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Page 1 of 31

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## 33 Abstract

Managing reactive nitrogen  $(N_r)$  to achieve a sustainable balance between production of 34 food, feed and fibre, and environmental protection is a grand challenge in the context of 35 an increasingly affluent society. Here, we propose a novel framework for national 36 nitrogen (N) assessments enabling a more consistent comparison of the uses, losses and 37 impacts of Nr between countries, and improvement of Nr management for sustainable 38 development at national and regional scales. This framework includes four key 39 components: national scale N budgets, validation of N fluxes, cost-benefit analysis and 40 Nr management strategies. We identify four critical factors for Nr management to 41 achieve the sustainable development goals: N use efficiency (NUE), Nr recycling ratio 42 (e.g., ratio of livestock excretion applied to cropland), human dietary patterns and food 43 waste ratio. This framework was partly adopted from the European Nitrogen 44 Assessment and now is successfully applied to China, where it contributed to trigger 45 policy interventions towards improvements for future sustainable use of Nr. We 46 demonstrate how other countries can also benefit from the application our framework, 47 in order to include sustainable Nr management under future challenges of growing 48 49 population, hence contributing to the achievement of some key sustainable development goals (SDGs). 50

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Key words: Cost-benefit analysis; Environmental protection; Food security; Nitrogen
budget; Nitrogen use efficiency; Socioeconomic barriers; Sustainable development
goals

55

## 56 Introduction

Human activities have more than tripled the global reactive nitrogen  $(N_r)$  creation rates 57 and inputs to terrestrial ecosystems through industrial N fixation (Haber-Bosch process, 58 HBNF), cultivated biological N fixation (CBNF) and unintended N oxide (NO<sub>x</sub>) 59 emissions from fossil fuel combustion, compared to the pre-industrial era<sup>1</sup>. The 60 elevated N inputs to agriculture and forestry have substantially increased the supply of 61 food, energy and materials, but also result in detrimental effects on the environment, 62 human health, ecosystem structure and function, and climate <sup>2, 3</sup>. About 50% of the 63 global population in the late-20<sup>th</sup> century was sustained by food production fertilized 64

with N derived from the HBNF process <sup>4</sup>. However, more than half of the N used in 65 agriculture is lost to the environment, largely as  $N_r^{1,5}$ . The same molecule of  $N_r$  can 66 cause a sequence of effects and result in N cascading across multiple scales and 67 environmental compartments <sup>6</sup>, involving complex anthropogenic, biological, chemical, 68 physical and geological processes <sup>7</sup> (Figure 1). A cost of 70-320 billion Euro have been 69 attributed to detrimental effects on the environment and human health in Europe<sup>8</sup>. 70 There is a substantial disparity in N use between global regions or countries, ranging 71 from too much N use in Europe, the United States (US), India or China, to too little in 72 Africa<sup>9</sup>, impacting on the sustainability and food security in those regions. The regional 73 differences in N use are determined by both human activities such as economic 74 development <sup>1, 5</sup>, and natural conditions such as climate and soil conditions <sup>10, 11</sup>. 75 Understanding and managing the disparity in regional N uses and both their beneficial 76 and adverse effects is crucial for global sustainable development. 77 Around the globe, substantial efforts have been made to understand and 78 quantitatively assess the N cycles from the field to local/regional scales and the 79 implication for food production and environmental protection<sup>12</sup>. The 7<sup>th</sup> international N 80 conference held in Melbourne, Australia, in 2016 published the "Melbourne Declaration 81 on Responsible Nitrogen Management for a Sustainable Future" 82 (www.ini2016.com/melbourne-declaration) emphasising the need to conduct regional 83 and global N studies to address the issues of  $N_r$  in food security, energy, health, 84 environment, biodiversity and climate change around the world. The US, the European 85 Union (EU) and India, have already completed comprehensive assessments of their N 86 sources, fluxes, and impacts <sup>7, 13, 14</sup>. Some regions have implemented a series of policy 87 measures to reduce Nr losses <sup>15, 16</sup>. However, specific findings and policies developed 88 e.g. in the US and Europe may not be applicable to other regions, because the N 89 challenges generally differ substantially between regions due to biogeochemical, socio-90 economical, cultural and political factors <sup>5</sup>. Other countries are conducting or intend to 91 conduct N assessments to improve their  $N_r$  management, such as China <sup>17</sup>. Here, we 92 propose a first comprehensive and uniform framework for the compilation of national N 93 assessments, in order to better understand N cycling at national (or regional) scale, and 94 to determine which policy can be developed and effectively implemented, as well as 95 improving the transferability of this understanding and knowledge between regions and 96

97 countries.

- 98 The framework and methodology for the national scale N assessment we have
- 99 developed addresses several N related Sustainable Development Goals
- 100 (www.undp.org/content/undp/en/home/sustainable-development-goals.html), in
- 101 particular Zero Hunger (SDG2), Good Health and Well-Being (SDG3), Sustainable
- 102 Cities and Communities (SDG11), Responsible Consumption and Production (SDG12),
- 103 Climate Action (SDG13) and Life on Land (SDG16), while contributing to other SDGs
- and global objectives, e.g. the Convention on Biological Diversity (CBD). The
- 105 framework includes four building blocks: (1) national N budget, (2) validation of N
- flux, (3) cost-benefit analysis, and (4)  $N_r$  management strategies. With these building
- 107 blocks, a pathway for N<sub>r</sub> sustainable management can be mapped out by integrating the
- 108 socioeconomic factors and scientific understanding of the key processes of N cycling.
- 109 With these approaches, N assessments in different countries can be compared on the
- same basis, contributing to a better understanding of regional N cycles, interactions
- 111 across spatial scales, their driving forces and consequences. This will aid policy makers
- in formulating national policies towards sustainable N<sub>r</sub> management in a wider regional
- 113 context and in interaction with international communities, while considering
- international trade and cross-transboundary pollution, which exhibit increasingly
- 115 important effects on global sustainability.
- 116

#### 117 Key scientific questions addressed through N assessments

# 118 How are N uses and losses affected by natural and human factors? (Q1)

Unlike the globally uniform effects of a unit emission of a greenhouse gas on climate 119 change  $^{18}$ , the environmental impact of a unit of N<sub>r</sub> emission depends on the source, 120 chemical form and location 8, 19. Therefore, the environmental impacts of changes in N 121 cycles are highly spatially explicit and thus mainly expressed at a regional or even local 122 scale, and also with cross-boundary impacts<sup>2</sup>. Anthropogenic N inputs from different 123 sources are generally well known, but the variation in their magnitude for some fluxes 124 such as the biological N fixation (BNF) is still quite uncertain<sup>1</sup>. These uncertainties 125 inevitably cascade through the N cycle on regional scale and result in multiple 126 consequences, except for climate and ozone depletion impacts of N<sub>2</sub>O emissions leading 127 to effects on global scale <sup>6</sup>. 128

Economic development increases fertilizer use through easier access to and reduced 129 prices of fertilizer products, whereas improved management practices reduce N 130 fertilizer use via increasing N use efficiency (NUE) <sup>5</sup>. NO<sub>x</sub> emissions from fossil fuel 131 combustion are directly related to energy consumption and technological development 132 levels that control the emission rate during the combustion process <sup>20</sup>. NH<sub>3</sub> emissions 133 show a strong temperature dependence, with more NH<sub>3</sub> emitted in warmer temperatures 134 <sup>21</sup>. Generally, natural factors such as precipitation and temperature affect N cycling 135 through changing N transformation rates and cycling pathways <sup>11</sup>. For instance, in dry 136 regions more N losses through NH<sub>3</sub> emissions to the air occur, while transportation to 137 groundwater and surface water is commonly found to be higher in humid regions <sup>22</sup>. 138 Overall, regional variations of the N fluxes and their environmental impacts may be 139 attributed to anthropogenic factors such as economic growth, technological 140 advancement, policy innovation, and natural factors such as air temperature and 141 precipitation <sup>5</sup> (Figure 2). These variations result in substantial uncertainties in N flows 142 and fates <sup>9</sup>. Although recent studies have improved our understanding and hence the 143 more accurate quantification of the global N flows and fates <sup>1</sup>, uncertainties regarding N 144 fluxes are still substantial due to inadequate understanding of N fates and their driving 145 factors on a regional scale<sup>17</sup>. More regional work is needed to better understand the 146 147 mechanisms and constrain the uncertainties.

148

#### 149 *How to quantify the societal costs and benefits of N uses? (Q2)*

Quantifying the costs and benefits of N use is one way to address multiple, adverse as 150 well as beneficial impacts of different chemical forms and sources of Nr (Figure 2). A 151 consistent approach to the valuation of multiple impacts allows a weighted comparison 152 of various uses (inputs) and emissions of Nr. Studies on the cost-benefit analysis of N 153 cycles on a regional scale are scarce, and only selected studies<sup>8, 19, 23</sup> attempted a 154 comprehensive assessment of costs and benefits in monetary terms. Large ranges and 155 uncertainties exist in these previous estimates, e.g., the cost per kilogram of Nr loss 156 ranges over one order of magnitude  $^{8, 24}$ . The variation of population exposure to  $N_r$ 157 pollutants in different regions might equally be a major cause of this large range <sup>19</sup>. 158 Moreover, effects on ecosystem function and services are important, but typically not 159 included <sup>24, 25</sup>. 160

A substantial element of human health costs arising from atmospheric N<sub>r</sub> pollution 161 is the quantification of the loss of healthy life years due to exposure to fine (N-162 containing) particulate matter. NO<sub>x</sub> contributing to the formation of tropospheric ozone 163 and to a lesser extent, especially in urban areas, direct exposure to ambient 164 concentrations of NO<sub>2</sub> also affects human health <sup>8</sup>. The societal cost of human health 165 impacts depends on the statistical value attributed to a healthy life year, which is highly 166 related to the income level of a country <sup>26</sup>. This illustrates that national assessments on 167 the cost and benefit of Nr uses and losses are crucial, as they take into consideration 168 national characteristics, such as population exposure rate and value of a healthy life 169 year, and determine the most appropriate parameters for the analysis. To make the 170 results from different countries comparable, a tiered approach is proposed to quantify 171 the cost-benefit of N<sub>r</sub> uses and losses. We distinguish five tiers that can be developed 172 from criteria for environmental quality (Tier 1), to impacts of Nr pollution on 173 ecosystems and health (Tier 2), nationally agreed policy objectives for environmental 174 quality (Tier 3), metrics to aggregate impacts Nr on human health (e.g. Disability 175 Adjusted of Life Years, DALY) and ecosystems (e.g. biodiversity or services) (Tier 4) 176 and a loss or gain of prosperity or welfare in monetary terms (Tier 5). More details 177 about the five tiers can be found in SI text. 178

179

# 180 What are the socioeconomic barriers constraining the more sustainable use of N? 181 (Q3)

To mitigate N<sub>r</sub> pollution, previous studies mainly focused on technological options, with 182 little consideration of the context or the implementation costs of regulations <sup>17</sup>. For 183 instance, the 4R approach (applying N fertilizer with the right type and right amount, at 184 the right time and right location) has been recommended for many years in the US and 185 EU<sup>16</sup>, but has not been widely adopted in China owing to local constraints <sup>27</sup>. Excessive 186 N fertilization is common in China, as a consequence of the average farm size being 187 smaller than 0.1 hectare, which restricts the usage of advanced machineries, 188 communication, information and training required for the implementation of the 4R 189 approach <sup>27</sup>. Cui et al <sup>28</sup> engaged in training activities aiming to enable millions of 190 smallholder farmers to implement advanced technologies; however, due to the high cost 191 involved, it is difficult to maintain the skill levels after delivering training or extending 192

such capabilities to a wider base of smallholder farmers <sup>29</sup>. Meanwhile, income from 193 such small farms accounts for very little fraction of family income, reducing the 194 incentive of better agronomic management practices. It implies that the socioeconomic 195 barriers are sometimes critical for the sustainable use of N, even if technological or 196 management approaches exist at zero or negative cost to the farmer. Meanwhile, the 197 cost per unit of prevented N<sub>r</sub> emission normally increases with cumulative emission 198 reduction<sup>8</sup>, suggesting it may be necessary to gradually adjust approaches to mitigate 199 N<sub>r</sub> pollution as more stringent emission controls and methods for N<sub>r</sub> management are 200 implemented. Hence, more work is needed to better understand the driving forces and 201 barriers underlying these non-linear mitigation pathways. Also more work is needed on 202 cost efficiency and cost-benefit analysis, including considering national and socio-203 economic characteristics (Figure 2). 204

Apart from the implementation costs of regulations, other barriers such as human 205 behaviour related factors, e.g., culture, dietary preference, also affect the sustainable use 206 of N<sup>30</sup> and opportunities to improve management. For instance, western diets (e.g., in 207 the US) prefer beef over pork, while eastern diets (e.g., in China) prefer pork over beef. 208 209 This implies that dietary structure regulations must consider the dietary culture in 210 different countries to be effective. Policies or regulations developed in some countries may not transfer well to other countries due to the difference in socioeconomic barriers 211 212 that constrain the sustainable use of N. Identifying and quantifying these barriers for the implementation of sustainable use of Nr through social surveys, sensitivity analyses and 213 cost-benefit analyses on national scale, working with social science experts and 214 economists in transdisciplinary contexts, can benefit a wider adoption of N regulation 215 216 measures.

217

## 218 Methodology of the framework

Here, we introduce a hierarchical approach of how to conduct N assessments to achieve

a sustainable and efficient use of  $N_r$ . The methodology broadly relates to the <u>D</u>river-

221 <u>Pressure-State-Impact-Response</u> (DPSIR) concept <sup>31</sup> (Figure 3). It comprises four

stages (Figure 4). <u>At the first stage</u>, an integrated N budget is compiled using a mass

balance model to analyse interactions between major components. All the sources,

flows, losses, and the NUE in a country are quantified, including time series to reflect

the past and current status and trends of N<sub>r</sub> use. The first research questions (Q1), and 225 the Driver and Pressure of the DPSIR concept are addressed at this stage (Figure 3). 226 At the second stage, data compiled from independent national field monitoring 227 programs, remote sensing, and published data are used for comparison with those N 228 fluxes obtained from the mass balance model from Stage 1 in order to validate model 229 results. The results of the compiled N budget are total amounts of N fluxes, which differ 230 from environmental quality objectives, i.e. Nr concentrations. Thus, a downscaling of 231 national N fluxes to match with data from environmental quality monitoring at an 232 appropriate, comparable scale is needed. The Pressure and State of the DPSIR concept 233 will be addressed at this stage (Figure 3). 234

At the third stage, a cost-benefit analysis is applied to estimate both the costs and benefits of  $N_r$  use on ecosystems, human health and economy, considering the excess and accumulation, respectively the deficiency of  $N_r$  in the environment. The second key research question (Q2) and the *State* and *Impact* of the DPSIR concept are addressed at this stage.

At the final (fourth) stage, the results from the first three stages are integrated to assess how to build a sustainable future through improved  $N_r$  management (e.g., measures to maximize the benefits while minimizing the costs and damages of  $N_r$  uses) embedded in policies, institutions and regulations. The potential pathways to ensure an effective implementation of these regulations to overcome the socioeconomic barriers are also discussed. The last research question (Q3), and the *Response* of the DPSIR concept are addressed at this stage.

247

## 248 N budget modelling

To understand the changes of N fluxes, several datasets from both national statistics and 249 global databases such as the Food and Agriculture Organization (FAO)<sup>32</sup>and 250 International Fertilizer Industry Association (IFA) <sup>33</sup> can be accessed and compiled for 251 analysis. Two types of data should be taken into account: human activities and N 252 cycling parameters (e.g., NH<sub>3</sub> emission, runoff, and denitrification) (Figure 4). The 253 datasets of human activities cover historical changes of population, urbanisation, 254 production (industrial, crop, livestock and aquaculture), consumption (food, non-food 255 goods, energy), international trade (e.g., grains, animal products, fertilizers), land use 256

and management related to N cycling, such as manure recycling and wastewater 257 treatment. Long-term datasets provide the best basis for a comprehensive assessment, 258 e.g. starting from the year 1961 (the year when FAO database was available). 259 For N parameters, literature reviews are required to complement data on all of the 260 relevant N cycling parameters, such as NH<sub>3</sub> emission ratios (% of total N applied), 261 which are indicative of the local situation <sup>34</sup>, such as dry climates in Australia. Some 262 global models containing regionally specific parameters can also be used for the 263 construction of such a dataset, e.g., IMAGE <sup>35</sup>, IMAGE-GNM <sup>36</sup>, GLOBIOM <sup>37</sup> and 264 MAgPIE <sup>38</sup>. However, they may not contain all N related subsystems, for instance, as 265 many models do not include an explicit industry subsystem. To understand the complete 266 N cycling in a country, we propose a 14-subsystem model which enables a 267 comprehensive assessment how N flows among different functional units, such as from 268 cropland to livestock. This 14-subsystem model can also help to quantitatively assess 269 and subsequently reduce the uncertainties of N fluxes calculation through robustly 270 constraining the interacting fluxes among different subsystems<sup>17</sup>. The 14 subsystems 271 include industry, cropland, grassland, forest, urban green land, livestock, aquaculture, 272 pet, human, wastewater treatment, garbage treatment, surface water, groundwater, and 273 274 atmosphere. Comprehensive N cycling within the 14-subsystem has been formally implemented in the CHANS model (Coupled Human And Natural Systems) for China 275 276 <sup>17</sup>, which can be easily adapted and applied to other countries. Other models can also be 277 used to calculate a complete or parts of a N budget based on similar mass balance principles, such as IMAGE <sup>35</sup> and MAgPIE <sup>38</sup>. 278

The basic principle of mass balance for the whole system and 14 subsystems is:

280 
$$\sum_{h=1}^{m} IN_{h} = \sum_{g=1}^{n} OUT_{g} + \sum_{k=1}^{p} ACC_{k}$$

where  $IN_h$  and  $OUT_g$  represent the N inputs and outputs, respectively, and  $ACC_k$ represents the N accumulations. Most N<sub>r</sub> inputs transfer between subsystems, for example NO<sub>x</sub> emissions from fossil fuel combustion deposit onto three major domains, natural land (i.e., forest, natural grassland and extensive graze land), managed grassland (including intensive grazing land and cropland) and freshwater and marine water bodies, and it can undergo further transformations and result in a variety of fluxes in and from these landscapes. N inputs to a country include HBNF, BNF, fossil fuel combustion, imports of N-containing products, and transboundary transports through atmospheric

- 289 circulation and surface water flows. N outputs across national boundaries include
- riverine N transport to coastal waters, atmospheric circulation that advects N<sub>r</sub> away
- from a country, denitrification, and N-containing product exports. More details of the N
- budget calculation for the 14 subsystems can be found in *SI Text*.
- 293

## 294 *N fluxes validation and uncertainty*

The validation of N fluxes calculated in the N budget model requires monitoring data 295 from independent sources. The national scale N fluxes are downscaled to provincial, 296 county or watershed scale to match and compare with the monitoring data. Two 297 approaches can be used for downscaling: first, applying the national scale assessment to 298 provincial, county or watershed scale if the available data is sufficiently spatially 299 resolved; second, allocating total N fluxes on national scale to smaller scales through 300 proxy indexes or modelling. The calibration of the modelled N budget is required if the 301 validation results suggest a consistent and systematic bias. Then the newly calculated N 302 fluxes need to be revalidated with monitoring data until sufficient agreement is 303 304 achieved. Two types of monitoring data can be utilized for this: ground based monitoring and remote sensing (including Earth Observation) (Figure 4). Ground based 305 306 monitoring covers N<sub>r</sub> concentrations in the environment (air, water and soil). The data 307 can typically be obtained from openly accessible repositories of regulatory monitoring networks, as well as published or ongoing research activities. However, the Nr 308 concentrations monitored cannot be used to validate the N fluxes calculated within the 309 budget model directly, as metrics and spatial resolution often differ. Thus, spatial 310 patterns or temporal trends of these Nr concentrations in the environment are used to 311 validate and calibrate the calculation of N fluxes. Meanwhile, some models which 312 quantitatively assess atmospheric transmission of pollutants such as the Community 313 Multi-scale Air Quality model (CMAQ) can be used to link the N fluxes to the N<sub>r</sub> 314 concentrations in the environment <sup>39</sup>. 315 Remote sensing data can also be used to monitor the Nr concentrations in the air. 316

Column concentrations of NH<sub>3</sub> and NO<sub>2</sub> from satellite instruments such as Infrared
 Atmospheric Sounding Interferometer (IASI), MOderate resolution Imaging
 Spectroradiometer (MODIS) or Multi-angle Imaging SpectroRadiometer (MISR)

generate spatially explicit maps of  $N_r$  concentrations for comparison with N budget results <sup>34, 40-42</sup>. Some re-sampling and weighted mean methods are typically applied to make spatial correlations comparable <sup>40, 41</sup>. A spatial and temporal correlation analysis between N budgets and satellite monitoring results can be conducted for the validation (Figure 4). Normalized Difference Vegetation Index (NDVI) and land use data can be used to validate the national vegetation and crop production datasets to ensure spatial patterns of statistical data are robust <sup>43</sup>.

The accuracy of N fluxes is critical for the subsequent costs-benefit analysis and 327 design of management strategies. This accuracy is limited by our understanding of N 328 cycles, the quality of the data, and the applicability of the calculated coefficients. The 329 input-output calculations of the 14 subsystems are based on our current understanding 330 of N cycles, and uncertainty quantifications have been introduced in areas where we 331 consider our understanding to be less advanced, such as denitrification <sup>17</sup>. The quality of 332 basic official data e.g. on food production and population consumption is important to 333 the overall uncertainty of the estimation. We believe that the official statistics are a 334 sufficiently reliable source of data for the analyses, and the confidence rating for the 335 related N fluxes such as HBNF and N in food (and straw), goods production and 336 consumption (from FAO statistics) can be considered as very high<sup>32</sup>. Nevertheless, N 337 cycling parameters usually have large uncertainty ranges because they are affected by 338 339 many natural (e.g. temperature) and anthropogenic (e.g. technology) factors <sup>34</sup>. Thus, N fluxes that are calculated from statistical data and N parameters have much higher levels 340 of uncertainty compared to the primary N fluxes such as food production. To offset 341 these uncertainties, the independently-calculated or measured data can be used to 342 calibrate these N fluxes (Figure 4). For example, soil carbon sink can be used to validate 343 the uncertainties of estimates on soil organic N accumulation <sup>17</sup>. 344

345

## 346 Cost-benefit analysis

Benefits of N<sub>r</sub> use are the results of increased production of the "good", decreased production of the "bad" outputs or decreased costs of measures or practices <sup>8, 44</sup>. N<sub>r</sub> costs are the result of decreased production of the "good", increased production of the "bad" or increased cost of measures or practices. Cost-benefit analysis of N can be applied to estimate the societal cost associated with N mitigation strategies (including the cost of

implementation of measures) or to calculate values and trends of societal net and grosscost of N pollution (excluding the cost of measures).

To quantify the societal value of these goods such as crops, livestock, biofuel, and 354 non-fertilizer industrial N products, a straight forward approach would be to apply 355 market prices and purchasing power parity (PPP) correction. Unlike the goods that can 356 be valued by using marketing prices, other benefits such as ecosystem service (ES) need 357 to be estimated based on non-market valuation approaches such as those presented in 358 the framework of Millennium Ecosystem Assessment (MEA)<sup>45</sup>. Apart from the 359 valuation of  $N_r$  contributions to climate change effects that can also be estimated based 360 on carbon price on the global market, the non-market values of other services are 361 mainly quantified using willingness-to-pay (WTP) approaches, or restoration costs <sup>25</sup> 362 (Figure 4). 363

The costs include the market costs to produce synthetic ammonia and its 364 derivatives, the costs of damage to the environment, human health, ecosystems, climate, 365 and society (e.g., reductions of labour productivity and crop yield), and costs to mitigate 366 N<sub>r</sub> pollution. The costs of intended N<sub>r</sub> uses are typically straightforward to quantify by 367 including values based on market prices of resources and other inputs to produce these 368 products or services <sup>8</sup>. For human health damage costs, the values for mortality and 369 morbidity (e.g. loss of a healthy human life year), as well as costs arising from 370 371 healthcare systems need to be quantified through N critical loading experiments and dose-response-effects modelling <sup>8, 46</sup>. For ecosystems and resource degradation, the 372 restoration cost or WTP to prevent or restore biodiversity loss and associated ES can be 373 374 used <sup>25</sup>.

375

#### 376 Scenario analysis and management

The findings from the first three stages provide an integrated picture of the N cycling in a country from the past to the present, and their costs and benefits. To maximize the benefits of N<sub>r</sub> uses while minimizing its costs, scenario analyses can be conducted to understand how different measures affect the sustainable uses of N<sub>r</sub> (Figure 4). Firstly, sensitivity analysis tests which parameters exert dominant effects on the target N fluxes by using N budget models such as MAgPIE and CHANS <sup>17, 38</sup>. Then, the potential changes in N fluxes and their costs and benefits under different scenarios can be

assessed. Various management strategies can be identified based on the results of the
scenario analysis, considering both the potential of N flux changes and the related costs
and benefits.

*Scenario analysis.* For scenario development, several parameters, including NUE, N recycling ratio, dietary pattern and food waste ratio, have been identified to have substantial effects on the mitigation of  $N_r$  losses <sup>17, 38</sup>. Human N requirement (mainly food N) is determined by dietary patterns and food waste ratios, and the overall N requirement and loss (N budget) can be estimated through integrating the NUE and N recycling ratios<sup>17</sup>. These four parameters can be estimated based on following equations:

$$NUE = \frac{N \text{ in products}}{\text{Total N input for production}}$$

where NUE refers to the efficiency to produce N containing products in a system, such as cropland, livestock, aquaculture, etc. N in products refers to N contained in the final products such as crops, meat & eggs, fishes. Total N input for production refers to the N used such as N fertilizer in cropland and feed for livestock. High NUE refers to a higher ratio of N<sub>r</sub> contained in final useful products, and lower amounts of N<sub>r</sub> lost to the environment <sup>5</sup>.

401 N recycling ratio = 
$$\frac{\text{N reused}}{\text{Total N residue}}$$

where N recycling ratio refers to the ratio of  $N_r$  reused for production, such as the manure recycled to cropland for production. N reused refers to the part of  $N_r$  residue or waste reused for production, and Total N residue refers to total N residue generated such as total manure generated. The N recycling ratio mainly includes the recycled content of livestock and human excretion and straw returned to the agricultural production systems. This can increase the NUE indirectly through reducing the overall N losses.

409Dietary pattern = 
$$\frac{\text{Animal protein}}{\text{Total protein consumed}}$$
410Food waste ratio =  $\frac{\text{Food waste}}{\text{Food supply}}$ 

411 where dietary pattern refers to the ratio of human protein consumption provided by

animal products such as meat and eggs. Total protein consumed includes both vegetal

and animal proteins. The food waste ratio refers to how much of food supplied is not consumed, usually wasted during storage, distribution and disposal. Regulations of dietary pattern and food waste mainly benefit the sustainable  $N_r$  uses through reducing the overall  $N_r$  demands by the end-users<sup>30</sup>.

To simulate these Nr uses and losses, shared social pathway (SSP) storylines 417 present a useful approach to estimate the population, urbanization and per capita GDP in 418 the future <sup>38</sup>. Many studies on the mitigation of N losses followed the scenarios of IPCC 419 with regard to future projections of greenhouse gas emissions, although the behaviour of 420 carbon is not always consistent with that of N<sup>47</sup>. Therefore, the new scenarios 421 specifically designed with N in mind here are recommended for use in N assessments. 422 Management. Solutions for better managing Nr under different scenarios may not 423 be adopted solely due to high cost, if benefits are not recognised or quantified <sup>24</sup>. 424 Therefore the results from the cost-benefit analysis and scenarios analysis on the 425 optimisation of N fluxes to achieve cost-effective solutions for a sustainable future need 426 to be integrated. The costs and benefits of each scenario could be quantified to identify 427 optimal solutions. Furthermore, whether these selected solutions for the sustainable use 428 of N<sub>r</sub> are feasible requires further analysis of the socioeconomic barriers <sup>27, 48</sup>. These 429 barriers can be economic structure (e.g., farm size), population density, culture, religion, 430 consumer or even dry climate. Thus, social scientists should be involved in assessing 431 432 the feasibilities of the proposed solutions. Finally, several N sustainability indices can be used to translate the scientific results to the public and policy makers to make real 433 impacts, such as the N footprint concepts<sup>49</sup>. N fluxes and their costs and benefits are 434 incorporated into the indices to reflect the quality of products and their environmental 435 effects. 436

437

### 438 Implications of N assessment

This paper presents a generally applicable framework for a comprehensive, 4-stage
national scale N assessment, covering all relevant Nr issues towards a better N
management in a country. While national N budgets have already been completed in
some countries, N<sub>r</sub> related challenges vary with regions and countries and are intricately
linked with local socioeconomic development <sup>7, 14, 17</sup>. Parts of this framework have been
successfully applied to EU<sup>50</sup> and China<sup>17</sup>, compiling a comprehensive N budget and

illustrating potential future trends under different scenarios. We believe that a wider
application of the N assessment to other countries can contribute to a substantial
improvement of global sustainable use of N.

The framework and methodology proposed in this study are inherently 448 interdisciplinary and require the integration of expertise from both natural sciences (e.g., 449 Environmental Science, Soil Science, Ecology, Earth Science, etc.) and social sciences 450 (e.g., Economics, Management, Policy, Law, etc.). Unlike previous studies on N 451 cycling, this study designs an approach for the development of a comprehensive, 452 comparable N assessments, including the interaction between N cycling and 453 socioeconomic issues. Novel interdisciplinary methods proposed here range from site-454 scale monitoring and validation, to regional surveys and remote sensing datasets, 455 combined with budget calculation, modelling, econometric and policy assessment. This 456 framework will advance our knowledge on how to sustainably use Nr at a national scale, 457 and how to face the challenges of Nr releases under future global change and growing 458 459 population pressures.

460

#### 461 Application for China

To feed an increasingly affluent population, China uses about one third of global N<sub>r</sub> to 462 produce food. Nevertheless, it still imports over 100 million tons of grain to meet 463 China's national food demand<sup>32</sup>. Unfortunately, a large amount of N<sub>r</sub> is lost to the 464 environment during the production and consumption of food at the country scale, 465 resulting in serious environmental pollution in China. At the same time, substantial 466 socioeconomic developments have taken place since the late 1970s<sup>19, 51</sup>. To solve the 467 double challenge of producing more food with less pollution, we applied our framework 468 of N assessment to China for the period of 1980 to 2015. 469

470*Nitrogen budget.* Results showed that total  $N_r$  input to China increased from 25 to47171 Tg N yr<sup>-1</sup> between 1980 and 2015, of which 74% and 89% derived from472anthropogenic sources in 1980 and 2015, respectively (Figure 5). After input to the473boundary of China,  $N_r$  cascades through the 14 subsystems, and produces about 20 Tg474N yr<sup>-1</sup> in food and feed, while losses to the environment amount to around 50 Tg N yr<sup>-1</sup>475in 2015. Agricultural sources are responsible for approximately 91% of total NH<sub>3</sub>476emissions; fossil fuel combustion accounts for 90% of total NO<sub>x</sub> emissions; agricultural

and natural sources (forest and surface water) together dominate  $N_2O$  emissions in China. Agricultural sources and human sewage contribute most of the  $N_r$  discharge to surface water, while agricultural sources and landfill leaching are responsible for the bulk of  $N_r$  discharges to groundwater. More detailed information about the N fluxes in

481 China can be found in Figure S1.

Nitrogen flux validation. The modelled Nr fluxes to the environment were 482 validated using data from ground based national monitoring networks of air and water 483 quality and remote sensing data providing column concentrations of NH<sub>3</sub> and NO<sub>2</sub><sup>19, 34</sup>. 484 N fluxes to air (NH<sub>3</sub> and NO<sub>x</sub> emissions) were well validated using remote sensing data 485 with a regression  $(R^2)$  of more than 0.7, while N fluxes to water showed a less strong 486 regression ( $R^2 \sim 0.5$ ). This is due to the complex N cycling processes which occur in 487 water bodies and e.g. denitrification. Increasing the number of monitoring sites for 488 water N concentrations could help to reduce the uncertainty by providing more data for 489 a robust validation of N fluxes to water bodies. 490

491 *Cost-benefit analysis.* Integrating  $N_r$  emissions and population exposure 492 assessments, we calculated that using 2015  $N_r$  fluxes,  $N_r$  emissions to the air (including 493 NH<sub>3</sub>, NO<sub>x</sub> and N<sub>2</sub>O) caused the loss of 16.5 million life years annually in China, a much 494 higher value than the loss of 2.6 million life years found in EU <sup>8, 17</sup>. If converted, these 495 losses of life years equate to a monetary value of around 200 billion US dollars 496 annually. Further analyses of other cost-benefit assessment components, such as 497 ecosystem service impacts due to N<sub>r</sub> loss to water bodies are still ongoing.

498 *Scenario analysis.* An explicit consideration of the following four proposed factors 499 in the analysis could help to better explore potential interventions to mitigate  $N_r$  losses. 500 We found that increasing N use efficiency, optimising diets, increasing N recycling and 501 reducing food waste could decrease total N losses during food production and 502 consumption by about 50%, 25%, 30% and 10%, respectively. Combining feasible 503 changes in these four factors could reduce N losses by the year 2050 to about 60-70% of 504 2015 levels.

505 *Management.* However, to achieve these reductions, socioeconomic barriers need 506 to be addressed as indicated previously. We found that increasing farm size (current 507 average of <0.1 hectare each farm) is the crucial challenge for the implementation of 508 regulatory measures in China, especially interventions to increase NUE and recycling

509	ratio. High labour costs suggest a low level of mechanisation and automation in small
510	farms, which inhibits the application of precision agriculture and fertilization
511	technologies, as well as management based on scientific knowledge and information on
512	application methods <sup>27</sup> . Increasing farm size has been integrated as a viable intervention
513	into the recommendations to achieve the goal of a zero increase and even reduction in
514	fertilizer use in China. Nevertheless, more sophisticated assessments are still ongoing to
515	identify further socioeconomic barriers that inhibit the sustainable use of $N_r$ in China.
516	
517	SUPPORTING INFORMATION
518	SI text
519	Five tiers for the cost-benefit analysis
520	N budgets of 14 subsystems
521	SI methods
522	Nitrogen budget calculations for the 14 subsystems within the CHANS N cycle model
523	SI Figures
524	Figure S1. N cycling among the 14 subsystems in China in 1980.
525	Figure S2. Coupled Human And Natural Systems (CHANS) model structure.
526	SI Tables
527	Table S1-S10, parameters used within the CHANS model.
528	
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- 544

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#### 697 Figure Legend

Figure 1. Major transformation pathways of the global N cycle. Red arrows
represent the pathways of the human meditated N cycle; grey arrows represent the
pathways dominated by microbial activities; green arrows represent the pathways
dominated by plants; blue arrows represent the pathways dominated by atmospheric
chemical reactions. Abbreviations: *BNF*, biological N fixation; CBNF, cultivated
biological N fixation; *Denitri*, denitrification; *HBNF*, Haber-Bosch N fixation; *Nitri*,
nitrification.

705

706 Figure 2. Simplified view of managing N for sustainable development highlighting

the three major scientific questions. (Q1) How are N uses and losses affected by
natural and human factors? (Q2) How to quantify the societal costs and benefits of N
use? (Q3) What are the socioeconomic barriers constraining the more sustainable use of
N? Nr, reactive N.

711

712 Figure 3. Conceptual diagram depicting the linkage between the DPSIR scheme

and the framework of N assessment in this study. DPSIR, Driver–Pressure–State–
Impact–Response; Q1-Q3 represent the three major scientific questions need to be
addressed in Figure 2. (Q1) How are N uses and losses affected by natural and human
factors? (Q2) How to quantify the societal costs and benefits of N use? (Q3) What are
the socioeconomic barriers constraining the more sustainable use of N? NUE, N use
efficiency.

719

Figure 4. An overview of the methodological framework. It describes the approaches
 to mass balance modelling, validation, cost-benefit analysis and management for the
 sustainable future use of N<sub>r</sub>. BNF, biological N fixation; NDVI, normalized differential
 vegetation index; WTPs, willingness to pay.

724

**Figure 5. A simplified N cycling schematic for China for the year 2015.** Pathways

marked in green refer to 'natural' fluxes (to some extent altered by atmospheric  $N_r$ 

deposition), those in blue are intentional anthropogenic fluxes, and those in orange are

vunintentional anthropogenic fluxes. Not all N fluxes were included due to space limits.

729 Nat, Natural; exp., export; wwt, wastewater treatment.

730 Figure 1











