Managing nitrogen to restore water quality in China

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Abstract (290 words)

The nitrogen (N) cycle has been radically changed by human activities [1]. China consumes nearly 1/3 of the world's N fertilizers, and widespread excessive application of N fertilizers [2, 3] and increased discharge from livestock, domestic and industrial sources have resulted in pervasive pollution of water bodies. Yet challenges in monitoring and quantifying diffusion of N pollution from heterogeneous sources inhibit understanding of safe "boundaries" [4] of N use and effective N management for meeting local water-quality standards. We use a combination of water quality observations and simulated N discharge from agricultural and other sources to estimate spatial patterns of N discharge into water bodies across China for 1955-2014. We find that the critical surface-water quality standard (1.0 $mgN{\bullet}L^{-1}$) was exceeded in most provinces by the mid-1980s, and that current rates of anthropogenic N discharge (14.5±3.1 $MtN{\bullet}yr^{-1}$) to freshwaters are about 2.7 times the estimated 'safe' N discharge threshold (5.3±0.7 $MtN{\bullet}yr^{-1}$). Current efforts to reduce pollution through wastewater treatment (WWT) and improving cropland N management can partially remedy this situation. Domestic WWT has already helped to reduce net discharge by 0.7±0.1 MtN in 2014, but at high monetary and energy costs. Improved cropland N management could cut another 2.3±0.3 $MtN{\bullet}y^{-1}$, about 25% of the excess discharge to freshwaters. Successfully restoring a clean water environment in China will further require transformational changes to boost the national nutrient recycling rate from its current average of 36% to ~87%, a level typical of traditional Chinese agriculture. Though ambitious, such a high level of N recycling is technologically achievable at a capital cost of approximately \$100 billion and operating costs of \$17-25 $billion{\bullet}yr^{-1}$, and would have co-benefits including recycled wastewater for crop irrigation, improved environmental quality and ecosystem services, and new economic opportunities for farmers and fertilizer industries.

Keywords: Sustainability, nutrient recycling, wastewater treatment, thresholds, water quality

Main text (1840 words)

The Earth's biogeochemical cycles have been strongly affected by human activity. For example, the amount of reactive nitrogen (N) entering the global environment increased from ~15 million tons (Mt) in 1860 to 185 Mt in 2010 [1, 5], while agricultural use of N fertilizers has expanded from 12 Mt in 1961 to 110 Mt in 2014 [6]. Though critical for crop yields and food production, human inputs of reactive N to terrestrial and freshwater ecosystems cause water pollution (e.g. NO_3-N, NH_4-N) and air pollution (NH_3, NO_x), as well as global warming and stratospheric ozone depletion (N_2O) [7].

China has dramatically increased its food production over the past four decades. Domestic grain production has risen from 132 Mt in 1950 to 607 Mt in 2014 without expanding total planting area (Fig.S1). This has been achieved through massive increases in synthetic fertilizer use (Fig.1), along with increasingly efficient agricultural technologies, practices and expanded irrigation infrastructure. China now accounts for about 32% [6] of the world's consumption of N fertilizers. To illustrate the critical role of N inputs on crop production, we use the nitrogen-based DeNitrification and DeComposition (DNDC) biogeochemical model [8] and agricultural data from 2403 counties to simulate major grain crops (rice, wheat and maize) during the period 1955-2014 (Fig.S2, S3; Methods). Fig.1b shows that 45±3% of current grain yields can be attributed to use of synthetic N fertilizers. Similar conclusions have been reached following controlled long-term experiments in China [9] and other locations [10, 11] (Table S1).

Meanwhile, traditional practices of recycling organic wastes as fertilizers have been largely abandoned. The availability of subsidized synthetic N fertilizers and new sewage infrastructure have resulted in nutrient-recycling rates dropping sharply from >90% in the late 1970s to 36% in 2014 (Fig.1c, d). Changing N management practices have caused average nitrogen use efficiency (NUE) in China's croplands to decrease significantly from the 1960s to the 2010s [2, 3]. Increased N discharges from croplands and non-recycled organic wastes have resulted in pervasive water pollution.

Quantifying safe levels [4] of N discharge to the environment is a prerequisite for effective N management. Rockstrom et al. (2009) proposed 25% (35 MtN•yr^{-1}) of current human N fixation from the atmosphere as a global "safe boundary", but qualified this estimate as "a first guess" [12]. Recent studies have attempted to clarify the global safe boundary for N released by human activities using simplified N-budget models [5, 13], but relationships among N discharge, N concentrations in freshwater bodies and biotic responses to N in water bodies are complex and often differ by region. Thus, large uncertainties remain regarding regional 'safe' N discharge thresholds, owing to insufficient understanding of spatiotemporal heterogeneity in biogeochemical and hydrological processes. The concept of a safe boundary is most useful when applied to regional and local scales, where practical management options are available. Yet, local to regional safe N boundaries have not been established across representative regions. Here we use observations of water quality in representative rivers and lakes across China to characterize regional thresholds of total N discharge to the water environment, assess excess N, and evaluate the potential for reducing N pollution.

We collected observational data on concentrations of total N (TN) in water bodies and estimate provincial N discharge into the inland aquatic environment for 1955-2014. Here, N discharge comprises anthropogenic sources from croplands, human and livestock excrement, organic garbage, and industrial waste (Fig.2,3; Table S1; Fig.S4-6; Methods). N concentrations in water bodies were low before the 1980s (typically <1.0 mgN•L^{-1}), but increased rapidly to levels exceeding 15 mgN•L^{-1} in many catchments after the 1990s. Groundwater concentrations of NO_3-N also increased sharply after the 1980s, with stable isotope analysis implicating agricultural and domestic sources [14]. The Ministry of Water Resources classified 80.2% of groundwater samples (from 2103 wells; Fig.S7) as polluted in 2015 [15].

We then identified empirically the critical N discharge threshold for each province as total anthropogenic N discharge during the year when pollution levels first breached the water-quality standard (1.0 mgN•L^{-1} [16, 17], Methods; Fig.2,3; Table S3) in representative catchments. Aggregating N-discharge thresholds from catchment to national scale leads to a novel estimate of the national N discharge threshold (5.3±0.7 MtN•yr^{-1}; Fig.3b). This input can be conceptualised as China's share of the total planetary safe N boundary. The current national anthropogenic N discharge rate is 14.5±3.1 MtN•yr^{-1} (2010-2014 average), well above the threshold. Agricultural systems are responsible for 59% of current N discharge (35% croplands, 24% livestock). Another 39% is attributed to domestic waste (13% urban sewage, 8% rural sewage, 18% organic garbage) and the remaining 2% to industrial waste.

Although China's declared target of zero growth in chemical fertilizer use by 2020 [18] has already been achieved, synthetic N use was still 30.5 Mt in 2016. Proposed remedies for maintaining agricultural productivity and reducing negative environmental impacts have centered on improved N management (INM) in croplands: applying the right fertilizer products, at the right rate, at the right time, in the right place [2]. To assess the potential benefits of INM in China's croplands, we use the DNDC model to estimate the minimum synthetic N input for maintaining historical yields (Methods) through broadcasting of fertilizers (BF) or the use of controlled-release fertilizers (CRF). Fig.3 shows that INM could reduce cropland N discharge from current (2010-2014 average) 5.1±0.3 MtN•y^{-1} to 2.8-3.0 MtN•y^{-1} (range encompasses the CRF and BF scenarios), and also fluxes of NH_3 (by 39~72%) and N_2O emissions (by 47~55%). INM could thus reduce current total cropland N inputs from 36.9±0.4 Mt to 24.1~26.9 MtN•yr^{-1}, or the use of synthetic N from 29.8±0.3 MtN•yr^{-1} to 17.0~19.8 MtN•yr^{-1}. If all smallholder farmers adopted INM practices [19], this alone could reduce the excess anthropogenic N discharge (9.2±2.0 MtN•y^{-1}, Methods) by 23-25%.

Quantifying safe boundaries at small geographical scales is helpful for clarifying more detailed environmental issues and local management options. Fig.4a-c summarizes the critical challenges for N management at the provincial level. Under current N management (CNM), cropland N discharge by itself already exceeds critical pollution thresholds in 14 of 31 provinces. INM-CRF could lower cropland pollution to below the critical thresholds in 29 provinces, but two provinces cannot reach safe thresholds without lowering food production (Inner Mongolia by 36%, Shaanxi by 12%).

Wastewater treatment (WWT) is another primary solution for reducing point-source water pollution, not only by N, but also with by phosphorus and enteric bacteria in inland water bodies. Domestic WWT has expanded considerably since the 1980s. In 2014, about 49.43 billion m^3 (Bm^3) of municipal wastewater (75% of urban water use) was treated, with energy consumption of 14.8 billion kWh. The total economic cost (infrastructure and operations) of WWT was about \$20.8 billion (\$0.42/m^3 [20]), equivalent to 2.2% of national rural household income (population 619 million) or 0.2% of GDP in 2014. But the amount of N removed by WWT was only 0.70±0.1 MtN (~26% of total municipal sewage N), because only 56% of WWT plants had N-removal facilities [21], with an average N removal rate of 55% [22]. Average efflux concentrations in treated water released to the environment were 14.3-16.5 mgN•L^{-1} [21, 23]. Further improvements in N-removal efficiency (e.g. tertiary WWT) are feasible, and have been achieved in some wastewater treatment works, although at additional capital [24] and operational costs [25].

China needs a holistic strategy to mobilise and integrate all relevant socio-economic sectors to cut effectively N pollution, not just from croplands but also from livestock, domestic and industrial wastes. As 63% of current N discharge to freshwaters is from non-recycled livestock and domestic waste, future policies should pursue a transformational expansion of nutrient-recycling systems, together with water, sanitation and hygiene (WASH) programmes. Results from long-term (>20 years) fertilisation experiments in China indicate that combining synthetic fertilizers with manure can improve soil quality and generate 8.2-9.9% larger yields of rice, maize and wheat compared to synthetic fertilizers alone [9]. Fig.4c, d demonstrate that reducing N discharge by enough to return to provincial safe thresholds, though daunting, could nearly be achieved by implementing INM and increasing the national nutrient-recycling rate to 86-88%. Rates in nine provinces would need to exceed 95% under INM-CRF. Even with a recycling rate of 100%, Shanghai would still need to cut industrial N discharge by 37%, and Shanxi must either reduce food production by 3% or cut industrial N discharge by 80%. Raising national nutrient-recycling rates to 86-88% could allow food productivity to be maintained at current levels while further reducing synthetic N requirements under INM from 17-19.8 MtN•yr^{-1} to 10.8~13.6 MtN•yr^{-1} (assuming equivalent NUE for organic and synthetic N). Side effects of increased recycling might include a rise in N_2O emissions [26].

We evaluate the costs of three strategies to achieve our proposed increase in nutrient recycling: 1) traditional wet manure recycling, 2) dry compost recycling, and 3) direct wastewater recycling, with consideration of the potential impacts of new toilet technologies and infrastructure changes (Methods). We estimate that the operational costs of recycling organic waste would range from \$8-26 billion•yr^{-1} based on 2010 prices (Table.S4). Traditional recycling is the most expensive option and carries WASH risks. A more practical approach is to deliver domestic wastewater for irrigation by separating industrial wastewater from domestic wastewater and connecting household sewage systems with existing irrigation systems. This approach would cost ~\$100 billion for infrastructure, with operational costs of \$17-25 billion•yr^{-1}.

Overall, the costs of building nutrient recycling systems are small relative to the annual cost of water pollution at current levels, estimated at 1.5% of national GDP in 2010 (\$91 billion; \$20 billion from treatment and \$71 billion from impacts of environmental degradation) [27]. Farmers would benefit more from nutrient recycling than from WWT because the former offers potential increases in job opportunities and income in rural areas. The human health risks of enhanced nutrient recycling could be reduced by sealing domestic wastewater systems to minimize physical contact and by enhancing wastewater disinfection (e.g., chlorination, aerobic treatment, ozonation and/or UV light). Further improvements in livestock-waste management (~14.3 MtN•yr^{-1} in excretion), such as manipulating animal diets, trapping particulate emissions and applying methane tanks, could also reduce air and water pollution [28], increase manure-N recycling rates and improve sanitation. These initiatives would create new market opportunities for the fertiliser industry [29] and farmers.

The massive challenges that China must overcome to restore safe and sustainable levels of N in water bodies are also shared by other regions where water pollution is increasing because of excess nutrient discharge. This includes many parts of Asia, South and Central America, and sub-Saharan Africa [7]. Well-defined targets for N releases into the local environment are essential for formulating effective regional policies to reduce pollution, which are in turn essential for progress at the global level (for example staying within "planetary boundaries"). Given the many environmental problems related to different types of N compounds and the complexity of the N cycle, coordinated approaches to nutrient management contribute to remaining within multiple planetary boundaries (climate, air quality, biodiversity) and have widespread co-benefits, including conserving water resources, lowering both air and water pollution, reducing N_2O-induced stratospheric ozone depletion, and increasing rural incomes.

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Figures

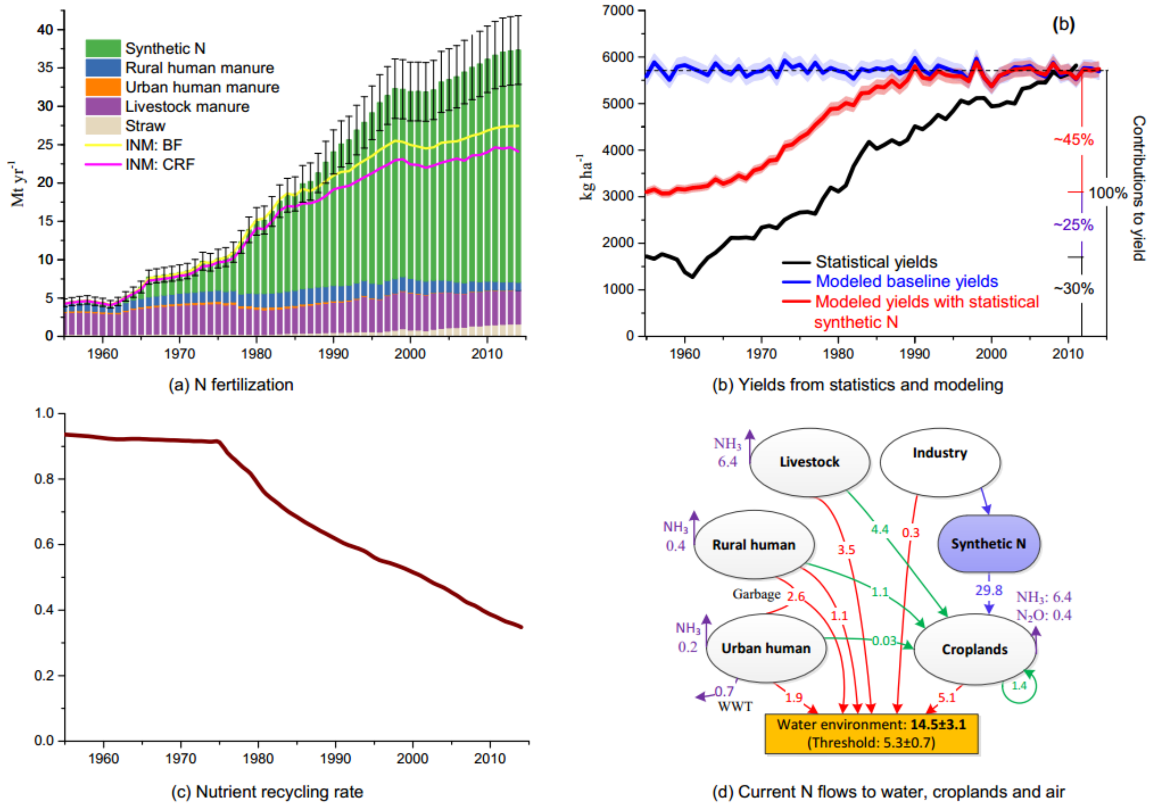

Fig.1 The changing nitrogen cycle and its contributions to food production in China during 1955-2014. (a) Nitrogen application in croplands (stacked bars); the yellow and purple lines mark our model-based estimates of the minimum nitrogen required to maintain annual food production under the two INM scenarios without changes in nutrient recycling rates (BF: broadcasting of fertilizer; CRF: controlled-release fertilizers). Error bars indicate upper and lower bounds on total N applied to croplands (see Methods). (b) Contribution of synthetic nitrogen (~45%, red line) to grain productivity. The blue line shows climate-driven variations in modeled grain yields (rice, maize and wheat) under the baseline scenario (current cropping with average agricultural inputs for 2007-2011; see Methods). The red line shows variations in modeled grain yields due to variations in climate and historical synthetic nitrogen inputs. The black line shows actual yields as recorded in the statistical yearbooks. The unexplained 25% growth in grain yields between 1955 and 2010 is attributed to additional contributions from irrigation expansion and technological advances that coupled with increased N inputs. (c) The broken nitrogen cycle: increases in synthetic nitrogen application are associated with declining rates of nutrient recycling since the late 1970s. The recycling rate is defined as the ratio of nitrogen from organic waste returned to croplands against the amount of nitrogen in organic waste, excluding emissions to the atmosphere. (d) The major anthropogenic N flows to croplands (green arrows: organic N, blue line: synthetic N), water (red arrows) and the atmospheric

(purple arrows) environment (2010-2014 average). N removal via WWT is based on data from 2014. Imported N is counted in our assessment of excretion (Fig.S8).

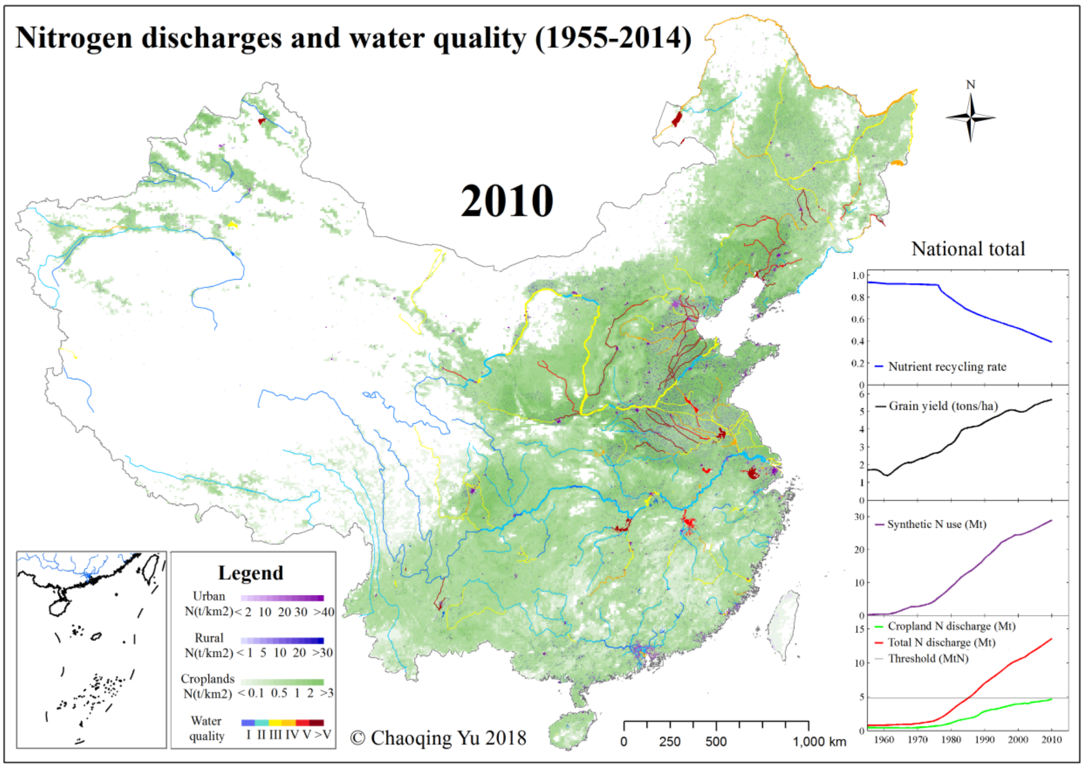

Fig.2 Reconstruction of N discharge during 1955-2014 and the associated evolution of surface-water quality in terms of TN (data sources in Methods). Raster maps of cropland N are based on leaching and runoff simulated by the DNDC model. Urban N includes discharges of human sewage, industrial waste and garbage at provincial levels. Rural N includes discharges of rural human sewage, livestock manure and garbage at provincial levels. Observed national grain yields, synthetic N use, and estimated nutrient recycling and discharge rates are shown in the panels to the right of the map. To view an animation of this figure, open the attached powerpoint file, and initiate the slide show. The animation will begin automatically and step through successive years.

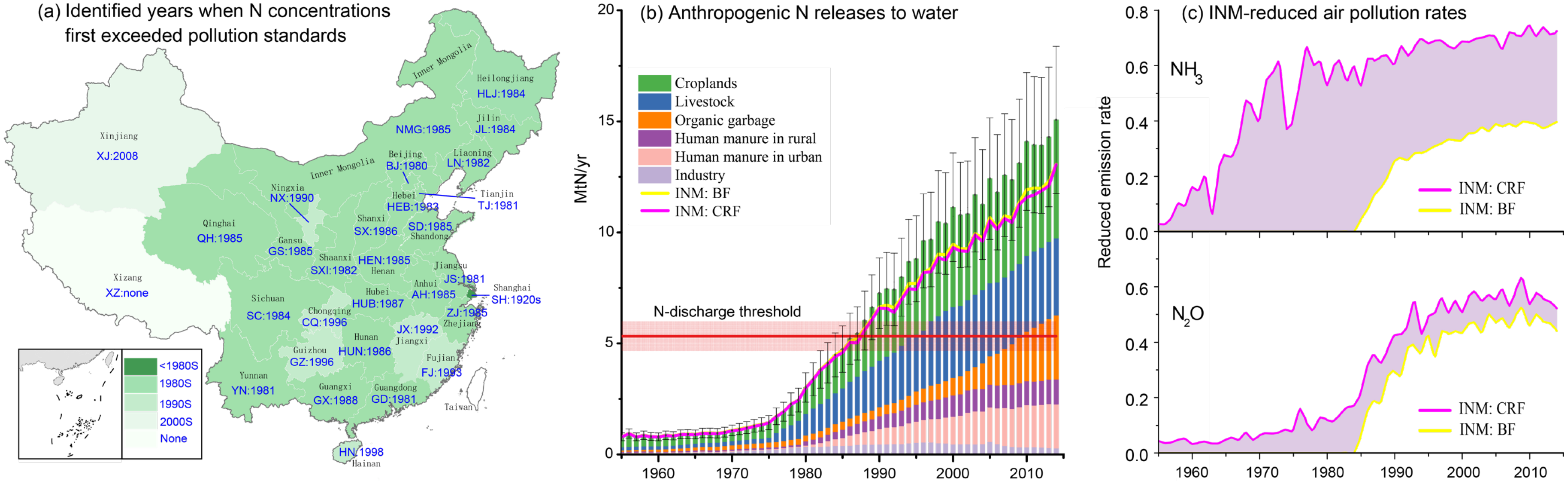

Fig.3 Thresholds of N discharge to the water environment and the potential contributions of INM in reducing N pollution. (a) The years in which N pollution first exceeded the national standard in each province (see Table S3 for representative catchments and references). (b) 60-year records of nitrogen discharge (runoff and leaching) to the water environment in mainland China from anthropogenic sources. The yellow and purple lines mark simulated minimum N discharge amounts through INM under the BF and CRF scenarios, respectively. The critical threshold is estimated by aggregating provincial-level N discharge amounts from the years marked in (a). Error bars indicate estimated upper and lower bounds of total N discharge into the water environment (see Methods). (c) Predicted reductions in emissions of air pollution (NH_3 and N_2O) from croplands are estimated by substituting CNM with INM. Controlled-release fertilizers (CRF) cannot significantly reduce N discharge to the aquatic enviroment relative to broadcasting of fertilizer (BF) under INM, but do reduce N loss in the form of NH_3.

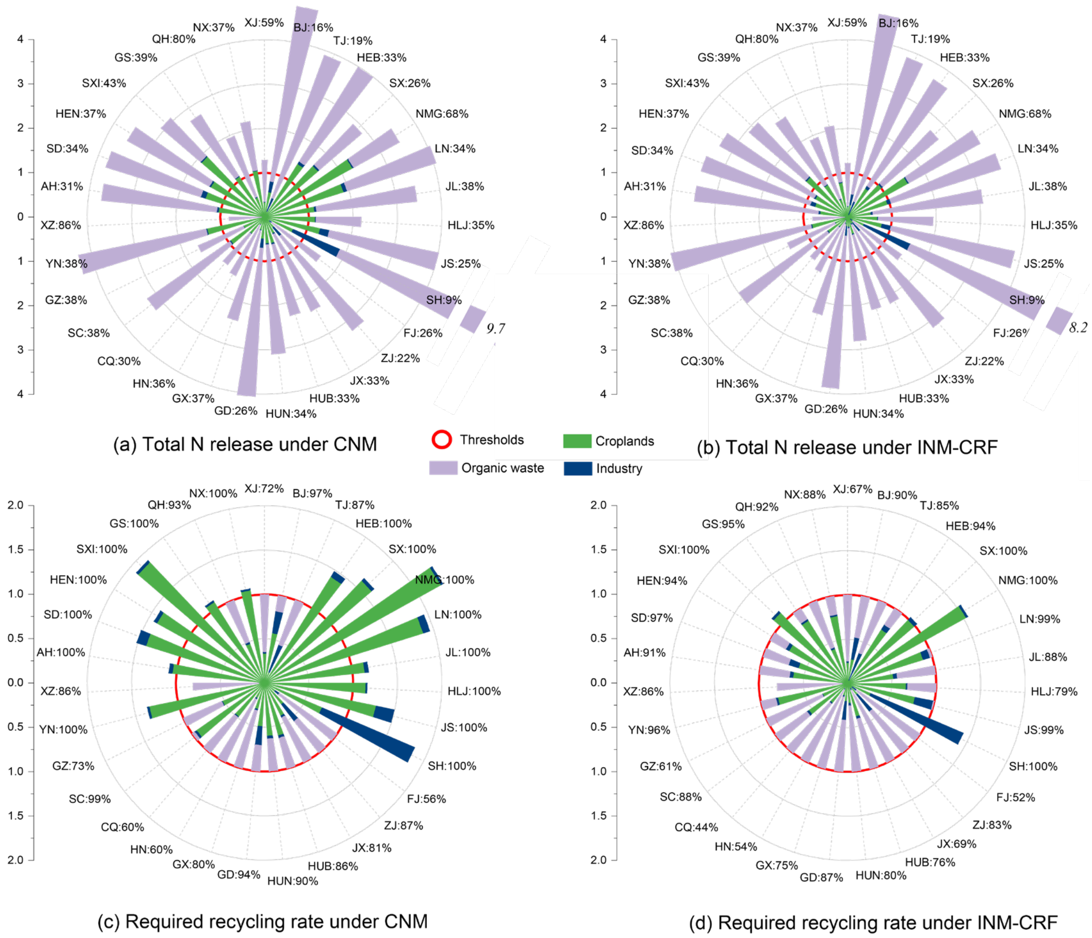

Fig.4 Anthropogenic nitrogen discharge and requirements to meet the critical threshold in each province of mainland China. Upper panels show provincial-level N discharge (see Fig.3a for locations) into the water environment under (a) current nitrogen management (CNM) and (b) improved nitrogen management (INM-CRF) in croplands based on 2010-2014 mean values. Percentages listed in (a) and (b) are current nutrient recycling rates. Lower panels list the minimum N recycling rates (in percentage of recyclable solid-liquid organic wastes) required for each province to meet its critical pollution threshold under (c) CNM and (d) INM-CRF. All discharge and recycling rates are normalized against the critical pollution threshold (marked as a red circle).

This Figure will appear in SI, for enhancing Fig.1d. It was required by the reviewer #4.

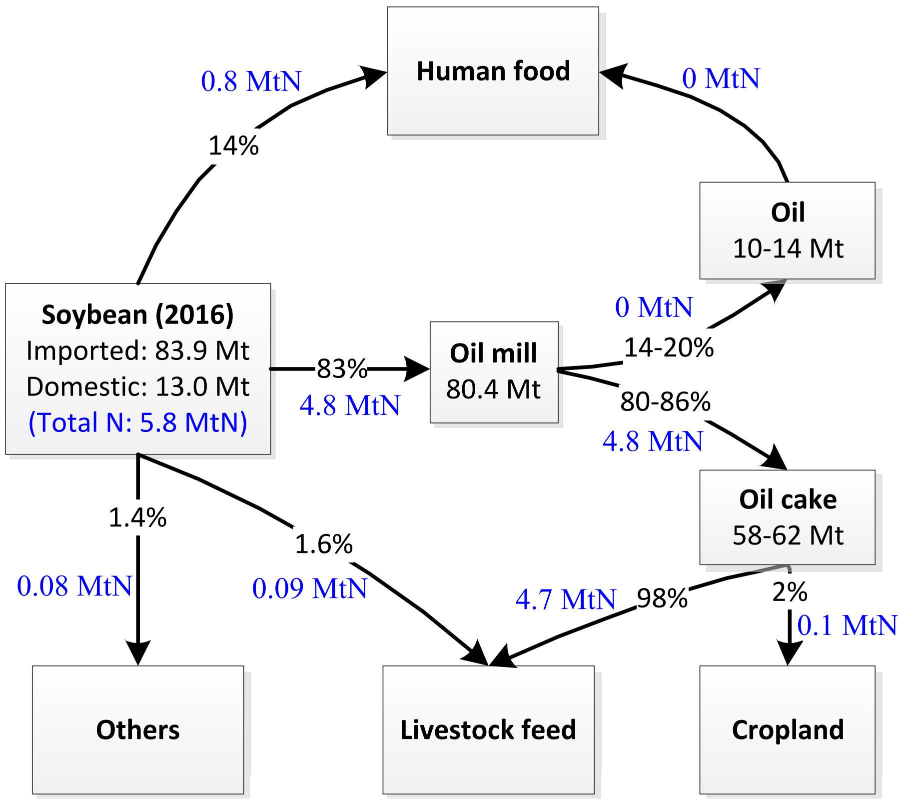

Fig.S8 An approximate estimate of China's soybean consumption (biomass values in black text) and N budget (in blue text) in 2016. In recent years imported grain has increased substantially in China, and reached 114.6 Mt in 2016. Most of the imported grain was in the form of soybeans (73%). This imported N has been considered in our study in estimates of both human diet and livestock feed.

(Sources: (1) Imported grain data, http://www.yuanbangtouzi.com/a/job/shichanggenzong/2017/0125/448.html
(2) fraction of soybean consumption, http://www.chyxx.com/industry/201805/638760.html, in Chinese, access on Sep.17, 2018,
(3) fraction of oil cake consumption, http://www.feedtrade.com.cn/technology/nutrition/ingredient/2012-08-22/2007111_2.html, access on Sep.17, 2018)

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