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1 **Managing nitrogen to restore water quality in China**

2

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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·L⁻¹) was exceeded in most provinces by the mid-1980s, and that current rates of anthropogenic N discharge (14.5±3.1 MtN·yr⁻¹) to freshwaters are about 2.7 times the estimated 'safe' N discharge threshold (5.3±0.7 MtN·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·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·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

58

59 **Main text** (1840 words)

60

61 The Earth's biogeochemical cycles have been strongly affected by human activity.
62 For example, the amount of reactive nitrogen (N) entering the global environment
63 increased from ~15 million tons (Mt) in 1860 to 185 Mt in 2010 ^[1, 5], while agricultural
64 use of N fertilizers has expanded from 12 Mt in 1961 to 110 Mt in 2014 ^[6]. Though
65 critical for crop yields and food production, human inputs of reactive N to terrestrial
66 and freshwater ecosystems cause water pollution (e.g. NO₃-N, NH₄-N) and air
67 pollution (NH₃, NO_x), as well as global warming and stratospheric ozone depletion
68 (N₂O) ^[7].

69

70 China has dramatically increased its food production over the past four decades.
71 Domestic grain production has risen from 132 Mt in 1950 to 607 Mt in 2014 without
72 expanding total planting area (Fig.S1). This has been achieved through massive
73 increases in synthetic fertilizer use (Fig.1), along with increasingly efficient
74 agricultural technologies, practices and expanded irrigation infrastructure. China now
75 accounts for about 32% ^[6] of the world's consumption of N fertilizers. To illustrate the
76 critical role of N inputs on crop production, we use the nitrogen-based DeNitrification
77 and DeComposition (DNDC) biogeochemical model ^[8] and agricultural data from
78 2403 counties to simulate major grain crops (rice, wheat and maize) during the period
79 1955-2014 (Fig.S2, S3; Methods). Fig.1b shows that 45±3% of current grain yields can
80 be attributed to use of synthetic N fertilizers. Similar conclusions have been reached
81 following controlled long-term experiments in China ^[9] and other locations ^[10, 11]
82 (Table S1).

83

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

91

92 Quantifying safe levels ^[4] of N discharge to the environment is a prerequisite for
93 effective N management. Rockstrom et al. (2009) proposed 25% (35 MtN·yr⁻¹) of
94 current human N fixation from the atmosphere as a global "safe boundary", but
95 qualified this estimate as "a first guess" ^[12]. Recent studies have attempted to clarify
96 the global safe boundary for N released by human activities using simplified N-budget
97 models ^[5, 13], but relationships among N discharge, N concentrations in freshwater
98 bodies and biotic responses to N in water bodies are complex and often differ by region.
99 Thus, large uncertainties remain regarding regional 'safe' N discharge thresholds,
100 owing to insufficient understanding of spatiotemporal heterogeneity in biogeochemical
101 and hydrological processes. The concept of a safe boundary is most useful when

102 applied to regional and local scales, where practical management options are available.
103 Yet, local to regional safe N boundaries have not been established across representative
104 regions. Here we use observations of water quality in representative rivers and lakes
105 across China to characterize regional thresholds of total N discharge to the water
106 environment, assess excess N, and evaluate the potential for reducing N pollution.

107
108 We collected observational data on concentrations of total N (TN) in water bodies
109 and estimate provincial N discharge into the inland aquatic environment for 1955-2014.
110 Here, N discharge comprises anthropogenic sources from croplands, human and
111 livestock excrement, organic garbage, and industrial waste (Fig.2,3; Table S1; Fig.S4-6;
112 Methods). N concentrations in water bodies were low before the 1980s (typically <1.0
113 $\text{mgN}\cdot\text{L}^{-1}$), but increased rapidly to levels exceeding $15 \text{ mgN}\cdot\text{L}^{-1}$ in many catchments
114 after the 1990s. Groundwater concentrations of $\text{NO}_3\text{-N}$ also increased sharply after the
115 1980s, with stable isotope analysis implicating agricultural and domestic sources ^[14].
116 The Ministry of Water Resources classified 80.2% of groundwater samples (from 2103
117 wells; Fig.S7) as polluted in 2015 ^[15].

118
119 We then identified empirically the critical N discharge threshold for each province
120 as total anthropogenic N discharge during the year when pollution levels first breached
121 the water-quality standard ($1.0 \text{ mgN}\cdot\text{L}^{-1}$ ^[16, 17], Methods; Fig.2,3; Table S3) in
122 representative catchments. Aggregating N-discharge thresholds from catchment to
123 national scale leads to a novel estimate of the national N discharge threshold (5.3 ± 0.7
124 $\text{MtN}\cdot\text{yr}^{-1}$; Fig.3b). This input can be conceptualised as China's share of the total
125 planetary safe N boundary. The current national anthropogenic N discharge rate is
126 $14.5\pm 3.1 \text{ MtN}\cdot\text{yr}^{-1}$ (2010-2014 average), well above the threshold. Agricultural
127 systems are responsible for 59% of current N discharge (35% croplands, 24%
128 livestock). Another 39% is attributed to domestic waste (13% urban sewage, 8% rural
129 sewage, 18% organic garbage) and the remaining 2% to industrial waste.

130
131 Although China's declared target of zero growth in chemical fertilizer use by 2020
132 ^[18] has already been achieved, synthetic N use was still 30.5 Mt in 2016. Proposed
133 remedies for maintaining agricultural productivity and reducing negative
134 environmental impacts have centered on improved N management (INM) in croplands:
135 applying the right fertilizer products, at the right rate, at the right time, in the right
136 place ^[2]. To assess the potential benefits of INM in China's croplands, we use the
137 DNDC model to estimate the minimum synthetic N input for maintaining historical
138 yields (Methods) through broadcasting of fertilizers (BF) or the use of
139 controlled-release fertilizers (CRF). Fig.3 shows that INM could reduce cropland N
140 discharge from current (2010-2014 average) $5.1\pm 0.3 \text{ MtN}\cdot\text{y}^{-1}$ to $2.8\text{-}3.0 \text{ MtN}\cdot\text{y}^{-1}$ (range
141 encompasses the CRF and BF scenarios), and also fluxes of NH_3 (by 39~72%) and
142 N_2O emissions (by 47~55%). INM could thus reduce current total cropland N inputs
143 from $36.9\pm 0.4 \text{ Mt}$ to $24.1\text{-}26.9 \text{ MtN}\cdot\text{yr}^{-1}$, or the use of synthetic N from 29.8 ± 0.3
144 $\text{MtN}\cdot\text{yr}^{-1}$ to $17.0\text{-}19.8 \text{ MtN}\cdot\text{yr}^{-1}$. If all smallholder farmers adopted INM practices ^[19],
145 this alone could reduce the excess anthropogenic N discharge ($9.2\pm 2.0 \text{ MtN}\cdot\text{y}^{-1}$,

146 Methods) by 23-25%.

147

148 Quantifying safe boundaries at small geographical scales is helpful for clarifying
149 more detailed environmental issues and local management options. Fig.4a-c
150 summarizes the critical challenges for N management at the provincial level. Under
151 current N management (CNM), cropland N discharge by itself already exceeds critical
152 pollution thresholds in 14 of 31 provinces. INM-CRF could lower cropland pollution
153 to below the critical thresholds in 29 provinces, but two provinces cannot reach safe
154 thresholds without lowering food production (Inner Mongolia by 36%, Shaanxi by
155 12%).

156

157 Wastewater treatment (WWT) is another primary solution for reducing point-source
158 water pollution, not only by N, but also with by phosphorus and enteric bacteria in
159 inland water bodies. Domestic WWT has expanded considerably since the 1980s. In
160 2014, about 49.43 billion m³ (Bm³) of municipal wastewater (75% of urban water use)
161 was treated, with energy consumption of 14.8 billion kWh. The total economic cost
162 (infrastructure and operations) of WWT was about \$20.8 billion (\$0.42/m³ [20]),
163 equivalent to 2.2% of national rural household income (population 619 million) or
164 0.2% of GDP in 2014. But the amount of N removed by WWT was only 0.70±0.1 MtN
165 (~26% of total municipal sewage N), because only 56% of WWT plants had
166 N-removal facilities [21], with an average N removal rate of 55% [22]. Average efflux
167 concentrations in treated water released to the environment were 14.3-16.5 mgN·L⁻¹ [21,
168 23]. Further improvements in N-removal efficiency (e.g. tertiary WWT) are feasible,
169 and have been achieved in some wastewater treatment works, although at additional
170 capital [24] and operational costs [25].

171

172 China needs a holistic strategy to mobilise and integrate all relevant
173 socio-economic sectors to cut effectively N pollution, not just from croplands but also
174 from livestock, domestic and industrial wastes. As 63% of current N discharge to
175 freshwaters is from non-recycled livestock and domestic waste, future policies should
176 pursue a transformational expansion of nutrient-recycling systems, together with water,
177 sanitation and hygiene (WASH) programmes. Results from long-term (>20 years)
178 fertilisation experiments in China indicate that combining synthetic fertilizers with
179 manure can improve soil quality and generate 8.2-9.9% larger yields of rice, maize and
180 wheat compared to synthetic fertilizers alone [9]. Fig.4c, d demonstrate that reducing N
181 discharge by enough to return to provincial safe thresholds, though daunting, could
182 nearly be achieved by implementing INM and increasing the national
183 nutrient-recycling rate to 86-88%. Rates in nine provinces would need to exceed 95%
184 under INM-CRF. Even with a recycling rate of 100%, Shanghai would still need to cut
185 industrial N discharge by 37%, and Shanxi must either reduce food production by 3%
186 or cut industrial N discharge by 80%. Raising national nutrient-recycling rates to
187 86-88% could allow food productivity to be maintained at current levels while further
188 reducing synthetic N requirements under INM from 17-19.8 MtN·yr⁻¹ to 10.8~13.6
189 MtN·yr⁻¹ (assuming equivalent NUE for organic and synthetic N). Side effects of

190 increased recycling might include a rise in N₂O emissions [26].

191
192 We evaluate the costs of three strategies to achieve our proposed increase in
193 nutrient recycling: 1) traditional wet manure recycling, 2) dry compost recycling, and 3)
194 direct wastewater recycling, with consideration of the potential impacts of new toilet
195 technologies and infrastructure changes (Methods). We estimate that the operational
196 costs of recycling organic waste would range from \$8-26 billion·yr⁻¹ based on 2010
197 prices (Table.S4). Traditional recycling is the most expensive option and carries WASH
198 risks. A more practical approach is to deliver domestic wastewater for irrigation by
199 separating industrial wastewater from domestic wastewater and connecting household
200 sewage systems with existing irrigation systems. This approach would cost ~\$100
201 billion for infrastructure, with operational costs of \$17-25 billion·yr⁻¹.

202
203 Overall, the costs of building nutrient recycling systems are small relative to the
204 annual cost of water pollution at current levels, estimated at 1.5% of national GDP in
205 2010 (\$91 billion; \$20 billion from treatment and \$71 billion from impacts of
206 environmental degradation) [27]. Farmers would benefit more from nutrient recycling
207 than from WWT because the former offers potential increases in job opportunities and
208 income in rural areas. The human health risks of enhanced nutrient recycling could be
209 reduced by sealing domestic wastewater systems to minimize physical contact and by
210 enhancing wastewater disinfection (e.g., chlorination, aerobic treatment, ozonation
211 and/or UV light). Further improvements in livestock-waste management (~14.3
212 MtN·yr⁻¹ in excretion), such as manipulating animal diets, trapping particulate
213 emissions and applying methane tanks, could also reduce air and water pollution [28],
214 increase manure-N recycling rates and improve sanitation. These initiatives would
215 create new market opportunities for the fertiliser industry [29] and farmers.

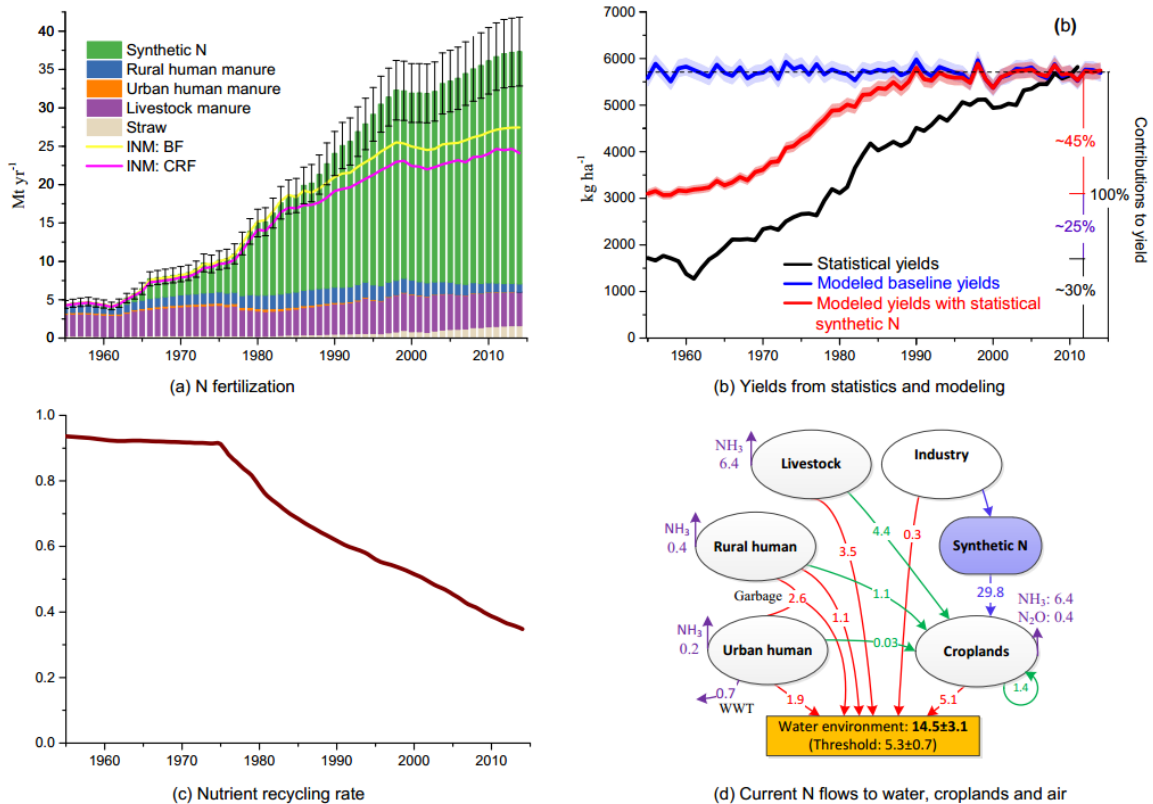
216
217 The massive challenges that China must overcome to restore safe and sustainable
218 levels of N in water bodies are also shared by other regions where water pollution is
219 increasing because of excess nutrient discharge. This includes many parts of Asia,
220 South and Central America, and sub-Saharan Africa [7]. Well-defined targets for N
221 releases into the local environment are essential for formulating effective regional
222 policies to reduce pollution, which are in turn essential for progress at the global level
223 (for example staying within “planetary boundaries”). Given the many environmental
224 problems related to different types of N compounds and the complexity of the N cycle,
225 coordinated approaches to nutrient management contribute to remaining within
226 multiple planetary boundaries (climate, air quality, biodiversity) and have widespread
227 co-benefits, including conserving water resources, lowering both air and water
228 pollution, reducing N₂O-induced stratospheric ozone depletion, and increasing rural
229 incomes.

230
231
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236

237 **Figures**

238

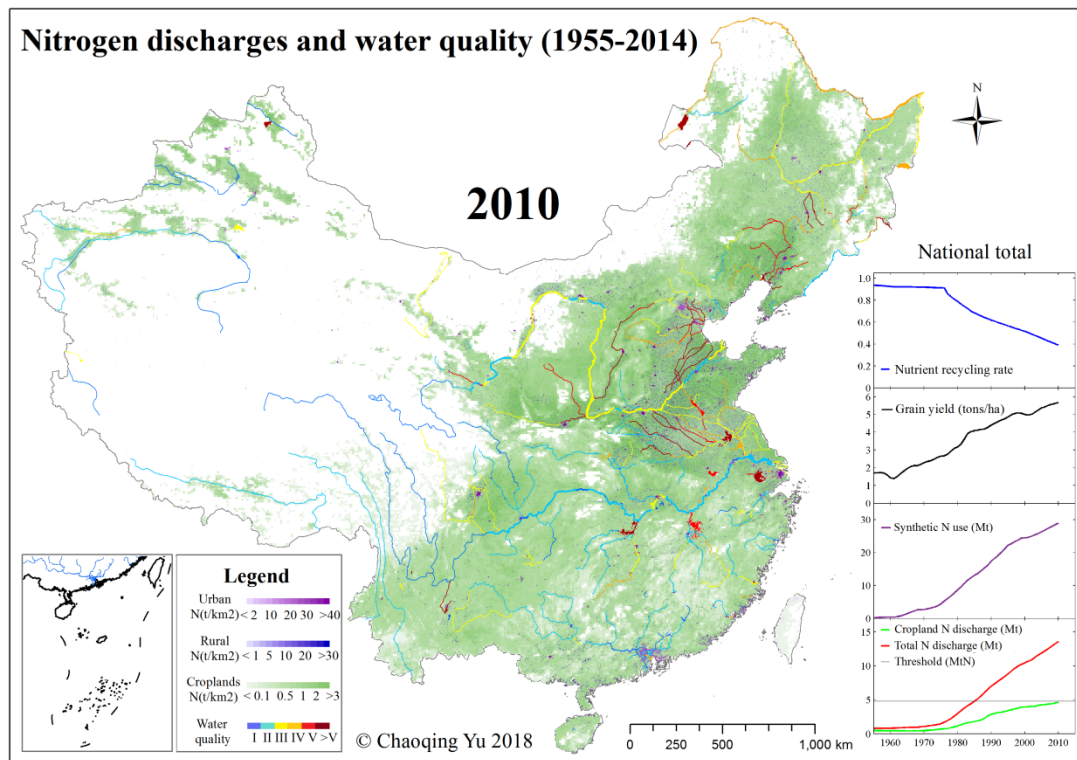


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240

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

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



263
 264 Fig.2 Reconstruction of N discharge during 1955-2014 and the associated evolution of
 265 surface-water quality in terms of TN (data sources in Methods). Raster maps of
 266 cropland N are based on leaching and runoff simulated by the DNDC model. Urban N
 267 includes discharges of human sewage, industrial waste and garbage at provincial levels.
 268 Rural N includes discharges of rural human sewage, livestock manure and garbage at
 269 provincial levels. Observed national grain yields, synthetic N use, and estimated
 270 nutrient recycling and discharge rates are shown in the panels to the right of the map.
 271 To view an animation of this figure, open the attached powerpoint file, and initiate the
 272 slide show. The animation will begin automatically and step through successive years.

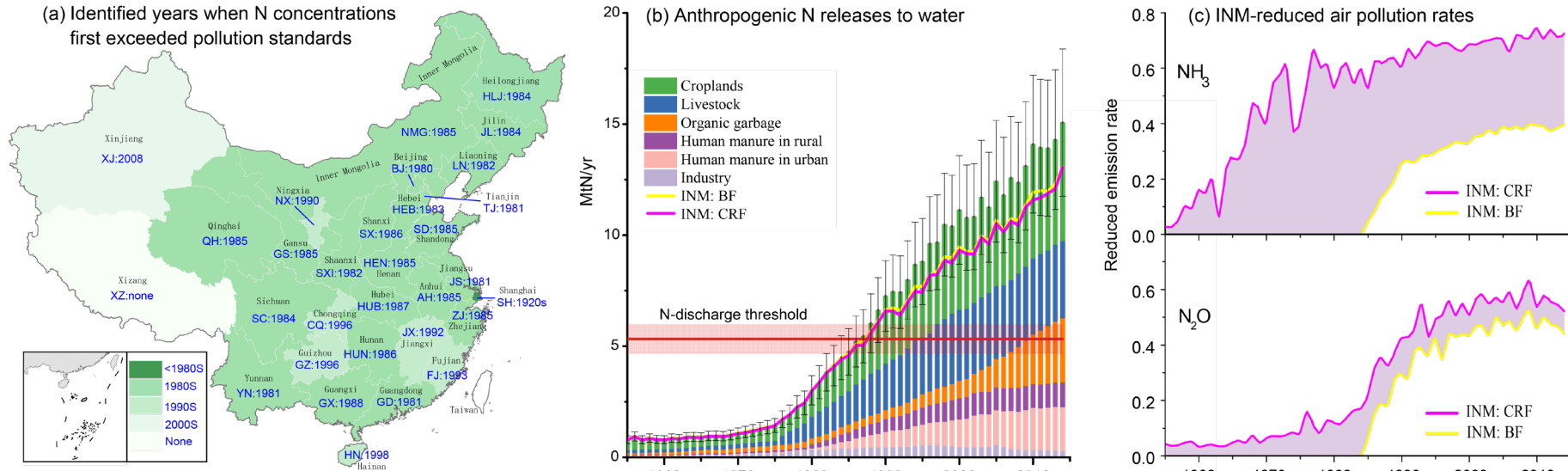
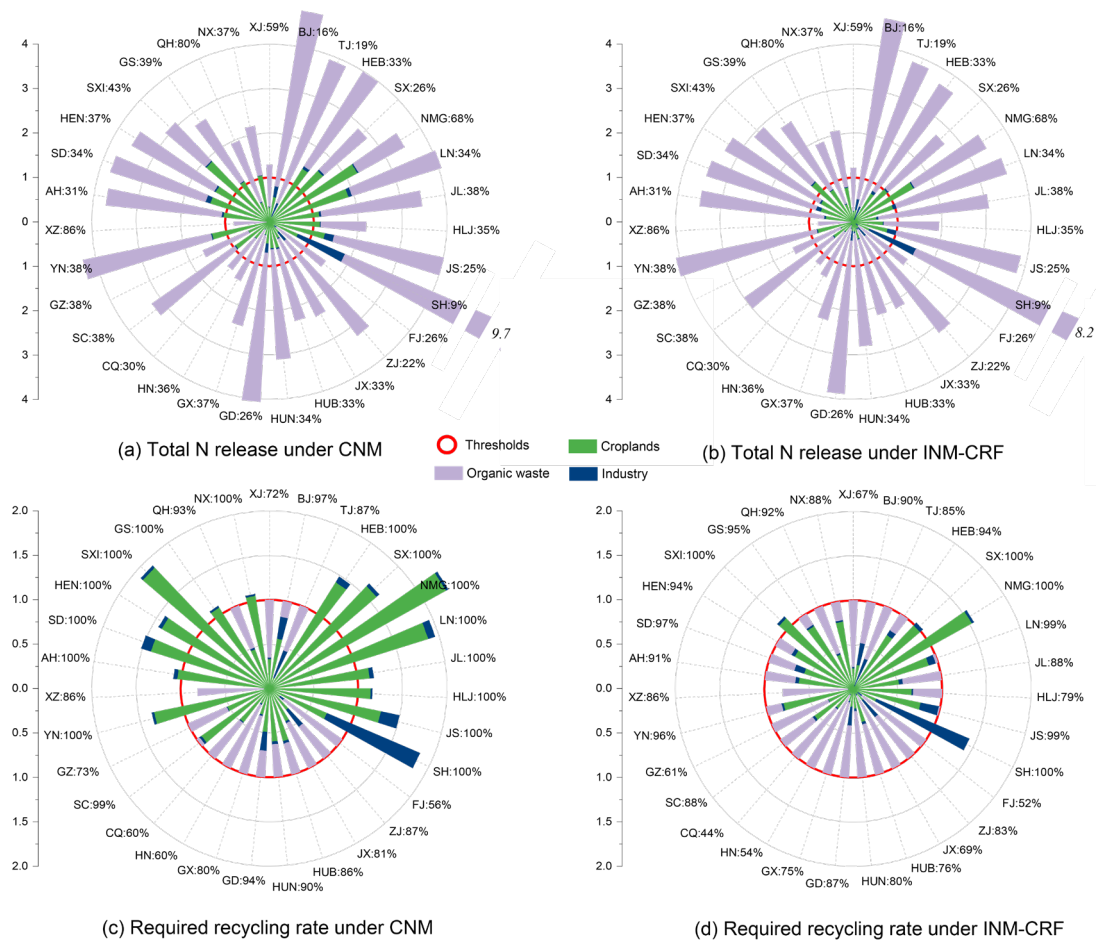


Fig.3 Thresholds of N discharge to the water environment and the potential contributions of INM in reducing N pollution. (a) The years in which N pollution first exceeded the national standard in each province (see Table S3 for representative catchments and references). (b) 60-year records of nitrogen 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 environment relative to broadcasting of fertilizer (BF) under INM, but do reduce N loss in the form of NH_3 .



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

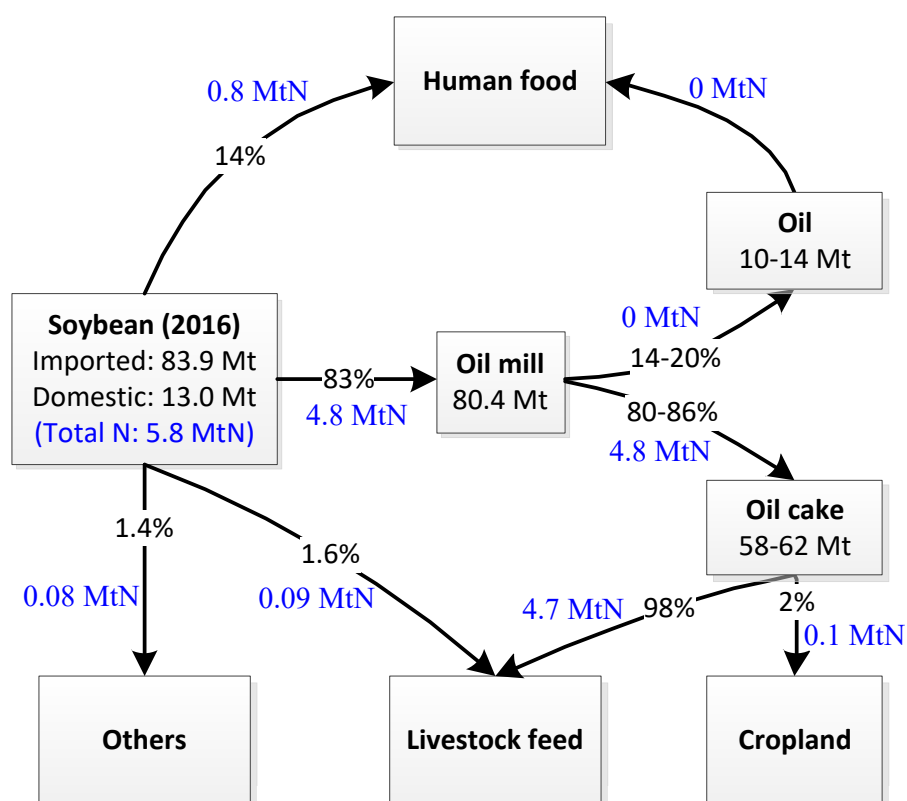


Fig.S8 An approximate estimate of China's soybean consumption (biomass values in black text) and N budget (in blue text) in 2016. In recent years imported grain has increased substantially in China, and reached 114.6 Mt in 2016. Most of the imported grain was in the form of soybeans (73%). This imported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