353	403
1995	1301	460	439	379	535	483	388	441
2005	1857	494	459	415	565	504	422	468
2015	2701	495	449	437	585	503	428	475

Table S9 N and P content in crop and straw, and ratio of straw to gain.[10-16]

Type	Crop (%)				Straw (%)				Ratio of straw to gain	
	N		P		N		P			
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
Rice	1.20	1.07-1.30	0.36	0.26-0.46	0.91	0.89-0.93	0.130	0.123-0.137	0.90	0.85-0.95
Wheat	1.95	1.90-2.10	0.41	0.33-0.49	0.65	0.62-0.68	0.080	0.072-0.088	1.30	1.06-1.54
Maize	1.40	1.30-1.50	0.27	0.24-0.30	0.92	0.89-0.95	0.152	0.138-0.166	1.10	0.85-1.35
Other cereals	1.60	1.50-1.80	0.35	0.26-0.49	0.82	0.64-1.00	0.101	0.07-0.132	1.40	0.96-1.84
Bean	5.70	5.60-6.00	0.48	0.40-0.50	1.81	1.67-1.95	0.196	0.171-0.221	1.60	1.23-1.97
Tuber	0.42	0.23-0.60	0.14	0.08-0.20	2.65	2.28-3.02	0.272	0.221-0.323	0.63	0.26-1.00
Peanut	3.20	2.90-3.50	0.31	0.25-0.33	1.82	1.72-1.92	0.163	0.147-0.179	1.50	0.88-2.12
Rapeseed	4.80	3.50-6.00	0.68	0.50-1.21	0.87	0.79-0.95	0.144	0.126-0.162	2.70	2.13-3.27
Sesame	3.30	3.00-3.50	0.59	0.33-0.67	1.31	1.18-1.44	0.150	0.135-0.165	2.82	2.29-3.35
Cotton	1.50	1.00-2.00	0.48	0.31-0.65	1.24	1.08-1.4	0.150	0.131-0.169	5.00	2.54-7.46
Sugarcane	1.10	1.00-1.90	0.14	0.12-0.16	1.10	1.00-1.20	0.140	0.117-0.163	0.08	0.06-0.10
Vegetable	0.24	1.60-3.20	0.06	0.04-0.08	-	-	-	-	-	-

Table S10 Loss rate of N and P applied in cropland, and storage.[17-27]

Category	Chemical Fertilizer (%)		Manure (%)	
	Upland	Paddy field	Applied	Storage
N				
NH_3	11±3	15±3	20±2	25±10
N_2	21±4	33±4	20±10	20±10
N_2O	1.3±1.0	1.3±1.0	1.3±1.0	1.3±1.0
Maximal runoff fraction		9±3		
P				
Maximal runoff fraction		6±3		

Table S11 Live animal weight, partitioning and nutrient content of every part.[10, 11, 28-50]

Type	Weight	% of live weight			kg N/100kg fraction				kg P/100kg fraction			
	kg	Edible	Bone	Other	Edible	Bone	Other	Total	Edible	Bone	Other	Total
Pig	90±9	65±3	9±1	26±3	2.11±0.21	2.95±0.30	2.7±0.30	2.34±0.30	0.162±0.019	3.95±0.70	0.09±0.00	0.50±0.05
Cattle	475±48	47±3	10±1	43±3	3.18±0.20	3.24±0.10	1.89±0.10	2.64±0.19	0.168±0.298	4.40±0.20	0.56±0.05	0.76±0.20
Sheep	40±4	39±4	11±1	50±2	3.04±0.40	3.57±0.40	1.53±0.08	2.35±0.24	0.146±0.040	6.40±0.10	0.48±0.04	1.01±0.50
Poultry	2.0±0.2	53±2	21±1	26±1	3.09±0.20	3.43±0.10	1.82±0.16	2.83±0.31	0.156±0.010	2.26±0.30	0.06±0.00	0.58±0.06
Milk		1						0.48±0.10				0.07±0.01
Eggs		1						2.13±0.12				0.13±0.05
Fish		53±5		47±4	2.66±0.20		2.128±0.201	2.41±0.18	0.202±0.021		0.05±0.00	0.13±0.01
Shrimp		53±5		47±4	2.77±0.21		2.216±0.213	2.51±0.21	0.232±0.300		0.06±0.00	0.15±0.01
Shells		53±5		47±4	2.02±0.30		1.616±0.301	1.83±0.11	0.177±0.060		0.04±0.00	0.11±0.01
Others		53±5		47±4	2.66±0.20		2.128±0.201	2.41±0.18	0.202±0.021		0.05±0.00	0.11±0.01

665 Table S12 N and P content in food.[10-15]

Type	N (%)		P (%)	
	Mean	Range	Mean	Range
Cereals	1.20	1.50-1.80	0.36	0.26-0.49
Vegetable	0.24	1.60-3.20	0.06	0.04-0.08
Oil	0.00	-	0.01	-
Meat	2.78	2.48-3.08	0.159	0.144-0.174
Poultry	3.09	2.89-3.29	0.156	0.146-0.166
Fish	2.66	2.46-2.86	0.202	0.181-0.223
Egg	2.13	2.01-2.25	0.13	0.08-0.18
Milk	0.48	0.38-0.58	0.07	0.06-0.08

666 Table S13 Straw utilization in the CLB from 1978 to 2015. Data are from our empirical
667 investigation and studies by Han et al. (2002) and Gu et al. (2013).[21, 51,53]

Utilization	1978-1985	1986-1995	1996-2005	2006-2015
Straw recycling (%)	33	45	60	72
Livestock feed (%)	27	25	13	8
Biofuel (%)	40	30	27	20

668 Table S14 Recycling rate of manure from 1978 to 2015. Data are from our empirical
669 investigation and studies by Chen et al. (1999), Gu et al. (2013) and Jiang et al. (2015).[21, 52, 53]

Type	1978-1985	1986-1995	1996-2005	2006-2015
Urban resident (%)	90	50	30	10
Rural resident (%)	95	90	85	75
Pig	70%			
Cattle	70%			
Sheep	40%			
Poultry	55%			

Table S15 The treatment rate of urban sewage in the CLB from 1978-2012. Data are from our empirical investigation.

Year	Heifei	Feidong	Feixi	Chaohu	Wuwei	Lujiang	Hanshan	Shucheng
1978-1997	0	0	0	0	0	0	0	0
1998	70	0	0	0	0	0	0	0
1999	70	0	0	0	0	0	0	0
2000	70	0	0	0	0	0	0	0
2001	70	0	0	0	0	0	0	0
2002	70	0	0	0	0	0	0	0
2003	70	0	0	0	0	0	0	0
2004	70	0	0	23	0	0	0	0
2005	70	0	0	23	0	0	0	0
2006	75	35	0	23	80	0	0	0
2007	75	35	0	63	80	0	0	0
2008	75	35	0	63	80	0	0	0
2009	90	35	75	63	80	75	75	75
2010	90	35	75	82	80	75	75	75
2011	90	35	75	82	80	75	75	75
2012	90	70	75	82	80	75	75	75
2013	90	70	75	82	80	75	75	75
2014	90	70	75	82	80	75	75	75
2015	90	70	75	82	80	75	75	75

Table S16 Animal excretion (g capita^{-1} day^{-1}).[14, 54, 55]

Type	Pig	Cattle	Sheep	Poultry
N	28.1±3.4	123.3±15.0	17.9±3.2	1.3±0.5
P	7.9±2.1	25.5±6.0	3.3±0.7	0.5±0.1

Table S17 Original and logarithmic data used in assessing driving factors. UR: urban rate (%). TIN: total N input (Gg N yr^{-1}). TIN: total P input (Gg P yr^{-1}). UR: urban rate (%). Pop: Population (million capita). Diet: proportion of N or P in food consumption from anima derived food (%). CE: proportion of N or P in crop food production for export (%). CE: proportion of N or P in animal food production for export (%).NUEc: nutrient use efficiency in crop farming. NUEa: nutrient use efficiency in animal breeding. The definition of the factors can be found in main text.

Year	N								P							
	TNI	**UR**	**Pop**	**Diet**	**CE**	**AE**	**NUEc**	**NUEa**	**TPI**	**UR**	**Pop**	**Diet**	**CE**	**AE**	**NUEc**	**NUEa**
Non-logarithmic data																
1978	62	13	6.4	7	24	38	47	11	11.8	13	6.4	1	24	43	58	3
1979	68	14	6.5	7	24	36	46	11	11.6	14	6.5	1	25	42	60	3
1980	78	14	6.6	7	12	33	38	10	14.2	14	6.6	2	12	39	48	3
1981	82	15	6.7	8	22	25	41	10	15.0	15	6.7	2	22	32	51	3
1982	87	15	6.8	8	25	18	41	10	16.2	15	6.8	2	25	26	51	3
1983	95	15	6.9	9	16	11	36	10	17.5	15	6.9	2	16	20	44	3
1984	101	16	7.0	10	29	13	39	11	17.1	16	7.0	2	29	21	52	3
1985	114	18	7.0	11	30	20	37	12	22.6	18	7.0	2	30	27	44	4
1986	126	18	7.1	11	33	22	35	13	21.7	18	7.1	3	33	29	47	4
1987	130	18	7.1	12	32	11	34	13	23.7	18	7.1	3	32	20	44	4
1988	139	19	7.3	12	30	16	31	14	23.0	19	7.3	3	30	24	44	4
1989	142	19	7.4	13	29	10	31	13	23.7	19	7.4	3	29	19	43	4
1990	144	19	7.5	13	30	12	33	14	25.8	19	7.5	3	30	20	43	4
1991	150	19	7.6	14	3	7	23	14	26.4	19	7.6	3	2	16	30	4
1992	156	20	7.6	15	29	8	29	14	26.5	20	7.6	3	29	17	39	4
1993	161	20	7.7	16	33	11	31	15	26.2	20	7.7	4	33	19	43	5
1994	172	21	7.8	17	32	20	28	17	29.0	21	7.8	4	31	27	38	5
1995	185	22	7.9	18	34	24	28	18	31.4	22	7.9	4	33	31	37	6

1996	190	23	7.9	20	39	36	27	21	32.3	23	7.9	5	38	41	37	6
1997	205	24	8.0	22	39	44	27	23	35.1	24	8.0	6	38	48	36	7
1998	204	25	8.1	23	43	47	27	24	37.7	25	8.1	6	42	51	35	7
1999	194	25	8.1	24	41	48	30	25	39.6	25	8.1	6	40	52	34	8
2000	211	26	8.2	24	25	46	26	26	43.0	26	8.2	6	24	49	28	8
2001	223	26	8.3	25	28	48	25	27	42.0	26	8.3	6	27	51	29	8
2002	220	27	8.4	27	37	46	26	26	43.7	27	8.4	7	36	49	29	8
2003	219	28	8.5	29	24	46	23	27	42.7	28	8.5	8	23	48	26	8
2004	201	29	8.6	29	52	52	32	28	42.2	29	8.6	8	52	54	36	8
2005	210	29	8.7	31	47	48	30	28	44.4	29	8.7	9	47	50	33	8
2006	214	31	8.7	32	54	48	30	30	45.8	31	8.7	9	54	50	34	9
2007	207	30	8.9	31	54	40	29	32	42.9	30	8.9	9	54	42	34	9
2008	218	31	9.0	32	57	43	29	31	45.0	31	9.0	9	57	45	34	9
2009	223	31	9.0	34	61	46	30	31	45.7	31	9.0	10	60	48	35	9
2010	232	31	9.1	35	61	46	29	31	47.6	31	9.1	10	61	48	34	9
2011	225	31	9.2	37	64	43	29	32	49.8	31	9.2	11	63	46	33	9
2012	219	32	9.2	40	65	44	30	32	49.4	32	9.2	12	65	47	33	9
2013	222	32	9.1	42	66	39	29	32	49.9	32	9.1	14	65	42	32	9
2014	222	33	9.0	45	67	34	30	32	50.2	33	9.0	15	67	38	33	10
2015	214	36	9.1	48	69	30	31	32	49.2	36	9.1	16	69	34	35	9

Logarithmic data

1978	4.13	2.59	1.86	1.90	3.17	3.63	3.85	2.38	2.47	2.59	1.86	0.38	3.19	3.77	4.07	1.21
1979	4.22	2.62	1.88	1.92	3.20	3.57	3.82	2.37	2.45	2.62	1.88	0.39	3.20	3.73	4.10	1.21
1980	4.36	2.67	1.89	1.94	2.50	3.50	3.65	2.34	2.66	2.67	1.89	0.41	2.50	3.67	3.87	1.17

1981	4.40	2.71	1.91	2.03	3.11	3.22	3.71	2.34	2.71	2.71	1.91	0.51	3.11	3.46	3.94	1.18
1982	4.47	2.74	1.92	2.12	3.23	2.88	3.71	2.34	2.78	2.74	1.92	0.61	3.24	3.25	3.93	1.19
1983	4.55	2.74	1.93	2.20	2.76	2.41	3.59	2.34	2.86	2.74	1.93	0.69	2.76	2.99	3.79	1.19
1984	4.62	2.79	1.94	2.30	3.36	2.54	3.66	2.41	2.84	2.79	1.94	0.79	3.36	3.02	3.94	1.23
1985	4.74	2.87	1.95	2.39	3.41	3.00	3.62	2.51	3.12	2.87	1.95	0.88	3.41	3.29	3.78	1.29
1986	4.83	2.89	1.96	2.44	3.51	3.10	3.57	2.56	3.08	2.89	1.96	0.94	3.51	3.37	3.85	1.34
1987	4.87	2.91	1.96	2.48	3.46	2.43	3.53	2.53	3.17	2.91	1.96	0.98	3.46	2.99	3.78	1.33
1988	4.93	2.92	1.98	2.52	3.39	2.79	3.43	2.62	3.14	2.92	1.98	1.03	3.39	3.18	3.77	1.41
1989	4.96	2.93	2.00	2.54	3.38	2.34	3.44	2.60	3.17	2.93	2.00	1.05	3.38	2.94	3.77	1.39
1990	4.97	2.92	2.01	2.56	3.41	2.47	3.49	2.63	3.25	2.92	2.01	1.08	3.41	3.00	3.75	1.42
1991	5.01	2.93	2.02	2.66	1.06	1.96	3.15	2.62	3.28	2.93	2.02	1.19	0.78	2.74	3.40	1.42
1992	5.05	2.98	2.03	2.70	3.37	2.13	3.36	2.67	3.28	2.98	2.03	1.24	3.36	2.81	3.66	1.46
1993	5.08	3.00	2.04	2.74	3.51	2.40	3.44	2.74	3.27	3.00	2.04	1.29	3.50	2.94	3.76	1.52
1994	5.15	3.05	2.05	2.83	3.45	3.01	3.34	2.85	3.37	3.05	2.05	1.39	3.43	3.30	3.64	1.65
1995	5.22	3.10	2.06	2.92	3.52	3.20	3.33	2.92	3.45	3.10	2.06	1.50	3.50	3.43	3.61	1.73
1996	5.25	3.14	2.07	2.99	3.65	3.59	3.30	3.05	3.48	3.14	2.07	1.60	3.64	3.71	3.62	1.87
1997	5.32	3.17	2.08	3.09	3.66	3.78	3.28	3.14	3.56	3.17	2.08	1.71	3.64	3.88	3.58	1.95
1998	5.32	3.20	2.09	3.12	3.76	3.85	3.30	3.19	3.63	3.20	2.09	1.74	3.74	3.93	3.54	2.00
1999	5.27	3.23	2.10	3.16	3.71	3.87	3.41	3.22	3.68	3.23	2.10	1.79	3.69	3.94	3.53	2.03
2000	5.35	3.26	2.11	3.17	3.24	3.83	3.24	3.24	3.76	3.26	2.11	1.81	3.20	3.90	3.34	2.04
2001	5.41	3.27	2.11	3.22	3.33	3.87	3.22	3.29	3.74	3.27	2.11	1.87	3.29	3.94	3.38	2.09
2002	5.39	3.31	2.12	3.31	3.61	3.82	3.24	3.26	3.78	3.31	2.12	1.99	3.59	3.89	3.38	2.06
2003	5.39	3.34	2.14	3.38	3.18	3.82	3.13	3.30	3.76	3.34	2.14	2.09	3.13	3.87	3.25	2.08
2004	5.30	3.38	2.15	3.38	3.95	3.95	3.46	3.33	3.74	3.38	2.15	2.09	3.94	3.99	3.57	2.09
2005	5.35	3.37	2.16	3.43	3.85	3.87	3.40	3.34	3.79	3.37	2.16	2.15	3.84	3.92	3.49	2.10
2006	5.36	3.42	2.16	3.48	3.99	3.87	3.42	3.40	3.82	3.42	2.16	2.22	3.98	3.91	3.53	2.15
2007	5.33	3.42	2.18	3.44	3.99	3.69	3.37	3.45	3.76	3.42	2.18	2.17	3.98	3.75	3.52	2.23
2008	5.39	3.43	2.19	3.47	4.05	3.77	3.38	3.43	3.81	3.43	2.19	2.22	4.04	3.81	3.54	2.19
2009	5.41	3.44	2.20	3.51	4.10	3.83	3.41	3.44	3.82	3.44	2.20	2.28	4.10	3.87	3.57	2.19
2010	5.45	3.44	2.21	3.55	4.12	3.82	3.35	3.44	3.86	3.44	2.21	2.33	4.11	3.87	3.52	2.21

2011	5.42	3.45	2.22	3.62	4.16	3.77	3.36	3.45	3.91	3.45	2.22	2.43	4.15	3.83	3.48	2.23
2012	5.39	3.46	2.22	3.68	4.18	3.79	3.40	3.46	3.90	3.46	2.22	2.51	4.17	3.86	3.49	2.24
2013	5.40	3.48	2.21	3.75	4.18	3.67	3.38	3.47	3.91	3.48	2.21	2.62	4.18	3.74	3.47	2.25
2014	5.40	3.49	2.20	3.81	4.20	3.51	3.39	3.47	3.92	3.49	2.20	2.71	4.20	3.63	3.49	2.25
2015	5.37	3.57	2.21	3.86	4.24	3.39	3.45	3.46	3.90	3.57	2.21	2.78	4.23	3.52	3.54	2.24

677 Table S18 The OLS result.

Factors	Coefficients	*t*-Statistic	Sig.	VIF
N				
Intercept	6.812	7.603	***	
Urban rate	0.858	2.334	**	201.350
Population	-0.183	-0.290		86.936
Dietary	-0.222	-1.335		169.376
Crop export	0.161	6.715	***	3.507
Animal export	-0.077	-1.758	*	12.266
NUE_c	-1.241	-12.964	***	4.707
NUE_a	0.190	0.774		200.422
R-square	0.99			
Sig.	0.000			
P				
Intercept	2.915	2.402	**	
Urban rate	1.043	2.963	***	179.488
Population	0.354	0.563		83.786
Dietary	-0.196	-1.81	*	104.035
Crop export	0.121	4.069	***	5.796
Animal export	-0.121	-2.094	*	8.713
NUE_c	-0.937	-7.03	***	12.672
NUE_a	0.134	0.637		130.753
R-square	0.99			
Sig.	0.000			

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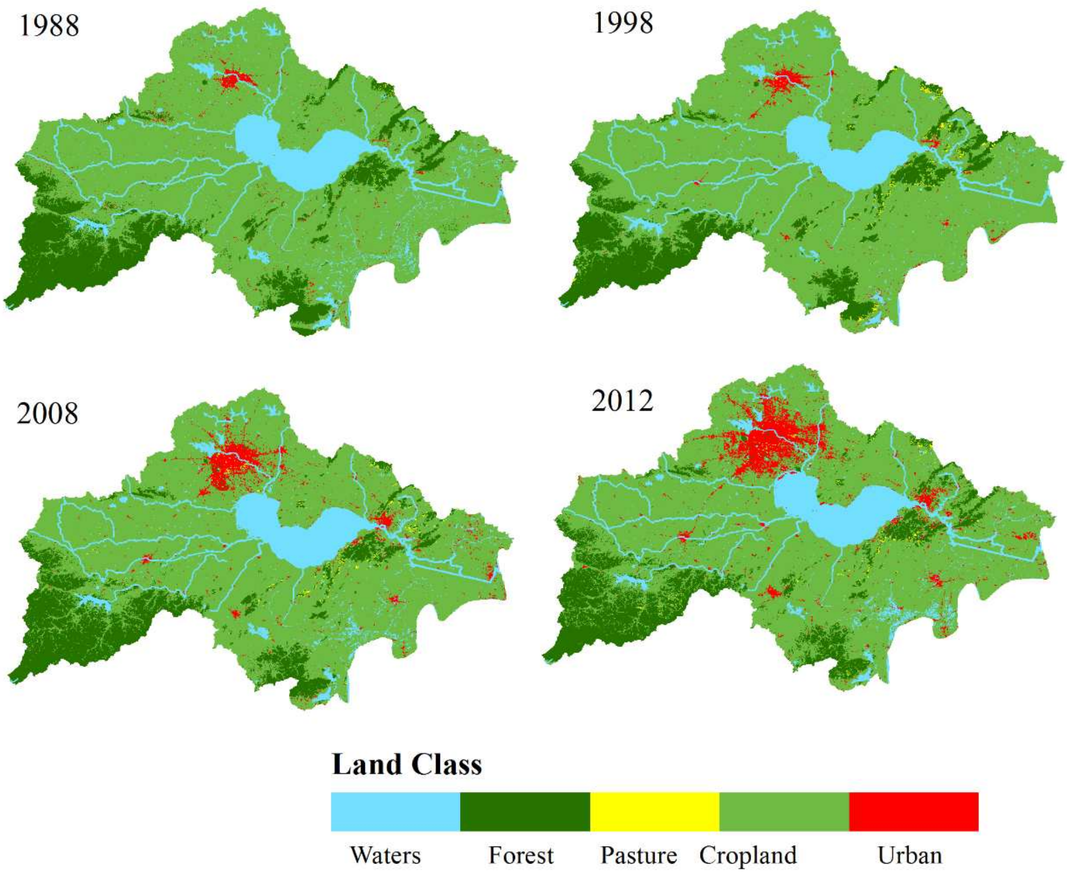

Figure S1 Changes of land use in the CLB in the past decades.

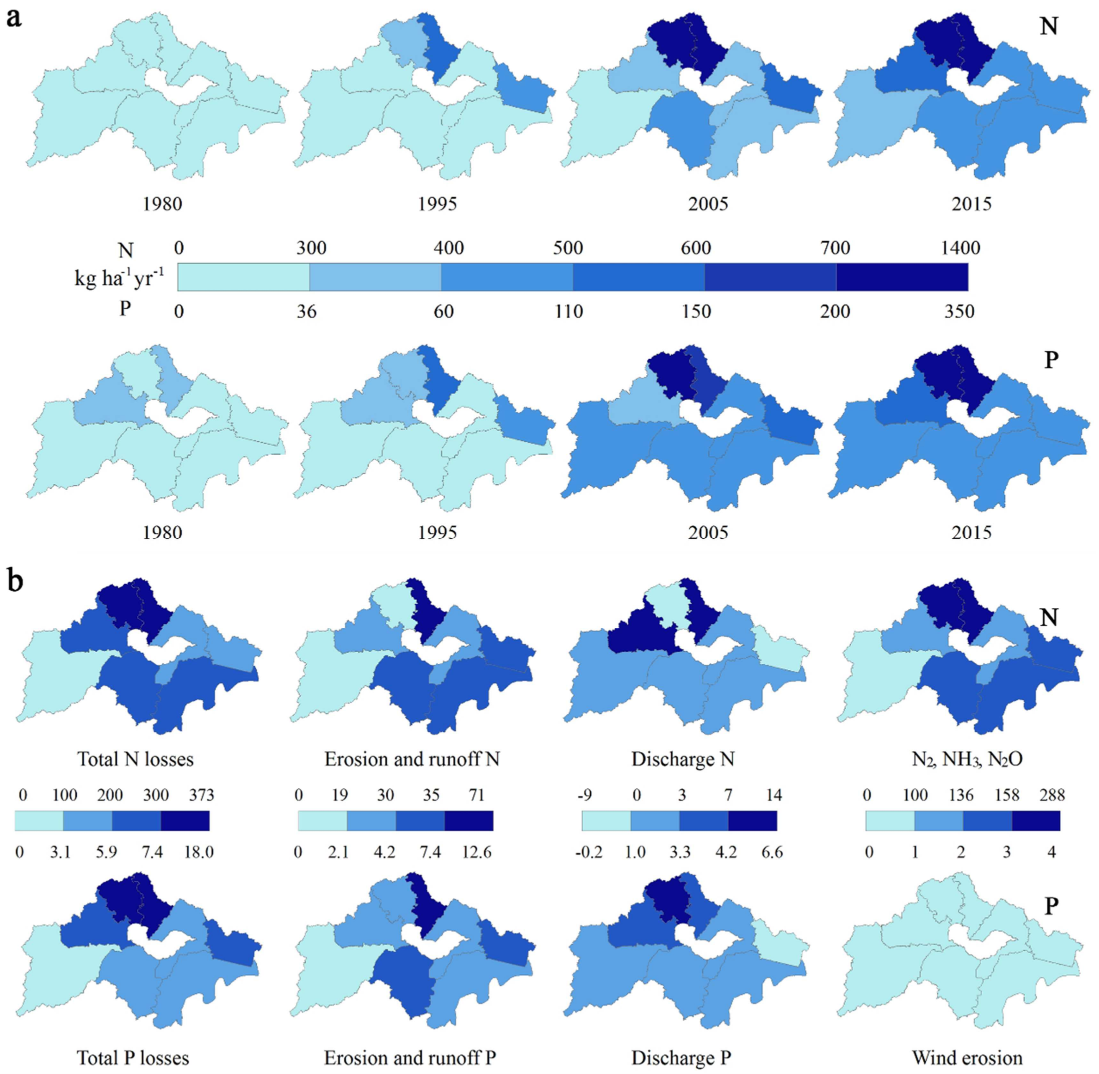

Figure S2 Spatial patterns in nutrient inputs and losses for the eight counties of the Chaohu Lake Basin. (a) TNI and TPI for 1980, 1995, 2005 and 2015. (b) Changes in total N and P losses from 1978 to 2015 for total losses and major sources of N and P loss. Data are presented as kg N or P per cropland area (ha^{-1}).

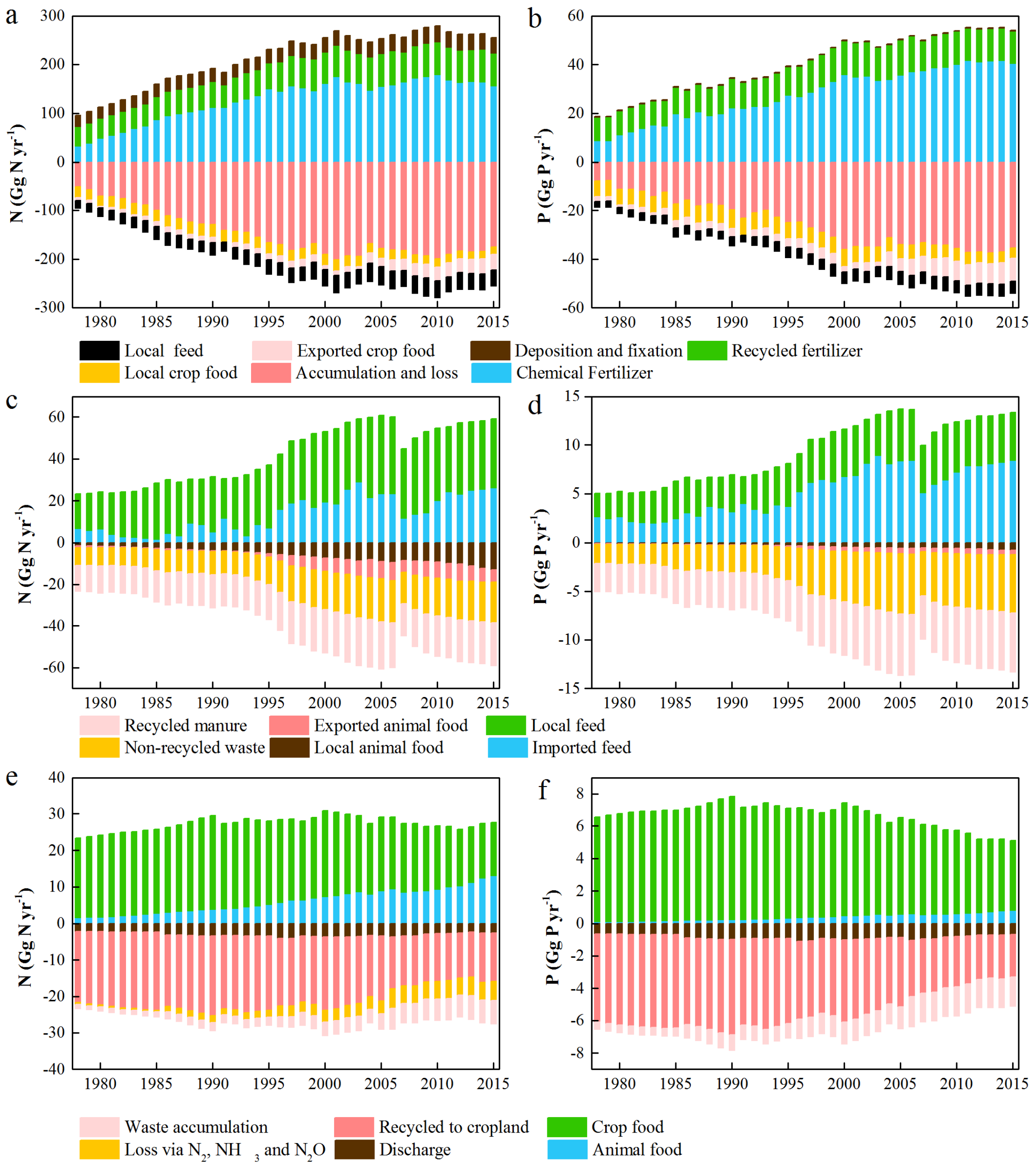

Figure S3 Nutrient inputs and outputs for crop farming, animal breeding and human consumption compartments. (a) N inputs and outputs for crop farming compartment. (b) P inputs and outputs for crop farming compartment. (c) N inputs and outputs for animal breeding compartment. (d) P inputs and outputs for animal breeding compartment. (e) N inputs and outputs for human consumption compartment. (f) P inputs and outputs for human consumption compartment.

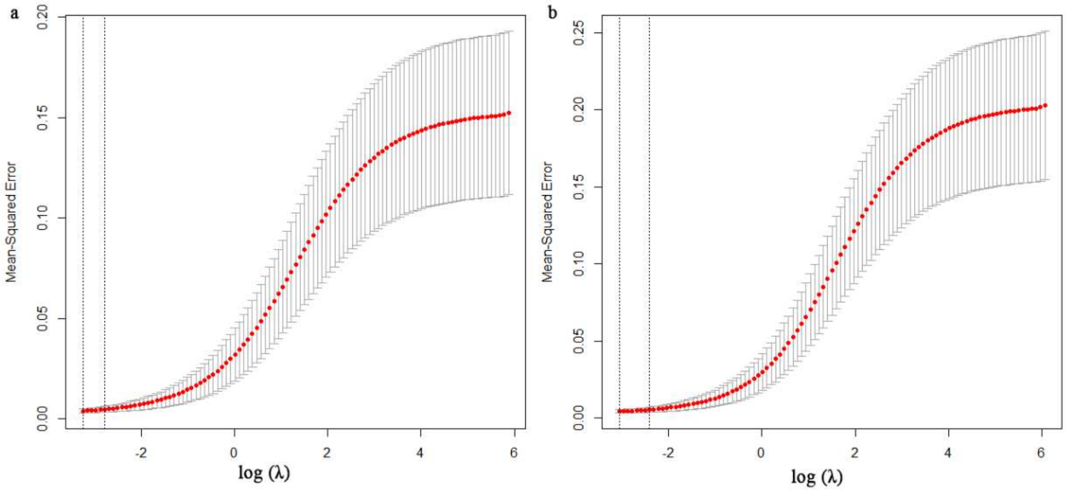

Figure S4 Relation between λ and mean squared error for (a) N and (b) P.

Highlights

- We evaluated changes in N and P flows and their stoichiometry for a basin.
- Human has greatly intensified N and P inputs and losses to the basin in 1978-2015.
- N:P ratio in input and accumulation in the basin declined since the mid-1990s.
- Declining N:P loss to water significantly ($p<0.05$) influenced TN:TP in the lake.
- Expansion of inefficient farming, diet change and urbanization drove the changes.