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2022. **Pollution controls in Lake Tai with the reduction of the
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1 **Pollution controls in Lake Tai with the reduction of the watershed nitrogen footprint**

3 **Abstract**

4 To feed an increasingly affluent population, the nitrogen input into global food production
5 systems is currently two times the safe planetary boundary leading to adverse impacts on the
6 local and global environments. The nitrogen footprint is an important index to understand the
7 impact of human activities on the environment, however, it is rarely applied at a watershed scale.
8 By using the Coupled Human And Natural System (CHANS) model, it was found that the total
9 nitrogen input to the Lake Tai watershed has increased from 141.1 Gg N yr⁻¹ in 1995 to 244.3
10 Gg N yr⁻¹ in 2010, and then decreased again to 201.2 Gg N yr⁻¹ in 2015. The study findings are
11 consistent with the change in the total nitrogen concentration observed in Lake Tai. While the
12 per capita nitrogen footprint remained stable at around 30 kg N yr⁻¹ before 2010, a substantial
13 decrease to approximately 25 and 20 kg N yr⁻¹ in 2010 and 2015, respectively, has occurred.
14 Dominant sources of nitrogen emissions contributing to the nitrogen footprint gradually have
15 changed from agricultural production to wastewater and nitrogen oxides emissions from fossil
16 fuel combustion. A reduction of the nitrogen footprint is beneficial for effective nitrogen
17 pollution control strategies and better wastewater treatment techniques should be prioritized for
18 future policymaking.

21 **Key words:** Water pollution, Nitrogen footprint, Agriculture, Wastewater, Land use, Watershed,
22 Lake Tai

24 **1. Introduction**

25 China is the most populous country in the world, and in recent years, rapid economic
26 development also has changed the dietary structure to more animal-product based nutrition,
27 leading to substantial pressures on food security (Zhai et al., 2014). To produce sufficient food
28 and animal feed, China uses about 30% of the global nitrogen fertilizers, and raises about 30-
29 40% of the global livestock (FAO, 2020). However, due to the small farm size, nitrogen is not
30 well managed on cropland, and two thirds of animal manure nitrogen also is lost to the
31 environment due to decoupling of livestock and cropland (Wu et al., 2018; Zhang et al., 2019;
32 Zhang and Hu, 2020). These factors have contributed to a rapid increase of nitrogen pollution
33 across China, resulting in water and air pollution, with consequences for biodiversity loss due
34 to nitrogen deposition (Zhang et al., 2015).

35 Besides the nitrogen losses during food production, nitrogen losses as a result of food

36 consumption, i.e., via domestic wastewater, is another important emission source. With the
37 increase of population and per capita consumption, wastewater including effluents of
38 wastewater treatment plants (WWTP) (Yu et al., 2019) has become a dominant source of water
39 pollution in many watersheds. In rural China, however, wastewater is rarely treated at all and
40 sludge from septic tanks is not reused as manure (MHURDPRC, 2005; Deng and Wheatley,
41 2016). This wastewater makes a significant contribution to the increase of nitrogen pollution.
42 Nitrogen oxide (NO_x) emissions from fossil fuel combustion are another important emission
43 source of nitrogen (Zhang et al., 2007). The increase in energy demand has led to a rapid growth
44 of fossil fuel usage in China. NO_x emissions have become increasingly important as a driver of
45 atmospheric nitrogen deposition and contribute to biodiversity loss in sensitive regions and also
46 nitrogen input to lakes in urbanized regions (Bobbink et al., 2010; Hobbs et al., 2016). This
47 atmospheric pathway can add up to 30% of the total nitrogen input to lakes, substantially
48 affecting water quality (Ti et al., 2018). Understanding the contribution of nitrogen losses
49 during wastewater discharge after food consumption and energy production has, thus, become
50 increasingly important for nitrogen management.

51 The nitrogen footprint is defined as the total amount of nitrogen released to the
52 environment as a result of individual or collective activities (Leach et al., 2012). It is used to
53 calculate the loss of reactive nitrogen (Nr; all species of nitrogen except N₂) from human
54 activities to the environment, providing information on how to reduce Nr losses (Ti et al., 2018).
55 Previous studies have calculated the national nitrogen footprint, e.g., for Australia (Liang et al.,
56 2016), China (Gu et al., 2013), and the United Kingdom (Stevens et al., 2014). In addition,
57 nitrogen footprints also can be used by institutions to improve their sustainability in nitrogen
58 consumption (Leach et al., 2013). The success of the universities of Virginia (Leach et al., 2013)
59 and Australia (Liang et al., 2018) shows that the nitrogen footprint can be used not only for
60 nitrogen evaluation at the national scale, but also for nitrogen analysis at the local scale. A new
61 study has applied a nitrogen footprint calculation approach at the city scale to understand how
62 to manage nitrogen regionally (Huang et al., 2019). However, nitrogen pollution is typically
63 constrained by natural boundaries such as watersheds, not administrative boundaries. Linking
64 administrative boundaries and natural boundaries on nitrogen management using a nitrogen
65 footprint is important.

66 In this paper, a new watershed-scale nitrogen footprint is developed taking into
67 consideration changes in land use and water quality. By using the Coupled Human And Natural
68 System (CHANS) model (Gu et al., 2015), which is a conceptual model that identifies the main
69 nitrogen fluxes in a given physical system, this study quantifies the contribution of different
70 sources to the nitrogen footprint for the Lake Tai watershed. This model has advantages in mass

71 balance calculation, which is beneficial to nitrogen management. Previous studies have proved
72 that it is feasible to calculate the nitrogen footprint through the CHANS model (Gu et al., 2015).
73 Meanwhile, the spatial variation of different sources contributing to the nitrogen footprint also
74 were estimated based on the land use changes to identify hotspots of nitrogen use and losses
75 within the watershed. The relationship between the overall nitrogen footprint and water quality
76 then is analyzed in particular to reveal the driving forces of human activities on the aquatic
77 environment. Finally, aspects relevant to policy implementation emerging from this study are
78 discussed to identify better management options for nitrogen to contribute to the attainment of
79 the sustainable development goals.

80

81 **2. Materials and Methods**

82 **2.1 Study area**

83 The Lake Tai watershed located in East China (30°05'-32°08'N, 119°08'-121°55'E) was
84 used as a case study. The lake has an area of 2,427 km², and the whole watershed area is about
85 36,900 km² (see Fig. 3 for more geographical information). The Lake Tai watershed is one of
86 the most developed regions in China, with a high population density and well-developed
87 industrial and agricultural production. The watershed includes part of Jiangsu, Zhejiang, and
88 Anhui provinces and Shanghai city. In 2015, the total population of the Lake Tai watershed was
89 about 60 million, accounting for 4.4% of the total population of China. The total gross domestic
90 product (GDP) is US\$ 945.9 billion, accounting for 9.9% of the national GDP. Since the 1990s,
91 water pollution events have frequently occurred in Lake Tai. One particular event in 2007 was
92 marked by a large scale cyanobacteria outbreak, which seriously affected drinking water quality
93 and aquaculture production.

94

95 **2.2 Land use analysis**

96 Land use maps for the Lake Tai watershed for the years 1995, 2000, 2005, and 2010 with
97 a resolution of 1 km were derived from Landsat TM/ETM, and for 2015, from Landsat 8
98 remote-sensing data. All data were provided by the Data Center for Resources and
99 Environmental Sciences, Chinese Academy of Sciences (RESDC) (<http://www.resdc.cn>). The
100 land use types were divided into cropland, forest, grassland, water, and residential and
101 commercial land to analyze the change of land use types across the Lake Tai watershed over
102 the period 1995 to 2015. In addition, the nitrogen footprint calculations were associated with
103 land use types to explore the change of the per capita nitrogen footprint during the study period.

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105 **2.3 Nitrogen flow analysis.**

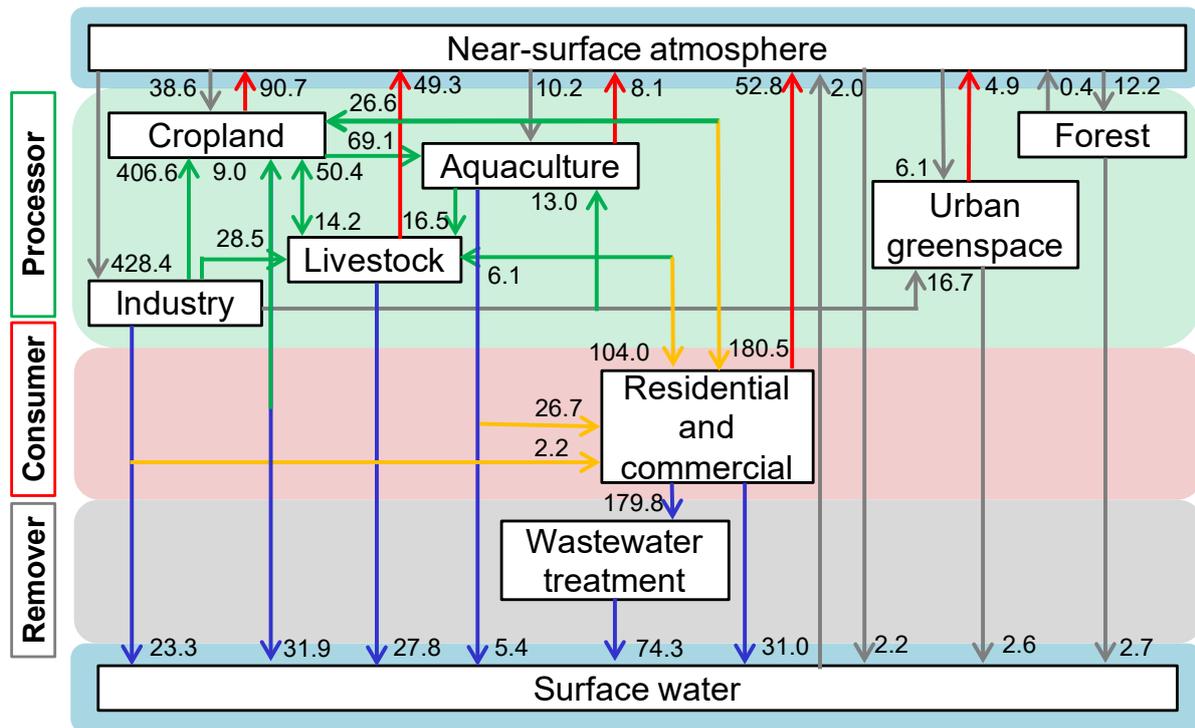
106 In order to quantify the nitrogen flows, the CHANS model was used to extract related
107 flows for the Lake Tai watershed (Fig. 1). The system boundary follows the watershed boundary
108 (Fig. 3), which includes the main channel and tributaries. Based on the relation between Nr
109 flows, the CHANS model is divided into four functional groups, which is further divided into
110 14 subsystems (Gu et al., 2015). The concept of subsystem and ecosystem is similar. Taking the
111 cropland subsystem as an example, it refers to biological (including human) activities and
112 natural activities on cropland. The nitrogen cycle for the Lake Tai watershed is primarily based
113 on the surface water subsystem. There are nine subsystems which directly interact with the
114 surface water subsystem through nitrogen flows: cropland, forest, livestock, aquaculture, urban
115 greenspace, industry, residential and commercial, wastewater treatment, and near-surface
116 atmosphere.

117

118 **2.4 Mass balance calculation**

119 Land use change and nitrogen flow analysis are related. The China Land Use/Land Cover
120 Remote Sensing Monitoring Database provides Chinese land use change data at intervals for
121 five years. The basic data for the Lake Tai watershed were collected for 5-year intervals for
122 1995, 2000, 2005, 2010, and 2015, and these were processed through the CHANS model to
123 calculate the local nitrogen flows (Fig. 1). Data used in this study can be divided into two
124 categories: (1) socioeconomic information for the Lake Tai watershed, such as cropland area,
125 urban/rural population, crop/livestock production, fertilizer usage, and sewage discharge, which
126 were mainly obtained from the annual statistics provided by yearbooks and bulletins, more
127 details can be found in the SI Appendix of Gu et al. (2015); and (2) coefficients and parameters
128 used for the calculation of nitrogen input to the surface water subsystem were adopted from the
129 CHANS model (Gu et al., 2015) with some modification for local parameters such as nitrogen
130 deposition (Liu et al., 2013) for each year.

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Fig. 1 Nitrogen cycle in the CHANS model used to calculate nitrogen fluxes. Data represent the N fluxes in 2015 with units of Gg N. The background colors represent different functional groups: blue represents life-supporter; green represents processor; red represents consumer; and gray represents remover. Arrows represent nitrogen (N) flows, which are all included in the calculation of the N cycle in this study. Blue arrows represent Nr discharge to the surface water; green arrows represent Nr flows associated with agricultural production; red arrows represent Nr emission to the atmosphere; and orange arrows represent Nr flows associated with human consumption and gray arrows mainly represent natural nitrogen processes. The two-way arrow indicates that the nitrogen flow is two-way. Taking the two arrows pointing to cropland and residential and commercial subsystems for example, the green arrow represents 27.2 Gg N from residential and commercial subsystem is input to cropland for agricultural production, and the orange arrow represents 184.3 Gg N from cropland subsystem is used by human residential and commercial activities. The natural nitrogen processes related to the nitrogen flow from industry to urban greenspace subsystems represent fertilization in urban greenspace.

The basic principle of the CHANS model is a mass balance approach, which is applicable to the whole system and subsystems (Gu et al., 2015):

150

$$\sum_{h=1}^m IN_h$$

$$= \sum_{g=1}^n OUT_g$$

$$+ \sum_{k=1}^p ACC_k \quad (1)$$

153 where IN_h and OUT_g represent the different nitrogen inputs and outputs, respectively, and ACC_k
 154 represents the different nitrogen accumulations. In the current study, this principle postulates
 155 that the nitrogen input from other subsystems to the surface water subsystem is equal to the sum
 156 of nitrogen output from the surface water subsystem to other subsystems and the amount of
 157 nitrogen accumulated by the surface water subsystem itself.

158

159 **2.5 Nitrogen footprint calculation**

160 The nitrogen footprint is defined as the total amount of nitrogen released to the
 161 environment as a result of individual or collective activities, expressed in total units of Nr (unit:
 162 kg N capita⁻¹ year⁻¹), which provides a novel method to link consumption behavior and
 163 management behavior to the nitrogen cycle (Leach et al., 2012). Based on the mass balance
 164 approach embedded in the CHANS model (eq.1), the data of nitrogen flows was calculated, and
 165 the data related to the system level nitrogen footprint (NF_{system}) of the Lake Tai watershed was
 166 extracted to calculate the footprint for a certain year as follows (Gu et al., 2013):

$$167 \quad NF_{system} = N_{BNF} + N_{CFA} + N_{FFC} + N_{FI} \quad (2)$$

168 where N_{BNF} indicates nitrogen from biological nitrogen fixation, N_{CFA} indicates chemical
 169 fertilizer application, N_{FFC} indicates NO_x emission from fossil fuel combustion, and N_{FI}
 170 indicates feed and food import from other regions (estimated based on the differences between
 171 consumption and production). In the calculation of the nitrogen footprint in the Lake Tai
 172 watershed, N_{BNF} mainly includes cropland BNF, aquaculture BNF, grassland BNF, forest BNF,
 173 and urban greenspace BNF.

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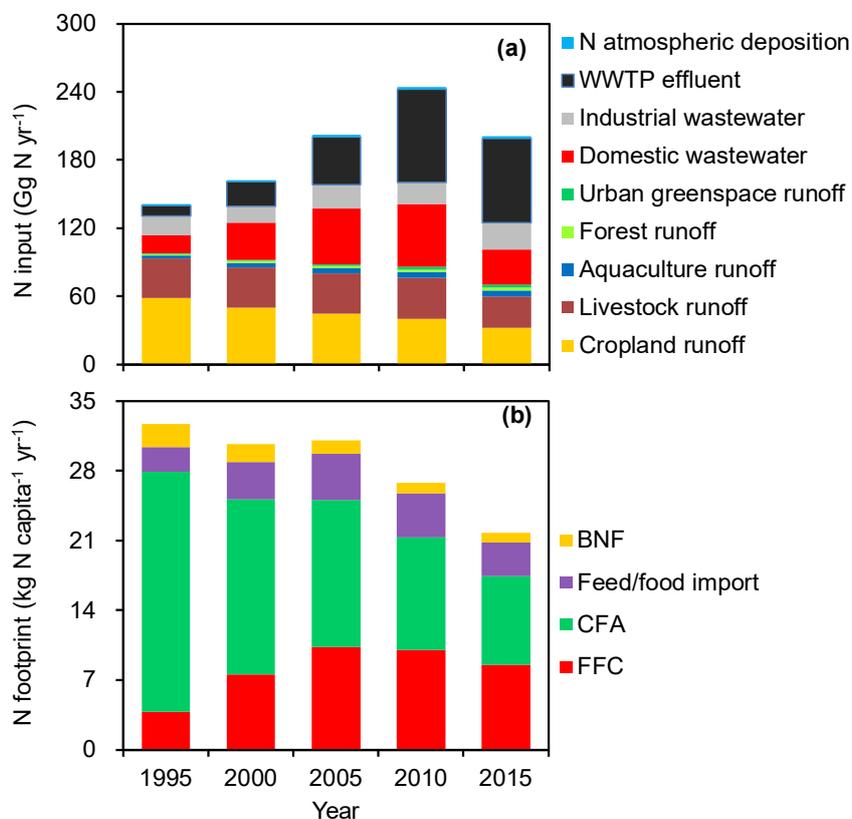
175 **3. Results**

176 **3.1 Nitrogen input and nitrogen footprint for the Lake Tai watershed**

177 The nitrogen input to the surface water subsystem Lake Tai watershed (including main
 178 channel and tributaries) increased by over 70% from 141.1 Gg N yr⁻¹ in 1995 to 244.3 Gg N yr⁻¹
 179 in 2010, and then declined to 201.2 Gg N yr⁻¹ in 2015 (Fig. 2a). Cropland runoff was the
 180 largest component of nitrogen input in 1995, while wastewater treatment plant (WWTP)

181 effluent has replaced cropland runoff as the largest source since about 2010, which was
 182 consistent with the changes in the socio-economic structure, industrial development, and
 183 population growth in the watershed. Cropland runoff decreased from 58.4 to 31.9 Gg N yr⁻¹
 184 between 1995 and 2015, mainly due to a reduction in fertilizer use. The Lake Tai watershed
 185 has attracted a large number of immigrants because of its well-developed economy and
 186 convenient transportation, leading to substantial population growth. These changes increased
 187 the amount of WWTP effluent, challenging the nitrogen removal rate of the WWTPs in the
 188 future.

189 A system-level nitrogen footprint for the Lake Tai watershed was estimated, with the per-
 190 capita footprint remaining relatively stable at around 30 kg N yr⁻¹ before 2010, and then
 191 reducing markedly to approximately 25 and 20 kg N yr⁻¹ in 2010 and 2015, respectively (Fig.
 192 2b). This change mainly occurs due to the decrease of N input and increase of population that
 193 results in a dilution effect. Chemical fertilizer application has decreased by 63.9% during the
 194 period from 1995 to 2015 because of the changes in agricultural production. The decrease of
 195 cropland area and fertilizer use per hectare led to an increased nitrogen use efficiency (NUE,
 196 harvested crop Nr divided by total Nr input), from 29% to 35% between 1995 and 2015, which
 197 reduced overall nitrogen fertilizer use. Meanwhile, an increase of animal feed and food import
 198 transferred part of the nitrogen footprint outside of the region (Fig. 2b).



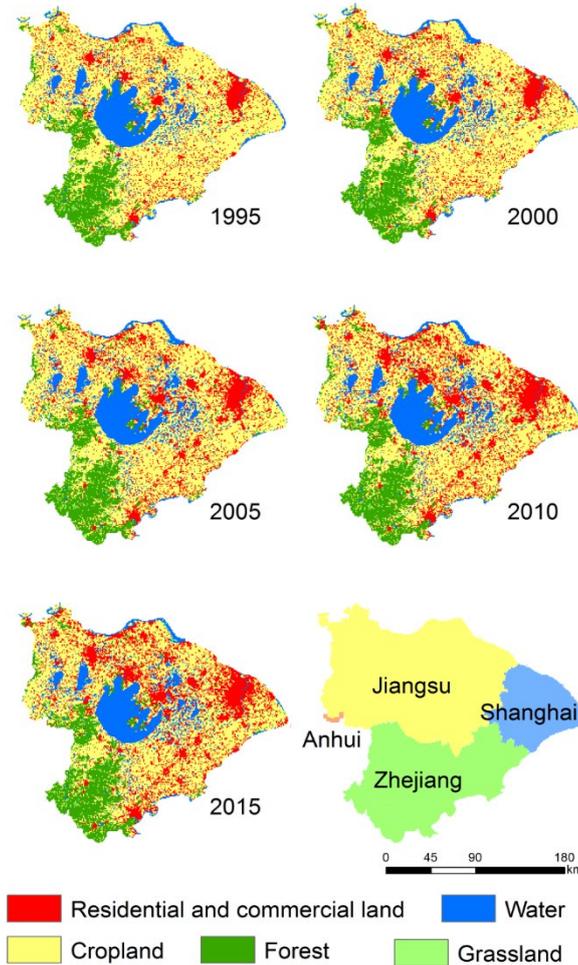
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200 **Fig. 2 Changes of nitrogen input and nitrogen footprint from 1995 to 2015.** (a) Nitrogen
201 input. (b) Nitrogen footprint. WWTP refers to wastewater treatment plant; BNF refers to
202 biological nitrogen fixation; CFA refers to chemical fertilizer application; FFC refers to NO_x
203 emission during fossil fuel combustion.

204

205 **3.2 Land use change and socioeconomic development**

206 The land-use types in the Lake Tai watershed were divided into five basic categories:
207 cropland, forest, grassland, water, and residential and commercial land to analyze of land use
208 changes from 1995 to 2015 (Fig. 3). Cropland, forest and grassland only consider nitrogen input
209 from their own subsystem in the CHANS model; water includes the aquacultural and surface
210 water subsystems; and residential and commercial land include nitrogen input from industry,
211 urban greenland, pet, livestock (animals raising normally used built-up area which is classified
212 in residential and commercial land), and wastewater treatment subsystems. Forest is mainly
213 distributed in the southwest of the Lake Tai watershed (within Zhejiang province), accounting
214 for 13% of the total watershed area. Water surface is mainly from Lake Tai, surrounded by some
215 smaller lakes and rivers (14%). Grassland area is relatively small (0.4%) and scattered around
216 other land use types. In the past 20 years, the area and distribution of forest, water, and grassland
217 have not changed significantly. On the other hand, cropland and residential and commercial
218 land have changed substantially. Between 1995 and 2015, about 16 percentage points of
219 cropland were converted into residential and commercial land. Residential and commercial land
220 was mainly distributed in the northeast of the watershed (within Shanghai city) in 1995, and
221 there were also small-scale residential and commercial land clusters in Jiangsu, Zhejiang, and
222 Anhui provinces. By 2015, not only had the area of the original residential and commercial
223 area become larger, but also many new residential and commercial areas had developed.



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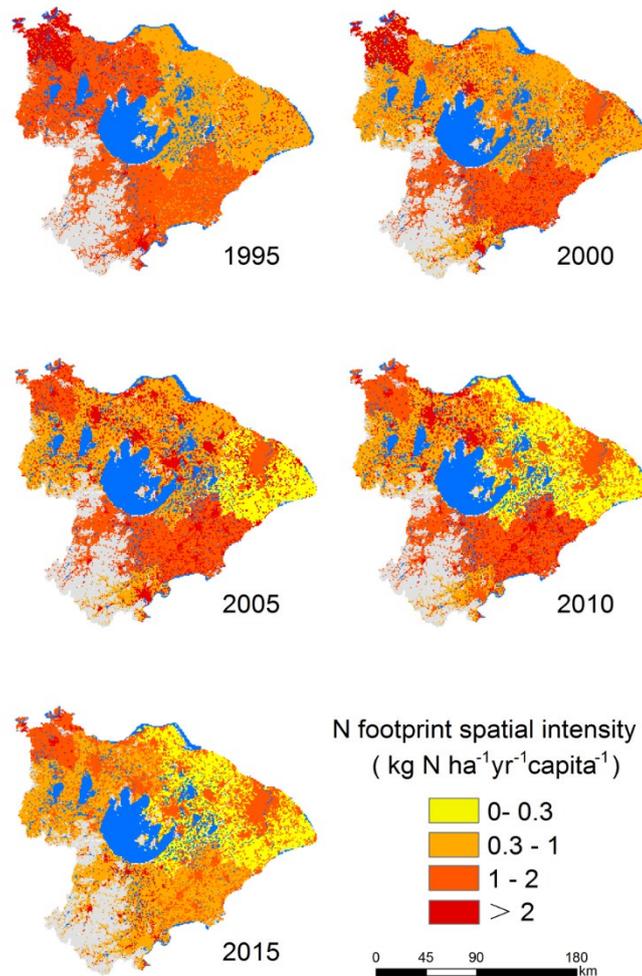
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229 3.3 Nitrogen footprint spatial intensity

230 In order to analyze the nitrogen footprint spatial intensity (NFSI), each source of the
 231 nitrogen footprint was assigned to different land use types (for example, the cropland BNF
 232 occurred on cropland, NO_x emission during fossil fuel combustion occurred on residential and
 233 commercial land), and the annual per capita nitrogen footprint per hectare of land was computed
 234 (Fig. 4). Because the nitrogen footprint of forest, grassland, and water is relatively small and
 235 has not changed much, Fig. 4 focuses on the analysis of the nitrogen footprint for cropland and
 236 residential and commercial land only.

237 As a whole, the per capita NFSI of cropland has gradually declined from 1995 to 2015,
 238 starting from between 0.3 and 2 kg N ha⁻¹ yr⁻¹ capita⁻¹ in 1995, with a decreasing gradient from
 239 west to east and the highest values in the northwest. By 2015, the NFSI of cropland was 0-0.3

240 kg N ha⁻¹ yr⁻¹ capita⁻¹ in the northeast of the watershed, 1-2 kg N ha⁻¹ yr⁻¹ capita⁻¹ in the
241 northwest, and 0.3-1 kg N ha⁻¹ yr⁻¹ capita⁻¹ in the remaining areas. In contrast, the per capita
242 NFSI of residential and commercial land showed an upward trend. With the continuous
243 expansion of the residential and commercial land areas, most have an NFSI higher than 1-2 kg
244 N ha⁻¹ yr⁻¹ capita⁻¹, which indicates that those areas have become nitrogen footprint hotspots
245 over the period from 1995 to 2015.



247 **Fig. 4 Spatial distribution of per capita nitrogen (N) footprint from 1995 to 2015.** Yellow
248 represents low nitrogen footprint regions, red represents high nitrogen footprint regions. The
249 more substantial the color change, the greater the change in the nitrogen footprint. Blue refers
250 to the water, and gray refers to forest and grassland, and their nitrogen footprints are small, and
251 the changes are slight, which are not identified in the figure. The base map is derived from
252 GADM data (<https://gadm.org/>).

253

254 4. Discussion

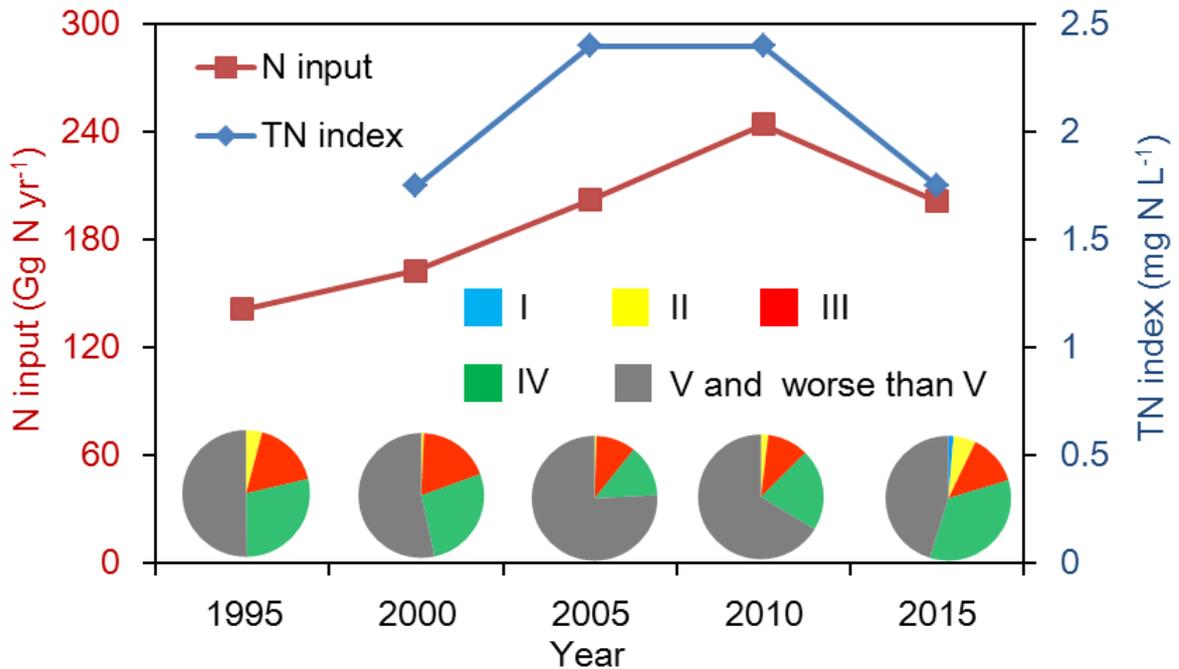
255 Generally, a substantial transition in the main flows of nitrogen use and loss within the
256 Lake Tai watershed was found over the study period. The improvement of Nr management for
257 cropland and the import of feed and food substantially reduced the nitrogen footprint of food
258 production, despite the population increasing by 20 million, for a growth of 53% compared with
259 the 1995 population levels. This illustrates that environmental improvements can be realized
260 despite increased population densities and food demand through better agricultural
261 management. The farm size in many regions within the Lake Tai watershed increased with the
262 implementation of the land transfer system (Ju et al., 2016). However, part of the region's
263 footprint has been transferred to other regions, with the food and feed imports. Information on
264 the origins of these imports currently is not available, which limits the ability to do a
265 comprehensive estimation of the Nr footprint change due to trade.

266 With the reduction in the relative share of agricultural land, more land areas were
267 converted to urban residential and commercial areas. As a result, a shift of increasing the
268 nitrogen footprint of urban land use is found, mainly corresponding to Nr emissions from food
269 and fossil fuel consumption. This is not only indicated by the total Nr losses to water bodies,
270 but also by the detailed analysis of the spatial distribution of Nr losses from different land use
271 types. Areas with urban land use have become the new hotspots compared to emissions from
272 cropland and natural land. This highlights the growing importance of an efficient management
273 of Nr for urban land use.

274 In the current study, a watershed nitrogen footprint based on the consumer side was
275 initiated. To directly link the nitrogen input and water pollution, the total nitrogen (TN) index
276 was used to analyze the nitrogen concentrations in water. It was found that the TN index and
277 Nr input in the watershed had the same trend over a 15-year period (Fig. 5): increasing from
278 2000 to 2010 and subsequently decreasing after 2010. The increasing demand for daily life
279 needs and industrial production has made the nitrogen input in the Lake Tai watershed increase
280 since 1995. This increase made the Chinese government realize the urgency and necessity of
281 controlling water pollution in the Lake Tai watershed. Since then, the government has invested
282 around US\$ 50 billion to reduce pollutant (e.g., nitrogen and phosphorus) input into the
283 watershed. The decline of nitrogen pollution from 2010 to 2015 directly reflects the
284 government's effectiveness in controlling nitrogen losses.

285 In addition, the water quality of Lake Tai also was mapped, and it followed a similar
286 trend: from 1995 to 2010, the proportion of water quality of Grade IV, Grade V, and worse than
287 Grade V continued to increase and by 2005 the worst category could be attributed to 75% of
288 the watershed. Starting around 2010, water quality gradually improved with the proportion of
289 Grade IV, Grade V, and worse than Grade V reducing, and a water quality level of Grade I being

290 observed for the first time (Fig. 5). In the current study, it is found that despite the timeline of
 291 nitrogen input not being completely consistent with respect to the TN index and water quality
 292 in the Lake Tai watershed, their overall patterns are similar on a two-decade scale. Therefore,
 293 these can be used to quantify the impact of different human activities, providing detailed data
 294 to support policy-makers in identifying suitable measures to control Nr pollution.



296 **Fig. 5 Temporal trend of the nitrogen input and water quality from 1995 to 2015.** The total
 297 nitrogen (TN) index in Fig. 5 represents the level of nitrogen concentration as an indicator of
 298 water quality, which includes nitrate nitrogen, ammonia nitrogen, organic nitrogen, etc.
 299 According to the Chinese classification of surface water environmental quality standard
 300 GB3838-2002, the middle value of the TN index range is taken as the value for the Lake Tai
 301 watershed (for example, the TN index in 2000 was class V, the range of class V is 1.5-2.0 mg
 302 N L⁻¹, and the middle value is taken as 1.75 mg L⁻¹ in the figure). The TN index in 2000 and
 303 2005 was from the literature (Qian and He, 2009), and the TN index in 2010 and 2015 was
 304 obtained from the Lake Tai health status report published by the Lake Tai Basin Authority of
 305 the Ministry of Water Resources (the TN index in 1995 was not available). The pie chart in Fig.
 306 5 shows the change of water quality in Lake Tai watershed from 1995 to 2015. The colors of
 307 the pie charts represent different water quality grades: blue, yellow, red, green, and gray
 308 represent Grades I, II, III, IV, and V and worse than V, respectively. According to GB3838-2002,
 309 Grades I-III are suitable for water supply, Grade IV waters are suitable for industrial water use
 310 and non-contact recreation areas, and Grade V waters are only suitable for agricultural water
 311 use and landscaping requirements, furthermore, the water worse than Grade V is not suitable

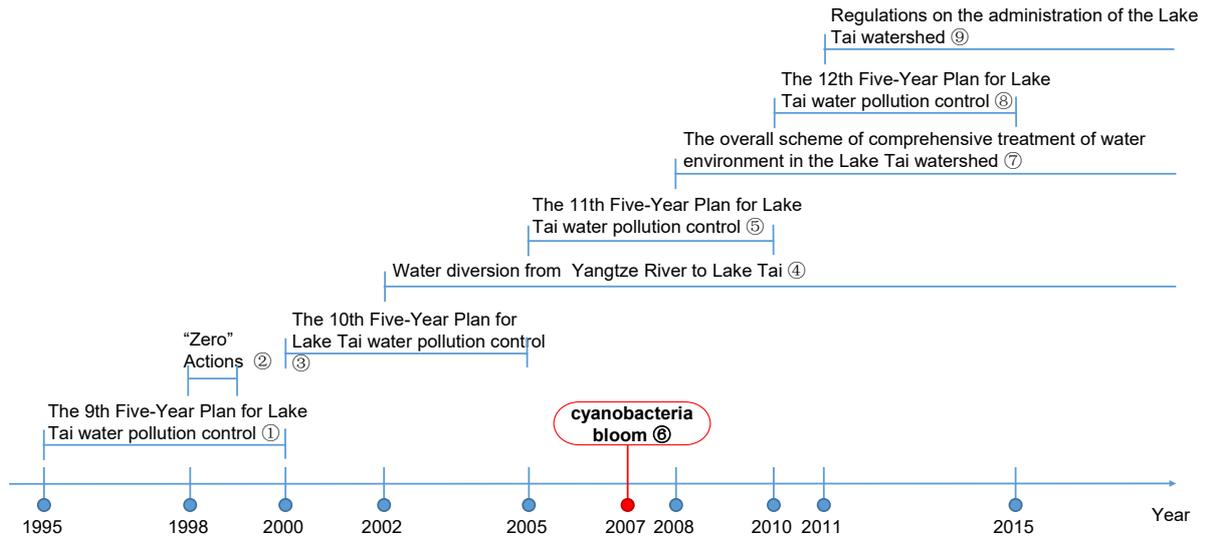
312 for any use. The data in 1995 and 2000 are from the literature (Jin et al., 2006), and the data of
313 2005, 2010, and 2015 were from the Lake Tai Watershed & Southeast Rivers Water Resources
314 Bulletin.

315

316 The Chinese government has paid close attention to nitrogen pollution in the Lake Tai
317 watershed, with substantial policies for the treatment of Nr pollution in the basin since 1995
318 (Fig. 6). These policies not only include the five-year plan, but also additional management in
319 combination with the actual situation of the Lake Tai watershed. Despite these actions, a large
320 cyanobacteria bloom occurred in 2007, causing irreparable damage to the livelihood of local
321 residents and biodiversity in the watershed, which reflects certain shortcomings in the
322 governance measures. Taking “zero o’clock” action in 1998 as an example (Fig. 6), although
323 it controlled the sewage discharge of industrial enterprises in the watershed, this action lasted
324 for a very short time, which was not conducive to the sustainable development of the Lake Tai
325 watershed.

326 In the aftermath of the 2007 cyanobacteria bloom, the government not only increased the
327 attention in environmental protection for the Lake Tai watershed, but also adjusted the focus of
328 the policy (Fig. 6). These policies not only require attention to agricultural non-point source
329 pollution, but also require the control of urban sewage discharge, taking the treatment of point
330 source pollution and non-point source pollution into account. During the past two decades, over
331 US\$ 50 billion has been invested by governments to control the water pollution in Lake Tai.
332 Although the pollution is reduced with the decline in the nitrogen footprint, still more effort is
333 needed to further reduce the Nr loading and other elements such as phosphorus (Gu et al., 2021).
334 Considering that the nitrogen footprint in residential and commercial land is higher than
335 cropland, the treatment of Nr pollution in the Lake Tai watershed needs to further reduce the
336 nitrogen input from residential and commercial lands, such as WWTP effluent in urban areas
337 and direct discharge of domestic wastewater from rural settlements.

338



340 **Fig. 6 The policy in nitrogen governance from 1995 to 2015.** All information is from Lake
 341 Tai Comprehensive Treatment Phase I Project, the overall plan of water environment
 342 comprehensive treatment in the Lake Tai watershed, the Five-Year Plan of China, and other
 343 public documents. ① In 1995, the 9th Five-Year Plan set the goal of total water pollutant
 344 discharge control in the Lake Tai watershed in 2000, and proposed a long-term goal was to
 345 solve the problem of eutrophication in Lake Tai by 2010. ② On December 31, 1998, nearly
 346 1,000 law enforcement officers conducted on-site inspections and enforcement of the industrial
 347 pollution from polluting enterprises in the Lake Tai watershed, known as the “zero” actions. ③
 348 In 2000, the 10th Five-Year Plan proposed to implement cleaner production in the Lake Tai
 349 watershed to control the total amount of pollutants entering the lake. The responsibility for the
 350 prevention and control of water pollution in Lake Tai was assigned to the people's governments
 351 at all levels to control water pollution. ④ In 2002, water from the Yangtze River was
 352 transferred from other areas to Lake Tai, which accelerated the flow in the water body of Lake
 353 Tai, improving the self-purification ability of the water body, and reducing the pollution. ⑤ In
 354 2005, the 11th Five-Year Plan carried out point source pollution control for heavily polluting
 355 industries, upgrading urban sewage treatment plants, implementing rural non-point source
 356 pollution control, and comprehensive improvement of rivers as part of the lake and ecological
 357 restoration work to improve the water quality in the Lake Tai watershed. ⑥ In 2007, a
 358 cyanobacteria bloom broke out in the Lake Tai watershed, affecting the normal supply of
 359 drinking water for residents and shutting down a large number of factories in the basin. ⑦ In
 360 2008, the overall scheme proposed industrial point source treatment and agricultural non-point
 361 source pollution treatment, implemented in the river chief system. ⑧ In 2010, the 12th Five-
 362 Year Plan proposed to harness the Lake Tai watershed for the following five aspects: promoting
 363 economic transformation and upgrading; controlling agricultural non-point source and urban

364 domestic pollution; restoring the Lake Tai ecosystem; promoting the collaborative management
365 of river basin; and advocating the construction of a water-saving society. ⑨ In 2011,
366 regulations were proposed to ensure drinking water safety, protection of water resources and
367 prevention of water pollution in the Lake Tai watershed through supervision and clear legal
368 responsibilities.

369

370 **5. Conclusions**

371 Nitrogen pollution has undergone marked changes with socioeconomic development.
372 Calculating a nitrogen footprint is a useful tool to link water pollution and human activities. In
373 this paper, a turning point of water quality in the Lake Tai watershed was identified around the
374 mid-2000s, which is well illustrated by the total nitrogen footprint despite an increase in the
375 overall population in this region. With urbanization, urban residential and commercial areas
376 have become new hotspots of the nitrogen footprint compared to agricultural land uses.
377 Meanwhile, part of the nitrogen footprint derived from agricultural production has been
378 exported to other regions with urbanization and land use change reducing the availability of
379 agricultural land. Therefore, government policies should focus on reducing the per capita
380 nitrogen footprint of residential and commercial areas going forward. Increasing the number
381 and effectiveness of WWTPs to reduce the direct discharge of Nr through wastewater should
382 be done, hence, improving and enforcing the standards of WWTPs to reduce the nitrogen
383 emissions in the effluent. Sustainable development at the watershed scale can be realized
384 through optimizing the nitrogen footprint across all relevant sources, as illustrated by this study.

385

386 **References:**

- 387 Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante,
388 M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J.W., Fenn, M.,
389 Gilliam, F., Nordin, A., Pardo, L., De Vries, W., 2010. Global assessment of nitrogen
390 deposition effects on terrestrial plant diversity: A synthesis. *Ecol. Appl.* 20, 30-59.
- 391 Deng, Y., Wheatley, A., 2016. Wastewater treatment in Chinese rural areas. *Asian Journal of*
392 *Water, Environment and Pollution* 13, 1-11.
- 393 FAO (Food and Agricultural Organization), 2020. FAOSTAT: FAO Statistical Databases, UN
394 Food and Agricultural Organization, Rome, Italy.
- 395 Gu, B., Ju, X., Chang, J., Ge, Y., Vitousek, P.M., 2015. Integrated reactive nitrogen budgets
396 and future trends in China. *Proceedings of the National Academy of Sciences* 112, 8792-
397 8797.
- 398 Gu, B., Leach, A.M., Ma, L., Galloway, J.N., Chang, S.X., Ge, Y., Chang, J., 2013. Nitrogen
399 footprint in China: Food, energy, and nonfood goods. *Environ. Sci. Technol.* 47, 9217-
400 9224.
- 401 Gu, B., van Grinsven, H.J.M., Lam, S.K., Oenema, O., Sutton, M.A., Mosier, A., Chen, D.,
402 2021. A credit system to solve agricultural nitrogen pollution. *The Innovation* 2, 100079.
- 403 Hobbs, W.O., Lafrancois, B.M., Stottlemeyer, R., Toczydlowski, D., Engstrom, D.R., Edlund,
404 M.B., Almendinger, J.E., Strock, K.E., VanderMeulen, D., Elias, J.E., Saros, J.E., 2016.
405 Nitrogen deposition to lakes in national parks of the western Great Lakes region: Isotopic
406 signatures, watershed retention, and algal shifts. *Global Biogeochem. Cy.* 30, 514-533.
- 407 Huang, W., Gao, B., Huang, Y., Zhang, Z., Xu, S., Xu, L., Cui, S., 2019. Transforming nitrogen
408 management of the urban food system in a food-sink city. *J. Environ. Manage.* 249,
409 109180.
- 410 Jin, X., Gao, J., Zhao, G., 2006. Impacts of 20 year socio-economic development on the trend
411 of aquatic environment of the Taihu Basin. *Resources and Environment in the Yangtze*
412 *Basin*, 298-302. (in Chinese)
- 413 Ju, X., Gu, B., Wu, Y., Galloway, J.N., 2016. Reducing China's fertilizer use by increasing
414 farm size. *Global Environmental Change* 41, 26-32.
- 415 Leach, A.M., Galloway, J.N., Bleeker, A., Erisman, J.W., Kohn, R., Kitzes, J., 2012. A nitrogen
416 footprint model to help consumers understand their role in nitrogen losses to the
417 environment. *Environmental Development* 1, 40-66.
- 418 Leach, A.M., Majidi, A.N., Galloway, J.N., Greene, A.J., 2013. Toward institutional
419 sustainability: A nitrogen footprint model for a university. *Sustainability: The Journal of*
420 *Record* 6, 211-219.

421 Liang, X., Leach, A.M., Galloway, J.N., Gu, B., Lam, S.K., Chen, D., 2016. Beef and coal are
422 key drivers of Australia's high nitrogen footprint. *Sci. Rep.-UK* 6, 1-8.

423 Liang, X., Ng, E.L., Lam, S.K., Castner, E.A., Leach, A.M., Gu, B., Healey, G., Galloway, J.N.,
424 Chen, D., 2018. The nitrogen footprint for an Australian university: Institutional change
425 for corporate sustainability. *J. Clean. Prod.* 197, 534-541.

426 Liu, X., Zhang, Y., Han, W., Tang, A., Shen, J., Cui, Z., Vitousek, P., Erisman, J.W., Goulding,
427 K., Christie, P., Fangmeier, A., Zhang, F., 2013. Enhanced nitrogen deposition over China.
428 *Nature* 494, 459-462.

429 MHURDPRC (Ministry of Housing and Urban-Rural Development of the People's Republic of
430 China), 2005. Present situation and problems of rural human settlements. Ministry of
431 Housing and Urban-Rural Development of the People's Republic of China, Beijing. (in
432 Chinese)

433 Qian, Y., He, P., 2009. An analysis on the changes in the water quality in Taihu Basin during
434 1998-2006. *Acta Agriculturae Universitatis Jiangxiensis* 31, 370-374. (in Chinese)

435 Stevens, C.J., Leach, A.M., Dale, S., Galloway, J.N., 2014. Personal nitrogen footprint tool for
436 the United Kingdom. *Environ Sci Process Impacts* 16, 1563-1569.

437 Ti, C., Gao, B., Luo, Y., Wang, S., Chang, S.X., Yan, X., 2018. Dry deposition of N has a major
438 impact on surface water quality in the Taihu Lake region in southeast China. *Atmos.*
439 *Environ.* 190, 1-8.

440 Wu, Y., Xi, X., Tang, X., Luo, D., Gu, B., Lam, S.K., Vitousek, P.M., Chen, D., 2018. Policy
441 distortions, farm size, and the overuse of agricultural chemicals in China. *Proceedings of*
442 *the National Academy of Sciences* 115, 7010-7015.

443 Yu, C., Huang, X., Chen, H., Godfray, H.C.J., Wright, J.S., Hall, J.W., Gong, P., Ni, S., Qiao,
444 S., Huang, G., Xiao, Y., Zhang, J., Feng, Z., Ju, X., Ciais, P., Stenseth, N.C., Hessen, D.O.,
445 Sun, Z., Yu, L., Cai, W., Fu, H., Huang, X., Zhang, C., Liu, H., Taylor, J., 2019. Managing
446 nitrogen to restore water quality in China. *Nature* 567, 516-520.

447 Zhai, F.Y., Du, S.F., Wang, Z.H., Zhang, J.G., Du, W.W., Popkin, B.M., 2014. Dynamics of
448 the Chinese diet and the role of urbanicity, 1991-2011. *Obes. Rev.* 15, 16-26.

449 Zhang, C., Hu, R., 2020. Does fertilizer use intensity respond to the urban-rural income gap?
450 Evidence from a dynamic panel-data analysis in China. *Sustainability-Basel* 12, 1-15.

451 Zhang, C., Liu, S., Wu, S., Jin, S., Reis, S., Liu, H., Gu, B., 2019. Rebuilding the linkage
452 between livestock and cropland to mitigate agricultural pollution in China. *Resources,*
453 *Conservation and Recycling* 144, 65-73.

454 Zhang, Q., Streets, D.G., He, K., Wang, Y., Richter, A., Burrows, J.P., Uno, I., Jang, C.J., Chen,
455 D., Yao, Z., Lei, Y., 2007. NO_x emission trends for China, 1995–2004: The view from the

456 ground and the view from space. *Journal of Geophysical Research* 112, 1-18.
457 Zhang, X., Davidson, E.A., Mauzerall, D.L., Searchinger, T.D., Dumas, P., Shen, Y., 2015.
458 Managing nitrogen for sustainable development. *Nature* 528, 51-59.
459