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1 **Towards a generic analytical framework for sustainable nitrogen management:**  
2 **application for China**

3

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## 33 **Abstract**

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

51

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

55

## 56 **Introduction**

57 Human activities have more than tripled the global reactive nitrogen ( $N_r$ ) creation rates  
58 and inputs to terrestrial ecosystems through industrial N fixation (Haber-Bosch process,  
59 HBNF), cultivated biological N fixation (CBNF) and unintended N oxide ( $NO_x$ )  
60 emissions from fossil fuel combustion, compared to the pre-industrial era <sup>1</sup>. The  
61 elevated N inputs to agriculture and forestry have substantially increased the supply of  
62 food, energy and materials, but also result in detrimental effects on the environment,  
63 human health, ecosystem structure and function, and climate <sup>2, 3</sup>. About 50% of the  
64 global population in the late-20<sup>th</sup> century was sustained by food production fertilized

65 with N derived from the HBNF process <sup>4</sup>. However, more than half of the N used in  
66 agriculture is lost to the environment, largely as  $N_r$  <sup>1,5</sup>. The same molecule of  $N_r$  can  
67 cause a sequence of effects and result in N cascading across multiple scales and  
68 environmental compartments <sup>6</sup>, involving complex anthropogenic, biological, chemical,  
69 physical and geological processes <sup>7</sup> (Figure 1). A cost of 70-320 billion Euro have been  
70 attributed to detrimental effects on the environment and human health in Europe <sup>8</sup>.  
71 There is a substantial disparity in N use between global regions or countries, ranging  
72 from too much N use in Europe, the United States (US), India or China, to too little in  
73 Africa<sup>9</sup>, impacting on the sustainability and food security in those regions. The regional  
74 differences in N use are determined by both human activities such as economic  
75 development <sup>1,5</sup>, and natural conditions such as climate and soil conditions <sup>10,11</sup>.  
76 Understanding and managing the disparity in regional N uses and both their beneficial  
77 and adverse effects is crucial for global sustainable development.

78       Around the globe, substantial efforts have been made to understand and  
79 quantitatively assess the N cycles from the field to local/regional scales and the  
80 implication for food production and environmental protection<sup>12</sup>. The 7<sup>th</sup> international N  
81 conference held in Melbourne, Australia, in 2016 published the “Melbourne Declaration  
82 on Responsible Nitrogen Management for a Sustainable Future”  
83 ([www.ini2016.com/melbourne-declaration](http://www.ini2016.com/melbourne-declaration)) emphasising the need to conduct regional  
84 and global N studies to address the issues of  $N_r$  in food security, energy, health,  
85 environment, biodiversity and climate change around the world. The US, the European  
86 Union (EU) and India, have already completed comprehensive assessments of their N  
87 sources, fluxes, and impacts <sup>7, 13, 14</sup>. Some regions have implemented a series of policy  
88 measures to reduce  $N_r$  losses <sup>15, 16</sup>. However, specific findings and policies developed  
89 e.g. in the US and Europe may not be applicable to other regions, because the N  
90 challenges generally differ substantially between regions due to biogeochemical, socio-  
91 economical, cultural and political factors <sup>5</sup>. Other countries are conducting or intend to  
92 conduct N assessments to improve their  $N_r$  management, such as China <sup>17</sup>. Here, we  
93 propose a first comprehensive and uniform framework for the compilation of national N  
94 assessments, in order to better understand N cycling at national (or regional) scale, and  
95 to determine which policy can be developed and effectively implemented, as well as  
96 improving the transferability of this understanding and knowledge between regions and

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](http://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  
104 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  
106 flux, (3) cost-benefit analysis, and (4)  $N_r$  management strategies. With these building  
107 blocks, a pathway for  $N_r$  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  
110 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  
112 in formulating national policies towards sustainable  $N_r$  management in a wider regional  
113 context and in interaction with international communities, while considering  
114 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)*

119 Unlike the globally uniform effects of a unit emission of a greenhouse gas on climate  
120 change<sup>18</sup>, the environmental impact of a unit of  $N_r$  emission depends on the source,  
121 chemical form and location<sup>8,19</sup>. Therefore, the environmental impacts of changes in N  
122 cycles are highly spatially explicit and thus mainly expressed at a regional or even local  
123 scale, and also with cross-boundary impacts<sup>2</sup>. Anthropogenic N inputs from different  
124 sources are generally well known, but the variation in their magnitude for some fluxes  
125 such as the biological N fixation (BNF) is still quite uncertain<sup>1</sup>. These uncertainties  
126 inevitably cascade through the N cycle on regional scale and result in multiple  
127 consequences, except for climate and ozone depletion impacts of  $N_2O$  emissions leading  
128 to effects on global scale<sup>6</sup>.

129 Economic development increases fertilizer use through easier access to and reduced  
130 prices of fertilizer products, whereas improved management practices reduce N  
131 fertilizer use via increasing N use efficiency (NUE)<sup>5</sup>. NO<sub>x</sub> emissions from fossil fuel  
132 combustion are directly related to energy consumption and technological development  
133 levels that control the emission rate during the combustion process<sup>20</sup>. NH<sub>3</sub> emissions  
134 show a strong temperature dependence, with more NH<sub>3</sub> emitted in warmer temperatures  
135<sup>21</sup>. Generally, natural factors such as precipitation and temperature affect N cycling  
136 through changing N transformation rates and cycling pathways<sup>11</sup>. For instance, in dry  
137 regions more N losses through NH<sub>3</sub> emissions to the air occur, while transportation to  
138 groundwater and surface water is commonly found to be higher in humid regions<sup>22</sup>.

139 Overall, regional variations of the N fluxes and their environmental impacts may be  
140 attributed to anthropogenic factors such as economic growth, technological  
141 advancement, policy innovation, and natural factors such as air temperature and  
142 precipitation<sup>5</sup> (Figure 2). These variations result in substantial uncertainties in N flows  
143 and fates<sup>9</sup>. Although recent studies have improved our understanding and hence the  
144 more accurate quantification of the global N flows and fates<sup>1</sup>, uncertainties regarding N  
145 fluxes are still substantial due to inadequate understanding of N fates and their driving  
146 factors on a regional scale<sup>17</sup>. More regional work is needed to better understand the  
147 mechanisms and constrain the uncertainties.

148

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

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

161 A substantial element of human health costs arising from atmospheric  $N_r$  pollution  
162 is the quantification of the loss of healthy life years due to exposure to fine (N-  
163 containing) particulate matter.  $NO_x$  contributing to the formation of tropospheric ozone  
164 and to a lesser extent, especially in urban areas, direct exposure to ambient  
165 concentrations of  $NO_2$  also affects human health<sup>8</sup>. The societal cost of human health  
166 impacts depends on the statistical value attributed to a healthy life year, which is highly  
167 related to the income level of a country<sup>26</sup>. This illustrates that national assessments on  
168 the cost and benefit of  $N_r$  uses and losses are crucial, as they take into consideration  
169 national characteristics, such as population exposure rate and value of a healthy life  
170 year, and determine the most appropriate parameters for the analysis. To make the  
171 results from different countries comparable, a tiered approach is proposed to quantify  
172 the cost-benefit of  $N_r$  uses and losses. We distinguish five tiers that can be developed  
173 from criteria for environmental quality (Tier 1), to impacts of  $N_r$  pollution on  
174 ecosystems and health (Tier 2), nationally agreed policy objectives for environmental  
175 quality (Tier 3), metrics to aggregate impacts  $N_r$  on human health (e.g. Disability  
176 Adjusted of Life Years, DALY) and ecosystems (e.g. biodiversity or services) (Tier 4)  
177 and a loss or gain of prosperity or welfare in monetary terms (Tier 5). More details  
178 about the five tiers can be found in *SI text*.

179

180 ***What are the socioeconomic barriers constraining the more sustainable use of N?***

181 ***(Q3)***

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

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

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

217

### 218 **Methodology of the framework**

219 Here, we introduce a hierarchical approach of how to conduct N assessments to achieve  
220 a sustainable and efficient use of N<sub>r</sub>. The methodology broadly relates to the Driver–  
221 Pressure–State–Impact–Response (DPSIR) concept<sup>31</sup> (Figure 3). It comprises four  
222 stages (Figure 4). At the first stage, an integrated N budget is compiled using a mass  
223 balance model to analyse interactions between major components. All the sources,  
224 flows, losses, and the NUE in a country are quantified, including time series to reflect



225 the past and current status and trends of  $N_r$  use. The first research questions (Q1), and  
226 the *Driver* and *Pressure* of the DPSIR concept are addressed at this stage (Figure 3).

227 At the second stage, data compiled from independent national field monitoring  
228 programs, remote sensing, and published data are used for comparison with those N  
229 fluxes obtained from the mass balance model from Stage 1 in order to validate model  
230 results. The results of the compiled N budget are total amounts of N fluxes, which differ  
231 from environmental quality objectives, i.e.  $N_r$  concentrations. Thus, a downscaling of  
232 national N fluxes to match with data from environmental quality monitoring at an  
233 appropriate, comparable scale is needed. The *Pressure* and *State* of the DPSIR concept  
234 will be addressed at this stage (Figure 3).

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

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

247

### 248 ***N budget modelling***

249 To understand the changes of N fluxes, several datasets from both national statistics and  
250 global databases such as the Food and Agriculture Organization (FAO)<sup>32</sup> and  
251 International Fertilizer Industry Association (IFA)<sup>33</sup> can be accessed and compiled for  
252 analysis. Two types of data should be taken into account: human activities and N  
253 cycling parameters (e.g.,  $NH_3$  emission, runoff, and denitrification) (Figure 4). The  
254 datasets of human activities cover historical changes of population, urbanisation,  
255 production (industrial, crop, livestock and aquaculture), consumption (food, non-food  
256 goods, energy), international trade (e.g., grains, animal products, fertilizers), land use

257 and management related to N cycling, such as manure recycling and wastewater  
 258 treatment. Long-term datasets provide the best basis for a comprehensive assessment,  
 259 e.g. starting from the year 1961 (the year when FAO database was available).

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

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

$$280 \quad \sum_{h=1}^m IN_h = \sum_{g=1}^n OUT_g + \sum_{k=1}^p ACC_k$$

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

288 imports of N-containing products, and transboundary transports through atmospheric  
289 circulation and surface water flows. N outputs across national boundaries include  
290 riverine N transport to coastal waters, atmospheric circulation that advects  $N_r$  away  
291 from a country, denitrification, and N-containing product exports. More details of the N  
292 budget calculation for the 14 subsystems can be found in *SI Text*.

293

#### 294 ***N fluxes validation and uncertainty***

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

316 Remote sensing data can also be used to monitor the  $N_r$  concentrations in the air.  
317 Column concentrations of  $NH_3$  and  $NO_2$  from satellite instruments such as Infrared  
318 Atmospheric Sounding Interferometer (IASI), MOderate resolution Imaging  
319 Spectroradiometer (MODIS) or Multi-angle Imaging SpectroRadiometer (MISR)

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

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

345

### 346 ***Cost-benefit analysis***

347 Benefits of  $N_r$  use are the results of increased production of the “good”, decreased  
348 production of the “bad” outputs or decreased costs of measures or practices<sup>8, 44</sup>.  $N_r$  costs  
349 are the result of decreased production of the “good”, increased production of the “bad”  
350 or increased cost of measures or practices. Cost-benefit analysis of N can be applied to  
351 estimate the societal cost associated with N mitigation strategies (including the cost of

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

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

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

375

### 376 ***Scenario analysis and management***

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

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

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

$$394 \quad \text{NUE} = \frac{\text{N in products}}{\text{Total N input for production}}$$

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

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

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

$$409 \quad \text{Dietary pattern} = \frac{\text{Animal protein}}{\text{Total protein consumed}}$$

$$410 \quad \text{Food waste ratio} = \frac{\text{Food waste}}{\text{Food supply}}$$

411 where dietary pattern refers to the ratio of human protein consumption provided by  
 412 animal products such as meat and eggs. Total protein consumed includes both vegetal

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

417 To simulate these  $N_r$  uses and losses, shared social pathway (SSP) storylines  
418 present a useful approach to estimate the population, urbanization and per capita GDP in  
419 the future<sup>38</sup>. Many studies on the mitigation of N losses followed the scenarios of IPCC  
420 with regard to future projections of greenhouse gas emissions, although the behaviour of  
421 carbon is not always consistent with that of N<sup>47</sup>. Therefore, the new scenarios  
422 specifically designed with N in mind here are recommended for use in N assessments.

423 **Management.** Solutions for better managing  $N_r$  under different scenarios may not  
424 be adopted solely due to high cost, if benefits are not recognised or quantified<sup>24</sup>.  
425 Therefore the results from the cost-benefit analysis and scenarios analysis on the  
426 optimisation of N fluxes to achieve cost-effective solutions for a sustainable future need  
427 to be integrated. The costs and benefits of each scenario could be quantified to identify  
428 optimal solutions. Furthermore, whether these selected solutions for the sustainable use  
429 of  $N_r$  are feasible requires further analysis of the socioeconomic barriers<sup>27, 48</sup>. These  
430 barriers can be economic structure (e.g., farm size), population density, culture, religion,  
431 consumer or even dry climate. Thus, social scientists should be involved in assessing  
432 the feasibilities of the proposed solutions. Finally, several N sustainability indices can  
433 be used to translate the scientific results to the public and policy makers to make real  
434 impacts, such as the N footprint concepts<sup>49</sup>. N fluxes and their costs and benefits are  
435 incorporated into the indices to reflect the quality of products and their environmental  
436 effects.

437

### 438 **Implications of N assessment**

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



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

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

460

#### 461 *Application for China*

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

470 **Nitrogen budget.** Results showed that total  $N_r$  input to China increased from 25 to  
471 71 Tg N yr<sup>-1</sup> between 1980 and 2015, of which 74% and 89% derived from  
472 anthropogenic sources in 1980 and 2015, respectively (Figure 5). After input to the  
473 boundary of China,  $N_r$  cascades through the 14 subsystems, and produces about 20 Tg  
474 N yr<sup>-1</sup> in food and feed, while losses to the environment amount to around 50 Tg N yr<sup>-1</sup>  
475 in 2015. Agricultural sources are responsible for approximately 91% of total  $NH_3$   
476 emissions; fossil fuel combustion accounts for 90% of total  $NO_x$  emissions; agricultural



477 and natural sources (forest and surface water) together dominate N<sub>2</sub>O emissions in  
478 China. Agricultural sources and human sewage contribute most of the N<sub>r</sub> discharge to  
479 surface water, while agricultural sources and landfill leaching are responsible for the  
480 bulk of N<sub>r</sub> discharges to groundwater. More detailed information about the N fluxes in  
481 China can be found in Figure S1.

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

491 ***Cost-benefit analysis.*** Integrating N<sub>r</sub> emissions and population exposure  
492 assessments, we calculated that using 2015 N<sub>r</sub> fluxes, N<sub>r</sub> 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<sub>r</sub> 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<sub>r</sub> 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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696



697 **Figure Legend**

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

705

706 **Figure 2. Simplified view of managing N for sustainable development highlighting**  
707 **the three major scientific questions.** (Q1) How are N uses and losses affected by  
708 natural and human factors? (Q2) How to quantify the societal costs and benefits of N  
709 use? (Q3) What are the socioeconomic barriers constraining the more sustainable use of  
710 N?  $N_r$ , reactive N.

711

712 **Figure 3. Conceptual diagram depicting the linkage between the DPSIR scheme**  
713 **and the framework of N assessment in this study.** DPSIR, Driver–Pressure–State–  
714 Impact–Response; Q1-Q3 represent the three major scientific questions need to be  
715 addressed in Figure 2. (Q1) How are N uses and losses affected by natural and human  
716 factors? (Q2) How to quantify the societal costs and benefits of N use? (Q3) What are  
717 the socioeconomic barriers constraining the more sustainable use of N? NUE, N use  
718 efficiency.

719

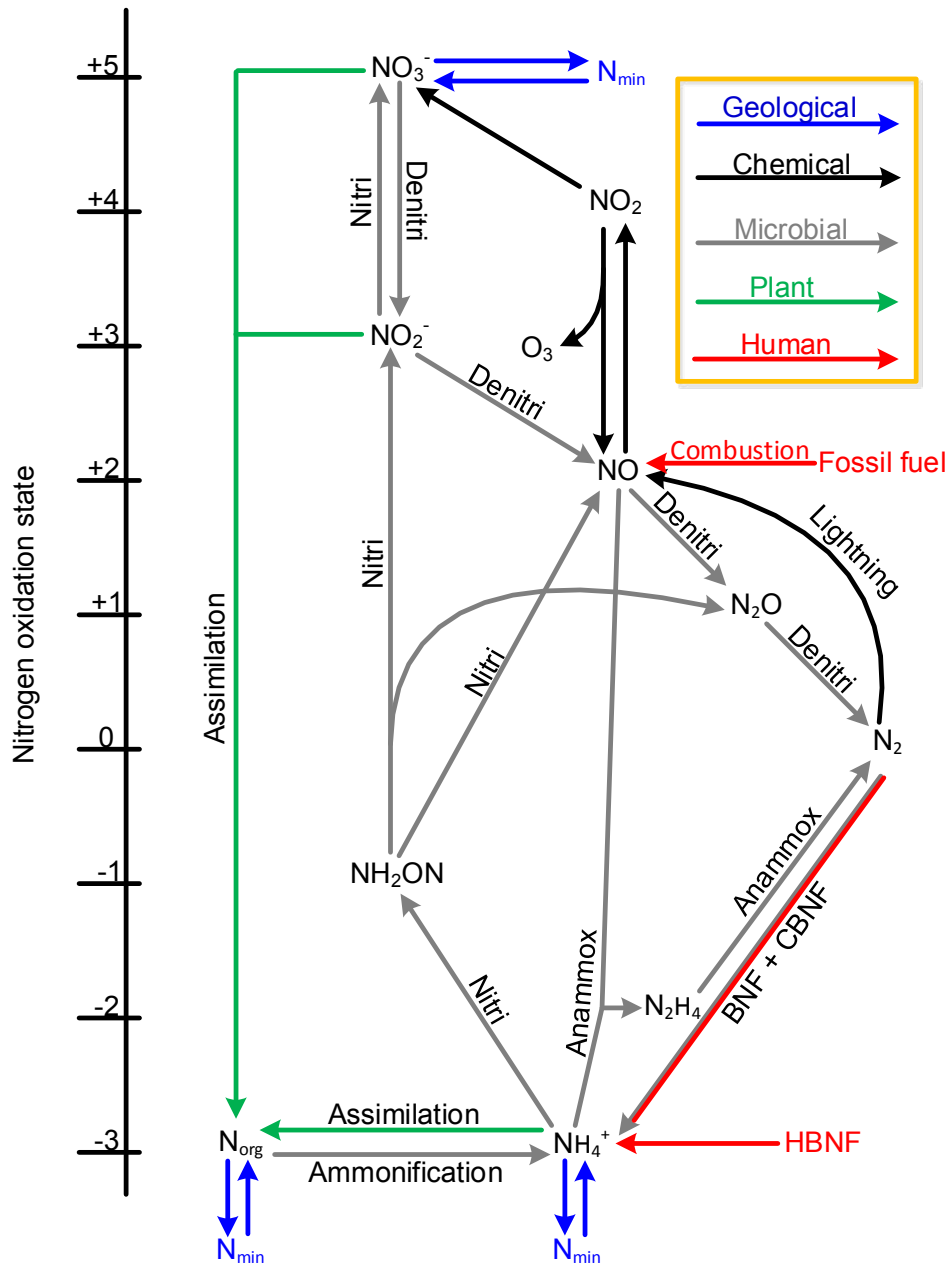
720 **Figure 4. An overview of the methodological framework.** It describes the approaches  
721 to mass balance modelling, validation, cost-benefit analysis and management for the  
722 sustainable future use of  $N_r$ . *BNF*, biological N fixation; *NDVI*, normalized differential  
723 vegetation index; *WTPs*, willingness to pay.

724

725 **Figure 5. A simplified N cycling schematic for China for the year 2015.** Pathways  
726 marked in green refer to ‘natural’ fluxes (to some extent altered by atmospheric  $N_r$   
727 deposition), those in blue are intentional anthropogenic fluxes, and those in orange are  
728 unintentional anthropogenic fluxes. Not all N fluxes were included due to space limits.

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

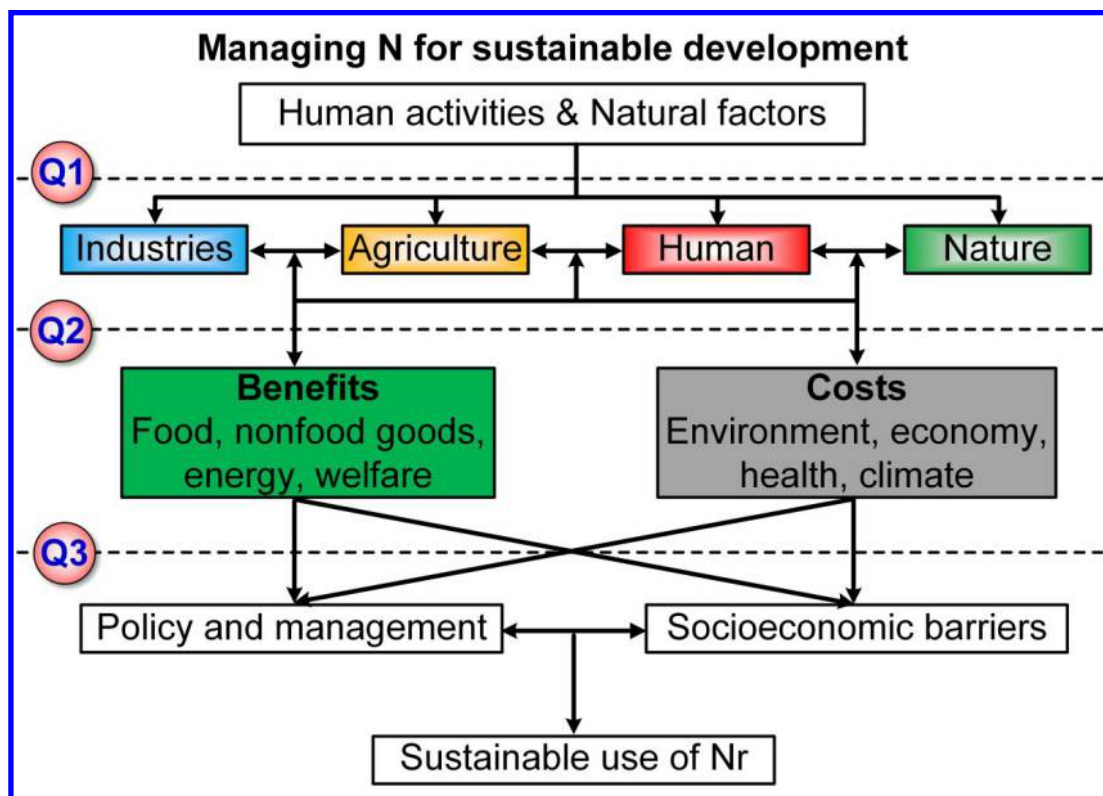
730 **Figure 1**



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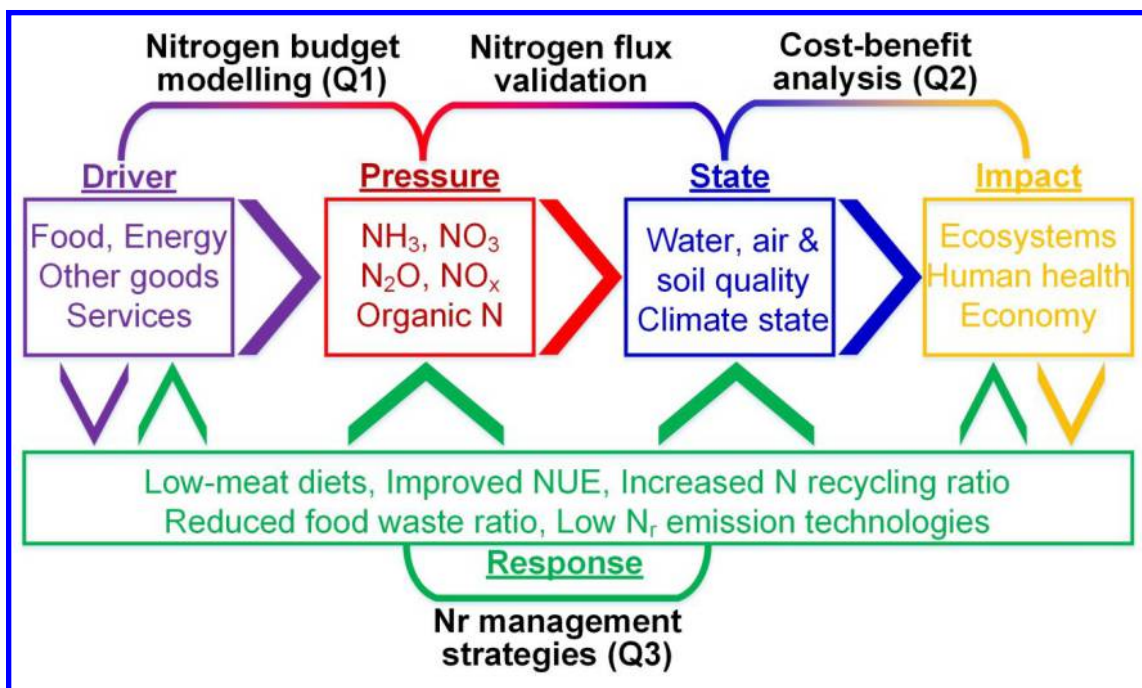
733 **Figure 2**



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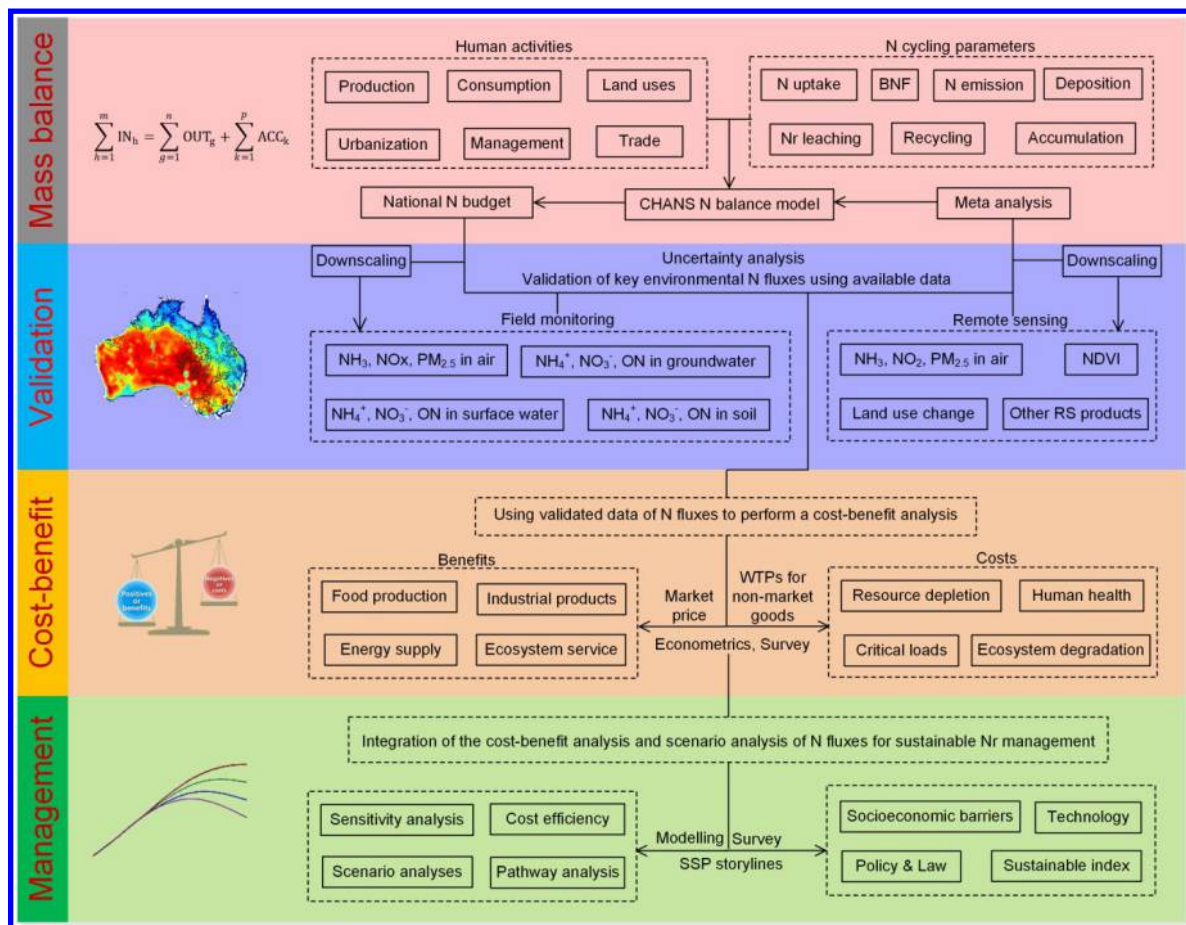
736 **Figure 3**



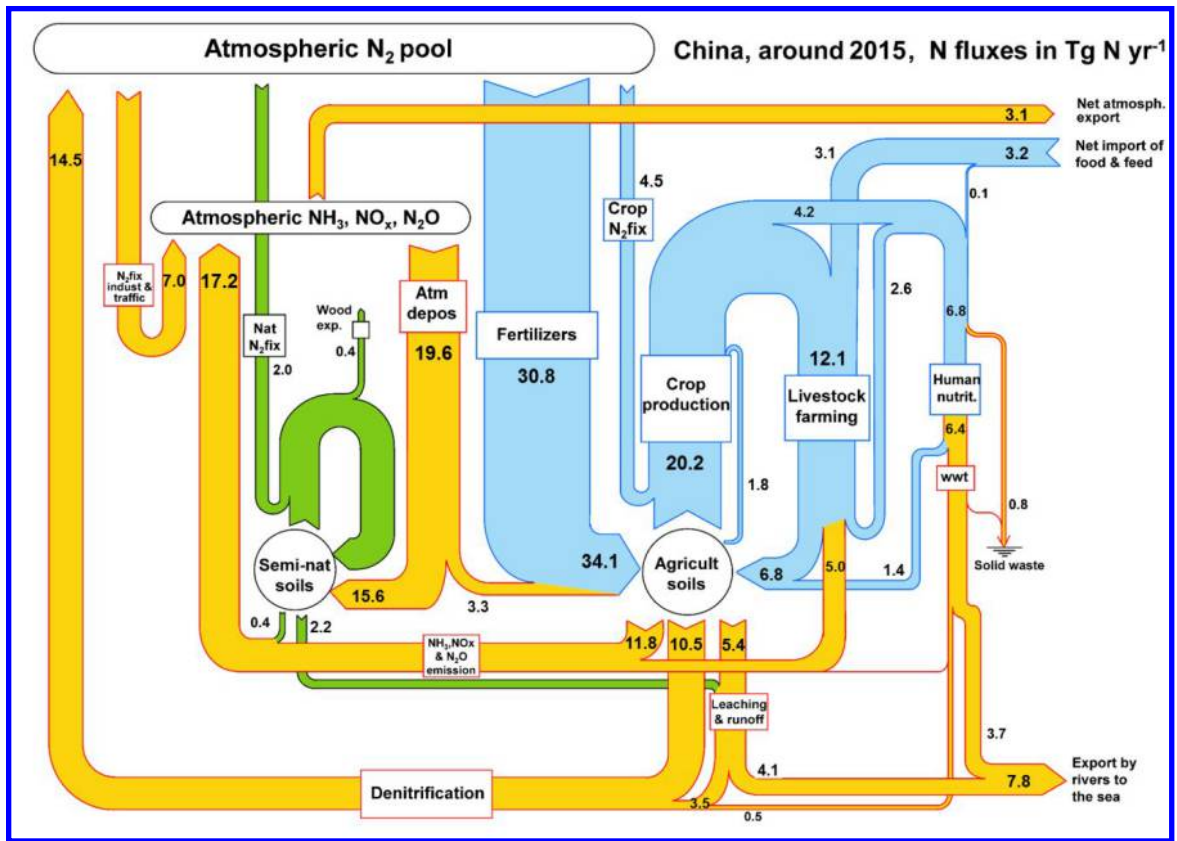
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740 **Figure 4**

741



742 **Figure 5**



743

