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4	Nitrogen surplus benchmarks for controlling N pollution in the main cropping									
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23 ABSTRACT: Nitrogen (N) surplus is a useful indicator for improving agricultural N management and controlling N pollution. Few studies have developed benchmark values for 24 25 cropping systems in China, a country with the largest N fertilizer use in the world. We 26 established N surplus benchmarks for 13 main cropping systems, at optimal N management, 27 which accounted for about 50% of total N fertilizer consumption in Chinese agriculture, using 28 results from >4,500 on-farm field experiments and a soil surface balance approach. The results 29 showed that N surplus benchmarks for single cropping systems ranged from 40 to 100 kg N ha⁻ 30 ¹ yr⁻¹ (average 73 kg N ha⁻¹ yr⁻¹), while for double cropping systems ranged from 110 to 190 kg N ha⁻¹ yr⁻¹ (average 160 kg N ha⁻¹ yr⁻¹), roughly twice that of single cropping systems. These 31 32 N surplus benchmarks could be further reduced as declines of N deposition and reactive N 33 losses towards to the "4R"s of nutrient stewardship through improving fertilization techniques 34 and agronomic managements. Our N surplus benchmarks could serve as realistic targets to improve the N management of current conventional practices, and thereby could lay the 35 36 foundations for a more sustainable N management in China.

37 KEYWORDS: N input, N output, N surpluses, N use efficiency, China, wheat, maize, rice,
38 rapeseed



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41 42 43 Nitrogen (N) is a main nutrient, and needed to boost crop growth and development. It is delivered to the crop mainly through fertilizers, manure and the mineralization of organic 44 45 matter and crop residues in soil. Nearly half of the world's population are currently nourished by crops grown with N fertilizers.^{1, 2} However, improper use of N fertilizer may result in poor 46 47 crop yield and/or crop quality, and lead to soil and environmental degradation, e.g., too little N 48 is used in sub-Saharan Africa which results in low crop yields and soil N mining; too much N is used in China which leads to serious environmental pollution.^{2, 3} Consequently, improving 49 agricultural N management is crucial for producing more nutritious food for a growing global 50 51 population, while maintaining or improving soil fertility and minimizing adverse environmental impacts. One of the key enabling steps for improving N management in 52 53 cropping systems is the development of critical indicators and realistic benchmarks for 54 evaluating the performance of N management.

55 Of the many indicators for assessing N management, two in particular may be useful for 56 policy, i.e. nitrogen use efficiency (NUE) and N surplus. There are many functional definitions 57 of NUE, e.g. agronomic efficiency, recovery efficiency, physiological efficiency and partial factor productivity,⁴ and they have mainly focused on the efficiency of fertilizer N input. In 58 59 contrast, the NUE concept used in the present study focused on the efficiency of all N inputs; 60 it was based on a soil nitrogen balance calculation and included the main N inputs such as 61 fertilizers, atmospheric deposition, and biological N fixation. Here, NUE is defined as the 62 efficiency of all the N inputs transferring to harvested crop N, which is consistent with the definition used by the EU Nitrogen Expert Panel⁵ and the approach used by Zhang et al.² NUE 63 is a simple indicator commonly used by researchers, policy makers and international 64 organizations to evaluate the relative transformation of N inputs into agricultural products.⁶⁻⁸ 65 Generally, for a given cropping system, low NUE over multiple years is an indicator of 66 significant N losses to the environment.⁶ NUE tends to decline with an increase of N input, and 67

reducing N input would increase NUE, but might not achieve the target yield. Therefore, combining NUE with other indicators such as N removed in harvested product and N surplus in a set of integrated indicators will be critical for evaluating the performance of the N management in cropping systems.⁵ In addition, estimations of changes in the soil N stock may be needed, because there is a risk that a high NUE is at the expense of soil N depletion, conversely, a low NUE may be the result of temporary soil N accumulation.^{5, 9, 10}

74 The N balance is a summary table which lists the main N inputs and outputs of a cropping system. The difference between N input and harvested N output is defined as the N surplus.^{11,} 75 ¹² N surplus and NUE are different indicators for evaluating the performance of N management, 76 77 but are related to each other. A benchmark for N surplus may be seen as a reference value for 78 the N surplus under optimum N management in a given cropping system. Calculating N surplus provides important information about the use efficiency of N inputs.^{11, 13, 14} The value of N 79 surplus can be positive, indicating a risk of N loss to the environment or negative, indicating 80 mining of soil N stock.¹¹ The N surplus is used as a management indicator by various countries 81 82 and organization. For example, the mineral accounting system (MINAS) in The Netherlands 83 was implemented at farm level in 1998 to achieve a step-wise decrease of the N (and phosphorus) surplus.¹⁵ Lowering the N surplus in agriculture in The Netherlands has made a 84 great contribution to improving the quality of groundwater and surface waters.¹⁵ The decline 85 86 of the nitrate concentrations in groundwater coincided well with the decline of the agricultural 87 N surplus in Denmark, which was attributed to the initiation of the Danish environmental action plan since 1985, of which the restrictions of maximum N application rates for specific crops 88 and minimum thresholds for utilization of N from animal manure are most important.¹⁶ 89

To improve N management in crop production, the EU Nitrogen Expert Panel⁵ proposed NUE as an easy-to-use indicator based on the N balance approach, but emphasized that NUE values need to be interpreted together with the quantity of N removed in harvested product (as a proxy for crop yield) and N surplus. They further suggested that these indicators are useful
tools for decision makers to compare N management between farms, cropping systems or
countries and to use this information to decide whether measures are needed to further improve
the N management of specific farms and/or regions.

97 In recent decades China has been successful in feeding 20% of the world's population with only 9% of the global cropland area.¹⁷ Grain yield has increased substantially, with 57% of this 98 increase being attributed to the use of synthetic fertilizer.¹⁸ The country is now consuming 99 around 30% of the global synthetic fertilizer,¹⁹ and had an overall N surplus of 27.6 ± 4.3 Tg 100 N for cropland in 2010,²⁰ which contributed to serious environmental pollution. More recently, 101 102 Chinese agricultural scientists have made great advances in finding technical solutions to 103 achieve relatively high crop yields with less environmental costs, under experimental conditions.²¹⁻²³ However, few studies have addressed the need for N management indicators at 104 farm level.²⁴ There are currently no clear guidelines to restrict the overuse or misuse of N in 105 106 farmers' practices, which is probably the main cause of severe N pollution from the agricultural sector over the last three decades.²⁵ In contrast, N management indicators in some Western 107 108 countries are well established and have served as useful tools for farmers and policy makers to achieve better N management.²⁶ 109

Over the last three decades a large numbers of studies have been carried out to establish economic optimum N fertilizer application rates based on yield response curves in experiments for widely differing cropping systems across China.^{21, 23} The results of these studies can be used now to establish N management indicators and N surplus benchmarks. The objectives of the present study are therefore: (1) to establish N surplus benchmarks for Chinese main cropping systems (wheat, maize, rice and rapeseed) at optimal N application rates; (2) to evaluate the N surplus and corresponding N harvest and NUE indicators under current farmers' practices; and (3) to propose effective ways for achieving the N surplus benchmarks throughimproved N management practices in crop production.

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120 MATERIALS AND METHODS

121 Main cropping systems and regions. Based on the "Integrated Regionalization and Planning of Agriculture"²⁷ and "Integrated Regionalization of Farming Systems",²⁸ we divided 122 Chinese cropland into 7 regions, each comprising several provinces, i.e. northeast (NE), 123 124 northwest (NW), North China Plain (NCP), middle and lower Yangtze River (MLYR), 125 southwest (SW), southeast (SE) and others. For 'others' there were no data available and so 126 these regions were not included in the present study, i.e., Tibet, Qinghai, Taiwan, Hong Kong 127 and Macao. The studied cropping systems of each region were identified according to the main Farming Systems in China²⁸ and data availability. In total, 4 crop types and 13 cropping 128 129 systems were selected (Figure 1). In the above 6 regions, grain production for the above 4 crops account for >95% of total grain production in China.²⁸ The proportions of the wheat, corn, rice 130 and rapeseed areas that were irrigated were 48%, 15%, 95% and 0, respectively.²⁹⁻³² The single 131 132 cropping systems (i.e. one crop per year) are mainly in NE and NW. In NCP, MLYR, SW and SE, double cropping systems are dominant with two harvests per year.³³ 133





Figure 1. The studied main cropping systems in different regions of China. The basemap wasdownload from Resource and Environment Data Cloud Platform (http://www.resdc.cn/).

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138 Calculation of N surplus. The main external N inputs to these cropping systems are fertilizer N (both synthetic fertilizer and manure) and N from atmospheric deposition and biological N₂ 139 fixation. Internal N transformations and minor N inputs (e.g., straw return, net soil organic 140 141 matter mineralization, irrigation and seed) were not considered. We assumed that all straw was returned to the soil in all cropping systems, due to the governmental ban on straw burning and 142 economic incentives to return straw since 2000.^{34, 35} We cannot exclude the possibility that 143 144 straw has been removed in some cases, but expect that our assumption here will not introduce 145 large bias in the N surplus estimation. Irrigation N input can be large in some cropping systems, 146 such as greenhouse vegetables in the North China Plain, but is small for the cropping systems of the present study.³⁶ Seed N is also very small and can be regarded as negligible compared 147 to N input from fertilization.³⁷⁻³⁹ 148

149 N output includes the N harvested in cereal grain. N surplus and NUE were calculated as:

 $N_{sur} = N_{fer} + N_{dep} + N_{fix} - N_{har}$ (1)

151
$$NUE = N_{har} / (N_{fer} + N_{dep} + N_{fix})$$
(2)

where N_{sur} and NUE are N surplus and N use efficiency, respectively; N_{fer} , N_{dep} , and N_{fix} represent the N input from fertilization, atmospheric deposition and non–symbiotic N fixation (all crops in the present study were non-leguminous crops), respectively; N_{har} is the N in harvested grain.

Approach for establishing N surplus benchmarks. Here we define the N surplus benchmark as 'the calculated N surplus value at economic optimum N management'. It has similar meanings in somehow with relevant references.^{2, 5, 40, 41}

159 Data and information on economic optimum N management for wheat, rice and maize were obtained from previous studies across different agro-ecological regions in China.^{31, 32, 42} On-160 161 farm experiments across the above regions (1575, 1177 and 1726 for wheat, rice and maize on 162 a national scale, respectively) were conducted between 2005 and 2010. All the field 163 experiments received the same treatments: no N fertilizer (N0), recommended N application 164 rate (RN), 50% RN, and 150% RN. None of these experiments had inputs of animal manure or 165 other organic N sources. The amount of N fertilizer for the RN treatment was determined by local agricultural extension employees. The RN was a starting point for setting the gradient of 166 167 N fertilizer rates (0, 50% RN, RN and 150% RN) for each field experiment, which allowed 168 reliable yield-N response curves to be established.

To determine the economic optimum N rates at the regional scale, a quadratic model (Table S1) was used to relate N input to grain yield in each on-farm experiment using SAS software (SAS Institute Inc., Cary, NC, USA). The quadratic model is typically used to describe N-yield response curves.⁴³ Next, the following variables were calculated at different N input levels: the yield increase (amount above the yield in the N0 treatment), gross return for the yield increase 174 (yield increase times grain price), cost of N fertilizer (N rate times fertilizer price), and the net return on N application (gross return minus fertilizer cost). Finally, the average net return for 175 each N increment across all N response curves was calculated. The N application rate with the 176 177 largest average net economic return from N fertilizer input was defined as economic optimum N rate in a region.^{31, 32, 42, 44} Although the fertilizer N price and market prices of rice, wheat and 178 179 maize showed variations in space and time, the fluctuation of these parameters was small compared to other countries We therefore used an average fertilizer N price of US\$0.59 kg⁻¹ 180 N, and mean grain prices of US\$0.27, 0.29, 0.32 kg⁻¹ for wheat, maize and rice, respectively 181 182 (see sensitivity analysis below). This approach is also known as 'Maximum return to N' (MRTN; Sawyer et al.⁴⁴), and used by Wu³¹ and Wu et al.^{32, 42} to calculate the economic 183 184 optimum N rates used in the current study. The relationships of economic optimum N rate (for 185 calculating N surplus benchmark), maximum yield N rate and the recommended N rate (RN) 186 was further illustrated in Figure S1.

As there were no data for rapeseed in the aforementioned studies, we obtained data on economic optimum N management for this crop from Ren et al.⁴⁵ who estimated the economic optimum N rates for rapeseed in the Yangtze River Basin between 2007 and 2009 using 60 onfarm experiments. Since there was no sub-region data in Ren et al.⁴⁵, fertilizer N and yield from Ren et al.⁴⁵ were used in both of the SW and MLYR regions in the present study.

For calculating the N surplus benchmarks, N input from deposition was obtained from 22 rural sites in the Nationwide Nitrogen Deposition Monitoring Network (NNDMN)⁴⁶. Each region in this study contains 2 to 6 monitoring sites in NNDMN. Regional N deposition rates were the average of measurements at all sites in each region. N input from biological N fixation was obtained from Bouwman et al.⁴⁷ N fixation rate associated with rice production was 25 kg N ha⁻¹ yr⁻¹, and 5 kg N ha⁻¹ yr⁻¹ for wheat, maize and rapeseed. Grain N harvest for each crop 198 was calculated by multiplying grain yield by grain N concentration: 1.9%, 2.3%, 1.4% and
199 3.9% of grain N content for rice, wheat, maize and rapeseed, respectively.⁴⁸

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Calculating reactive N (Nr) losses. Total Nr losses were related to N surplus but not
equivalent to N surplus; we examined the correlations between N surplus, NUE and Nr losses.
Reactive N (Nr) losses include NH₃ volatilization, N₂O emissions, nitrate leaching and runoff;
N₂ loss was not included since N₂ has no harmful impacts on the environment. Details about
calculation of the Nr losses can be found in SI and Table S2.

To compare N surplus and Nr losses under farmers' conventional N management with those under economic optimum N management, we collected data on fertilizer N rates and grain yields under farmer's conventional N management from the literatures (See SI for details data collection and calculation).

In the present study, we divided Chinese cropland into 7 regions, each comprising several provinces with the consideration of soil-climate similarity. There were differences of zoning method between the present study and literature data .^{31, 32, 42, 46, 49} Details on merging the regional data from literature to the present study can be found in SI.

Fertilizer N input, crop yield, N harvest and Nr losses of different crops in China under optimum and conventional N managements were summarized in Table S3. For the double cropping systems in each region, the items of N input and N output were calculated as the sum of each crop to convert the data to cropping systems.

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Sensitivity analyses. We did the sensitivity analysis to see how variation of fertilizer prices and product revenues affect the economic optimum N rates and the N surplus benchmarks, and predicted the changes of the crop yield, NUE, and Nr losses. As the crop price is quite stable in China due to the central government control⁵⁰ and the urea price range from 150 to 300 (fluctuation range: $\pm 33\%$) USD per ton during 2000-2010 in China,⁵¹ we therefore used a change of $\pm 40\%$ (only change of fertilizer price) for price ratio of fertilizer to crop to calculate the change of economic optimum N rates and corresponding crop yield based on the equation 3^{52} and the N-yield response curves in Table S2, respectively. N surplus benchmarks and NUE were calculated by using the above equation 1 and equation 2. Nr losses were calculated by using the regional N loss models of Cui et al.⁴⁹ (See SI and Table S2 for more details).

229 $X^*=X_{max} \times [1-2 \times R \times X_{max}/(Y_{max}-Y_0)]$ (3)

where X* is the economic optimum N rate; Y_{max} and X_{max} are maximum crop yield and corresponding N rate, respectively, which calculated from the N-yield response curves in Table S2; Y_0 is crop yield without N fertilizer application; R is the price ratio of fertilizer to crop

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234 **RESULTS**

N surplus benchmarks for main cropping systems. With N inputs of 192–455 kg N ha⁻¹ 235 yr⁻¹ and N harvests of 126–294 kg N ha⁻¹ yr⁻¹ under economic optimum N management, the N 236 surplus benchmarks range from 40 to 190 kg N ha⁻¹ yr⁻¹ (Table 1 and Table 2). In NE and NW 237 with single cropping systems, N surplus benchmarks are in the range of 40–100 kg N ha⁻¹ yr⁻¹, 238 239 with corresponding N input and N harvest values in the range of 192-224 and 126-154 kg N ha⁻¹ yr⁻¹, respectively. The N surplus benchmarks of double cropping systems are in the range 240 of 110–190 kg N ha⁻¹ yr⁻¹, with N inputs and N harvests in the range of 358–455 and 211–294 241 kg N ha⁻¹ yr⁻¹, respectively. The wheat-maize system in NCP and wheat-rice system in MLYR 242 are the two most intensive double cropping systems in China,²³ with relatively high N input, N 243 244 harvest and yield (Table 1 and Table 2). The N surplus benchmarks in the above two cropping systems have a similar value, 160 kg N ha⁻¹ yr⁻¹. The N surplus benchmarks for rapeseed-rice 245 systems in MLYR and rapeseed-maize in SW are higher than those for other cropping systems 246

247 and regions; these differences may be attributed to the low NUE of rapeseed, which contributes

248 to relatively large N surpluses⁵³ in the aforementioned two cropping systems.

- 249
- 250 Table 1. N surplus benchmarks for the main cropping systems of China as derived from
- 251 economic optimal N management^a

Designs	Constant	N input		N. h	N surpluses ^d	
Regions	Cropping systems	Fertilizer N ^b	Other N ^c	- N narvest		
Northeast	Rice	127	65	154	38	
Northeast	Maize	160	45	126	79	
NI	Wheat	166	40	138	68	
Northwest	Maize	184	40	128	96	
North China Plain	Wheat-Maize	361	71	270	162	
	Wheat-Rice	381	74	294	161	
Middle and lower Yangtze River	Rice-Rice	337	94	263	168	
	Rapeseed-Rice	377	74	264	187	
	Wheat-Maize	313	45	211	147	
C there at	Wheat-Rice	294	65	250	109	
Southwest	Rapeseed-Rice	343	65	252	156	
	Rapeseed-Maize	362	45	213	194	
Southeast	Rice-Rice	325	83	261	147	

^a the unit for all the numbers is kg N ha⁻¹ yr⁻¹

^b includes synthetic fertilizer and manure (but manure was not used in the experiments used here)

^c including N inputs from atmospheric deposition and biological N fixation

 d N surplus benchmark has been rounded to the nearest 10 kg ha⁻¹ yr⁻¹ for use in Table 2

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258 **Performance under economic optimum and conventional N management practices.** The

259 N input (including fertilizer N and other N) under conventional N management was in the range

of 206 to 532 kg N ha⁻¹ yr⁻¹, which is 7% to 27% higher than the N input under economic

261 optimum N management. However, crop yield under conventional N management was 2 to

262 30% lower than that under economic optimum N management. This was because N input under conventional N management far exceeded the N rate for the maximum yield (extreme N over-263 fertilization) which led to either lodging or increased susceptibility to diseases or pests.⁵⁴ (Table 264 2) The harvested N under conventional N management (range 104 to 259 kg N ha⁻¹ yr⁻¹) was 265 266 2% to 30% lower than that under economic optimum N management. The reason for higher N 267 input with relatively lower crop yield in conventional N management compared to economic optimum N management could be attributed to farmers' lack of knowledge and market 268 confusion as mentioned by Zhang et al.55 As a result, N surpluses under conventional N 269 management (range 59 to 349 kg N ha⁻¹ yr⁻¹) were 34% to 96% higher than the N surpluses 270 271 under economic optimal N management.

Expressing Nr losses on a yield-scaled basis (kg N (Mg of grain)⁻¹) provides an indication of Nr losses per ton of grain yield. Average yield-scaled Nr losses for conventional N management (7.4; range 2.4 to 13.0 kg N Mg⁻¹) were 42% higher than those at economic optimal N management (5.2; range 2.0 to 7.6 kg N Mg⁻¹). NUE at economic optimal and conventional N management were 52%–80% and 30%–71%, respectively. Average NUE of main cropping systems was 47% for conventional N management and 63% for economic optimum N management (Table 2).

279 Table 2 Nitrogen surpluses, NUE, and reactive N losses under economic optimum and farmer's conventional N management for different cropping

280 systems and regions

		Economic optimum N management					Farmer's conventional N management											
Regions	Cropping systems	N surplus bench- marks (kg N ha ⁻¹ yr ⁻ ¹)	NUE ª (%)	Yield (Mg ha ⁻¹ yr ⁻¹)	Nr losses ^b (kg N ha ⁻¹ yr ⁻¹)		Yield- scaled Nr	$ \begin{array}{c} N \text{ input} & N \\ \underline{(kg N ha^{-1} yr^{-1})} & harves \end{array} $		N harvest	N Surplus	NUE	Yield	Nr losses (kg N ha ⁻¹ yr ⁻¹)		Yield– — scaled Nr		
	- ,				NH3	N ₂ O	L&R °	losses ⁶ (kg N Mg ⁻ ¹)	Fertili- zer N	Other N	(kg N ha ⁻¹ yr ⁻ ¹)	(kg N ha ⁻ ¹ yr ⁻¹)	(%)	(Mg ha ⁻ ¹ yr ⁻¹)	NH3	N ₂ O	L&R	losses (kg N Mg ⁻¹)
N	Rice	40	80	8.13	16.5	0.5	5.3	2.7	141	65	147	59	71	7.75	17.9	0.5	5.6	3.1
Northeast	Maize	80	61	9.02	11.8	1.2	4.6	2.0	199	45	124	120	51	8.86	14.1	1.4	5.4	2.4
	Wheat	70	67	6.01	14.5	0.5	12.7	4.6	202	40	109	133	45	4.72	16.9	0.6	15.6	7.0
Northwest	Maize	100	57	9.16	13.2	1.3	5.1	2.1	238	40	104	174	37	7.42	16.3	1.6	6.3	3.3
North China Plain	Wheat-Maize	160	63	14.94	40.9	3.2	46.8	6.1	436	71	256	251	50	14.12	47.0	3.6	59.8	7.8
	Wheat-Rice	160	65	14.24	62.2	3.2	17.9	5.8	458	74	257	275	48	12.47	76.5	3.8	21.7	8.2
Middle and lower Yangtze River	Rice-Rice	170	61	13.85	66.4	2.6	19.7	6.4	390	94	259	225	54	13.60	77.0	2.8	21.9	7.5
8	Rapeseed-Ric e	190	59	11.06	50.8	3.2	30.4	7.6	442	74	214	302	41	9.25	63.3	3.6	33.7	10.9
	Wheat-Maize	150	59	12.25	31.3	2.7	29.0	5.1	395	45	165	275	38	9.33	37.9	3.3	41.7	8.9
	Wheat-Rice	110	70	12.24	49.1	1.8	13.0	5.2	345	65	224	186	55	10.96	57.2	2.1	14.5	6.7
Southwest	Rapeseed–Ric e	160	62	10.45	44.6	2.3	27.8	7.1	404	65	209	260	45	8.98	52.5	2.7	31.4	9.6
	Rapeseed–Mai ze	190	52	10.46	26.8	3.2	43.8	7.1	454	45	150	349	30	7.35	33.2	3.9	58.6	13.0
Southeast	Rice-Rice	150	64	13.76	65.2	1.0	16.7	6.0	434	83	257	260	50	13.53	83.7	1.5	19.9	7.8

281 a NUE=N harvest/N input×100%

^b Nr losses denote reactive N losses, Yield–scaled Nr losses=Nr losses/Yield

283 ^c L&R denote N leaching and runoff losses

284 Sensitivity analyses. The $\pm 40\%$ change of price ratio of fertilizer to crop gave a $\pm 2-23$ kg N ha⁻¹ yr⁻¹ ($\pm 2-6\%$) change of economic N rates and $\pm 2-22$ kg N ha⁻¹ yr⁻¹ ($\pm 6-15\%$) change of N 285 surplus benchmarks in all cropping systems (Figure 2a and 2b). It led to $\leq \pm 0.05$ Mg ha⁻¹ yr⁻¹ 286 ($\leq \pm 0.3\%$) change of crop yield, $\pm 1-3\%$ ($\pm 1-5\%$) change of NUE and $\pm 0-6$ kg N ha⁻¹ yr⁻¹ ($\pm 1-$ 287 6%) change of Nr losses (Figure 2c, 2d, 2e). The above analysis showed that the variation of 288 289 fertilizer prices and product revenues have little effect on the economic optimum N rates, N 290 surplus benchmarks, crop yield, NUE and Nr losses, given the low price ratio of fertilizer to 291 crop in China.



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Figure 2. Sensitivities of economic optimum N rates (a), N surplus benchmarks (b), crop yield
(c), NUE (d) and Nr losses (e) in response to ±40% change of price ratio of fertilizer to crop.
NE, NW, NCP, MLYR, SW, SE denote the agro–ecological regions of northeast, northwest,
North China Plain, middle and lower Yangtze River, southwest and southeast of China,
respectively. M, R, W, WM, WR, DR, RR, RM denote maize, rice, wheat, wheat–maize,
wheat–rice, double rice, rapeseed–rice and rapeseed–maize cropping systems, respectively.

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303 Assessment of N management. We plotted N input and N output (harvested N) of the 13 304 main cropping systems of China under both economic optimum and conventional N 305 management in Figure 3. The suggested minimum productivity level (N harvest=80 kg N ha⁻¹ yr⁻¹) and suggested ranges for NUE (50%–90%) according to the EU Nitrogen Expert Panel⁵ 306 307 are also shown in Figure 3. The N harvest of the main cropping systems under both economic 308 optimum and conventional N management were all above the minimum productivity level (80 kg N ha⁻¹ yr⁻¹) suggested by EU Nitrogen Expert Panel.⁵ Especially the N harvest values in 309 double cropping systems (range 150 to 294 kg N ha⁻¹ yr⁻¹) were much higher. Economic 310 optimum N management greatly reduced N surpluses (range 38 to 194 kg N ha⁻¹ yr⁻¹) compared 311 312 to conventional N management, and increased average (arithmetic mean value) NUE from 47% 313 for conventional management to 63% for economic optimal N management. The NUE of all 314 cropping systems under economic optimum N management was within the desirable range suggested by the EU Nitrogen Expert Panel,⁵ i.e., 50% to 90%, however, N surpluses of most 315 cropping systems were higher than the suggested mean (80 kg N ha⁻¹ yr⁻¹)⁵, except for single 316 317 rice and single maize in NE, and single wheat in NW.

318 The N inputs of some double cropping systems under conventional N management were 319 extremely high; the N input of the wheat-maize system in NCP, wheat-rice and rapeseed-rice in MLYR, rice-rice in SE all exceeded 500 kg N ha⁻¹ yr⁻¹. NUE values of single rice and single 320 321 maize in NE, rice-rice in MLYR, wheat-rice in SW under conventional N management were within the desirable range (50% to 90%), showing that high yield (high N harvest) were 322 323 obtained together with a desirable NUE level. However, the N surplus in these 4 cropping 324 systems was relatively high, which illustrates the importance of combining the above three 325 indictors for evaluating the overall performance of N management. In the other cropping 326 systems, high N harvests were associated with high N surpluses and low NUE; the N surplus

327 in 7 of the 9 double cropping systems under conventional N management exceeded 250 kg N



328 ha⁻¹ yr⁻¹, while NUE was \leq 50%.

Figure 3. Comparison of mean N input and output in harvested cereals under economic 330 331 optimum and conventional N management of main cropping systems and regions. N input 332 include fertilizer N, N deposition and biological N fixation; NE, NW, NCP, MLYR, SW, SE 333 denote northeast, northwest, North China Plain, middle and lower Yangtze River, southwest 334 and southeast regions; M, R, W, WM, WR, DR, RR, RM denote maize, rice, wheat, 335 wheat-maize, wheat-rice, double rice, rapeseed-rice and rapeseed-maize cropping systems. Open and solid circles denote data under economic optimum and conventional N management, 336 337 respectively (modified from EU Nitrogen Expert Panel⁵).

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339 **DISCUSSION**

Establishing N surplus benchmarks. In the present study, we established N surplus benchmarks for China's 13 main cropping systems. Benchmarks were derived at economic optimal N management. The cropping systems accounted for about 50% of total N fertilizer 343 consumption in Chinese agriculture in 2015.⁵⁶ Other important crops such as vegetables, fruits,
344 potatoes and cotton were not included due to lack of reliable experimental data.

345 For the soil surface N balance approach, N outputs (N removed by products) are usually easy 346 to obtain from yield and N concentration determinations. Nitrogen inputs may include many items, including fertilizer, manure, deposition, biological fixation, crop residue (straw), 347 348 irrigation, seed etc., and these data are not always easy to obtain accurately. There is no 349 accepted and universally applied protocol for establishing soil surface balances and the N input items considered differ therefore between studies. For instance, Norton et al.⁶ compared N 350 351 surplus and NUE of different countries, only considering synthetic fertilizer as N input. In the 352 crop section of the farm-gate N balance in MINAS, synthetic fertilizer and manure are taken into account, while deposition and biological fixation are accounted for as natural processes.⁵⁷ 353 354 Simplifications may make the N balance easier to calculate and use for policy orientation, but there is the risk of underestimating total N input.⁵⁷ At a global scale, Zhang et al.² calculated N 355 356 surplus and NUE for cropping systems in different regions/countries, using synthetic fertilizer, 357 manure, deposition and biological fixation as N input items.

The items of N input considered in the present study are the same as Zhang et al.² It is 358 359 important to include atmospheric N deposition because China (especially central-east China) is a global hotspot of N deposition with annual deposition rates of 23–71 kg N ha⁻¹ for rural 360 area.^{46, 58} Biological fixation is also an important N input item for some of the non-symbiotic 361 362 N fixation crops (e.g., rice in the present study). Although N fixation was relatively small for 363 maize, wheat, and rapeseed, we included N fixation in the N balance calculations of all crop 364 systems in order to maintain uniformity with rice-based crop systems. The seed N input was 365 neglected; it is lower than the associative N fixation rate. We also did not include animal 366 manure or other organic materials because these are mainly used in vegetables and fruits production, and seldom in maize, wheat, rice and rapeseed production.⁵⁹ 367

368 We assumed that straw was returned to the field, thus, straw N in the output was offset by 369 the input when calculated N surplus benchmark. Although the straw burning was banned and economic incentive for returning straw was provided by the Chinese government since 2000,³⁴, 370 ³⁵ it still has some cropping systems removing straw partly from the field in the practices. To 371 372 address these situations, we assumed that 1/3 of the straw was removed in all cropping systems, 373 and estimated how it affects N surplus benchmarks. The results showed the current N surplus benchmarks would be reduced 18-27 and 32-46 kg N ha⁻¹ yr⁻¹ for single and double cropping 374 systems, respectively (Table S4). Therefore, for the cropping systems in which only part of 375 straw was returned, the current N surplus benchmarks could be reduced around 20 kg N ha⁻¹ 376 377 per crop season.

By investigating the sensitivity of fertilizer price and product revenues, it showed that the economic optimum N rate and the N surplus benchmarks didn't depend strongly on them due to the low price ratio of fertilizer to crop in China.^{31, 32, 42} Thus, the change of fertilizer price (much lower compared with other countries in general)⁵¹ and the relative stable crop price⁵⁰ would cause little uncertainties of N surplus benchmarks.

383

384 **Comparison of N surplus benchmarks.** Losses of N are inevitable, especially in intensively managed crop systems, and the benchmark N surplus must reflect that. Our results show that 385 the N surplus benchmarks for China's main single cropping systems range from 40 to 100 kg 386 N ha⁻¹ yr⁻¹ (average 73 kg N ha⁻¹ yr⁻¹), which is close to the N surplus benchmark of the 387 Netherlands' MINAS (80 kg N ha⁻¹ yr⁻¹), and the N surplus benchmark of 80 kg N ha⁻¹ yr⁻¹ for 388 389 cropping systems proposed by the EU Nitrogen Expert Panel (Table 3). However, the N surplus benchmarks for Chinese main double cropping systems range from 110 to 190 kg N ha⁻¹ yr⁻¹ 390 (average 160 kg N ha⁻¹ yr⁻¹), which is roughly double that of single cropping systems (Table 391 3). There were large differences of N surplus benchmark in the 13 cropping systems, e.g. maize 392

of single cropping system in northeast China (80 kg N ha⁻¹ yr⁻¹) and the northwest China (100 kg N ha⁻¹ yr⁻¹). The main reasons for this relatively wide range are variations in soil-climate conditions and crop management. The high soil organic carbon content in northeast China compared to the northwest China³⁵ suggest that the soil N supply was larger in northeast than in northwest. There are also differences in annual average temperatures and rainfall between these regions,⁶⁰ which may have contributed to differences in Nr losses and N surplus.

In contrast to the top-down approach used by Zhang et al.², which derived N surplus 399 benchmarks for global and regional mean N surpluses by model simulations^{2,65}, we established 400 401 N surplus benchmarks for China's main cropping systems using on-farm field experiments: 402 this approach could be termed a bottom-up approach. The biggest difference between our study and the Zhang et al.² study is the data source. Our study used data from on-farm experiments 403 in specific cropping systems in specific regions, whereas the Zhang et al.² study used data from 404 405 the FAO and IFA statistical databases, and considered only one average Chinese cropping system. Zhang et al.² projected the total N surplus in China at 11 Tg N yr⁻¹ for the year 2050, 406 based on projected harvested N and target NUE. This translates to an N surplus benchmark of 407 65 kg N ha⁻¹ yr⁻¹, based on a harvested area of 170 million hectares in 2010 in China.⁶¹ Our 408 estimated average N surplus (77 kg N ha⁻¹ yr⁻¹) for main Chinese crops is close to the mean N 409 surplus target for 2050 suggested by Zhang et al.², in which they set higher NUE targets in all 410 411 crops. The economic optimum N rates between non-staple crops (e.g., fruits and vegetables) 412 and staple crops were similar. This suggests that the N surplus target for 2050 suggested by Zhang et al.² could be realized through economic optimal N input management. 413

We recognize that our N surplus benchmarks are still higher than the actual N surplus values already achieved in some western countries, which would suggest that our N surpluses could be further reduced in the future. Atmospheric deposition currently is an important N input, accounting for 8 to 21% of the total N input under economic optimum N management.⁴⁶ It is 418 likely that N deposition will reduce in the coming years due to the regulations and actions to 419 deliver cleaner air in China.⁶² Further, the Nr losses is still high even in the optimum N 420 management, and still have much space to be reduced when toward the "4R"s of nutrient 421 stewardship (using the right synthetic N fertilizer, at the right rate, right time, and in the right 422 place)⁶³. Hence, it is expected that N inputs may be reduced beyond economic optimal N inputs 423 and that N surpluses may be reduced further in future.

424

425 Table 3 Comparison of N surplus benchmarks between China and other countries/regions

Countries or Regions	N balance approach	N surplus benchmarks (kg ha ⁻¹ yr ⁻¹)	References	Notes
Netherlands	farm-gate	80	Hanegraaf and den Boer ⁶⁴	80 kg ha ⁻¹ yr ⁻¹ was the levy free N surplus for arable land in 2003.
Europe	n.a. ^{a)}	80	EU Nitrogen Expert Panel ⁵	Overal mean N surplus benchmark
World	soil surface	39	Zhang et al. ²	39 kg ha ⁻¹ yr ⁻¹ is the global mean N surplus benchmark in 2050
China	soil surface	65	Zhang et al. ²	Zhang et al. ² proposed a N surplus of 11 Tg yr ⁻¹ for China in 2050, which translates to about 65 kg ha ⁻¹ yr ⁻¹ .
China	soil surface	40-100	this study	N surplus benchmarks for single cropping systems
China	soil surface	110-190	this study	N surplus benchmarks for double cropping systems

^{426 &}lt;sup>a</sup> n.a. denotes not available

427

N surplus and Nr losses. The N surplus is a combination of the total N losses and changes of the soil N stock over time, but includes also possible errors associated with the determination of N inputs and N outputs. We expect that N surplus is a proxy indicator for N losses, and that changes in soil N stock over time were relatively small because of the long-term cultivation of farmland.⁹ However, the N surplus did not match with Nr losses in Table 2. For example, Northeast maize has an N surplus of 79 kg N ha⁻¹ yr⁻¹, while the Nr losses sum to only 18 kg N

ha⁻¹ yr⁻¹. There are three possible reasons for the difference between N surpluses and Nr losses. 434 435 Firstly, the assumption of returning straw may lead to a slight overestimation of N surplus as 436 discussed before. Secondly, we calculated only the Nr losses which have harmful 437 environmental effects, so denitrification loss to N2 was not accounted for in the N losses. 438 Finally, the region evaluation of Nr losses by model simulation was more uncertain than the 439 evaluation by site specific measurement. Nevertheless, the evaluation of Nr losses in the 440 present study gave an integrated impression for different pathways of Nr losses, especially for 441 the comparison of Nr losses between optimum and conventional N management.

442 Towards sustainable N management. To realize the proposed N surplus benchmarks for main cropping systems, N inputs (mainly synthetic N fertilizer) need to be reduced while 443 maintaining or improving current crop yields (N harvest).²³ Reducing synthetic N fertilizer 444 445 inputs must be done with complementary measures to minimize N losses. For instance, if we 446 optimize the N application rate according to crop demand while applying the fertilizer with 447 farmers' common practices (e.g. broadcast fertilizer with high N losses), large quantities of the 448 applied fertilizer N will be lost to the environment, and the rest of the applied fertilizer N will 449 not be enough to meet the crops' N demand (low harvested N), and N surplus will still be high.⁶⁶ The improvements could be expressed as full adoption of the "4R"s of nutrient 450 stewardship⁶³. and also include a better utilization of N from animal manures. By improving 451 452 fertilization techniques and agronomic managements to realize the N surplus benchmarks, the 453 target yields will be attained and the Nr losses will be minimized. We think that our N surplus 454 benchmark will be a valuable tool for policy makers and agricultural extension workers to evaluate and improve the N management of current farmers' practices, and to lay the 455 456 foundations for achieving the long-term goals for sustainable N management in China.

457

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464 Supporting Information Available

This file includes calculating N surplus under farmers' conventional N management, calculating reactive N (Nr) losses, merging the regional data from literatures to the present study, and Figure S1 and Table S1-S4.

468 This information is available free of charge via the Internet at http://pubs.acs.org.

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