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RESEARCH ARTICLE
10.1029/2022EF002870

Special Section:
Modeling MultiSector Dynamics to Inform Adaptive Pathways

Tingyu Li and Xin Zhang contributed equally to this work.

Key Points:

- *C*ropping, *A*nimal-crop, *F*ood, and *E*cosystem framework encompasses the complex N dynamics across systems and spatial scales to guide policies
- Priorities and key stakeholders for enhanced N management vary among countries
- Systematic solutions that improve the coupling of different system components were revealed

Supporting Information:

Supporting Information may be found in the online version of this article.

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Citation:

Li, T., Zhang, X., Zhong, Y., Davidson, E. A., Dou, Z., Zhang, W., et al. (2022). A hierarchical framework for unpacking the nitrogen challenge. *Earth's Future*, *10*, e2022EF002870. https://doi.org/10.1029/2022EF002870

Received 29 APR 2022
Accepted 25 OCT 2022

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A Hierarchical Framework for Unpacking the Nitrogen Challenge

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Abstract To feed the world population while mitigating pressing nitrogen (N) pollution problems, tremendous efforts have been devoted to developing and implementing N-efficient technologies in crop or livestock production, but limited progress has been made. The N management improvement on a farm does not necessarily translate to N pollution reduction on a broader scale due to complex responses of natural and human systems and lack of coordination among stakeholders. Consequently, it is imperative to develop an N management framework that encompasses the complex N dynamics across systems and spatial scales, yet simple enough to guide policies and actions of various stakeholders. Here, we propose a new framework, *CAFE*, that defines four N management systems (*C*ropping, *A*nimal-crop, *F*ood, and *E*cosystem) in a hierarchical manner, and apply it to 13 representative countries to partition N surpluses across systems in a simple and consistent manner, thereby facilitating the identification and prioritization of systems-based intervention strategies. Surprisingly, the *C*ropping system contributes less than half of the total N surplus within its *E*cosystem for most countries, highlighting the importance of N management beyond croplands. This framework reveals that the relevant priorities and key stakeholders for enhanced N management vary among countries, such as improving the *C*ropping-system efficiencies in China, adjusting the animal-crop portfolio in the Netherlands, reducing food wastage in the U.S., and lowering crop storage losses and increasing overall production capacities in African countries. As N surplus increases along the *CAFE* hierarchy, systems-based intervention strategies are revealed: (a) coupling chemical fertilizers with other N sources by maintaining half of the N from manure and biological N fixation; (b) coupling animal-crop production by reducing animal density to lower than 1.2 livestock units per hectare, and increasing self-sufficiency of animal feed to above 50%; (c) coupling food trade with domestic demand and production; and (d) coupling population needs for economic opportunities with environmental capacity of the region. This novel framework can help unpack the "wicked" N management challenges across systems to provide new insights and tools for improving N management on and beyond farms.

Plain Language Summary To tackle the "wicked" nitrogen (N) challenge, N-efficient technologies in crop or livestock production alone are insufficient. A broader understanding of the complex dynamics and interactions across different N cycle systems and among various stakeholders is needed. We propose a new framework, *CAFE*, that defines four N management systems (*C*ropping system, *A*nimal-crop system, *F*ood system, and landscape *E*cosystem) in a hierarchical manner, and the quantitative relations represented by N surplus changes between adjacent *CAFE* systems are illustrated. Potential driving factors for N surplus increases across the *CAFE* systems are also identified, such as animal and population density, feed and food self-sufficiency, etc. This framework can be used to partition N surpluses across systems and spatial scales in a simple and consistent manner, thereby facilitating the identification and prioritization of systems-based intervention strategies. Applying *CAFE* to 13 representative countries reveals that the cropping system contributes to less than half of the total N surplus of its respective ecosystem for most countries, highlighting the importance of N management beyond croplands, and the relevant priorities and key stakeholders for

Funding acquisition: Xin Zhang, Weifeng Zhang
Methodology: Tingyu Li, Xin Zhang
Project Administration: Weifeng Zhang
Supervision: Weifeng Zhang
Validation: Tingyu Li, Yuxiu Zhong
Visualization: Tingyu Li, Yuxiu Zhong
Writing – original draft: Tingyu Li, Xin Zhang, Eric A. Davidson, Zhengxia Dou, Weifeng Zhang, Paulo S. Pavinato, Luiz A. Martinelli, David R. Kanter, Jianguo Liu, Fusuo Zhang
Writing – review & editing: Tingyu Li, Xin Zhang, Eric A. Davidson, Zhengxia Dou, Weifeng Zhang

enhanced N management vary among countries. Potential systems-based intervention strategies are proposed with new insights for achieving sustainable N management.

1. Introduction

Sustainably feeding the growing population amid climate change is a grand challenge in the 21st century (Godfray et al., 2010), and improving nitrogen (N) management is at the core of addressing this challenge. N is essential for growing food for humans and feed for livestock, the latter in turn producing nutrient-dense foods such as meat, milk, and eggs to serve a surging demand, especially in developing countries. However, anthropogenic N inputs to the biosphere have increased nearly fivefold in the last 60 years (Battye et al., 2017), exceeding the estimated "planetary boundary" that marks a safe operating space (Steffen et al., 2015). The same atom of reactive N (Nr) added to the biosphere can be transformed and transported across multiple systems (e.g., terrestrial ecosystems, freshwater and marine systems, and the atmosphere), and consequently cause multiple effects, ranging from eutrophication to biodiversity loss to air pollution and stratospheric ozone depletion and to climate change (Galloway et al., 2003; Liu et al., 2013; Reay et al., 2012; Yu et al., 2019). This sequence of effects, also known as the N cascade (Galloway et al., 2003), has aggravated N pollution and has becomes one of the most pervasive environmental issues. As the world strives to feed 10 billion people by 2050, what can be done differently from previous or existing approaches regarding N management in order to produce more food with less N pollution?

Efforts in managing N have mostly focused on enhancing management practices in the agricultural domain, emphasizing the improvement of N use efficiency (NUE) while reducing N losses (e.g., $NH_3/NO_3^-/NO_X/N_2O/N_2$). Examples of these approaches include education-extension programs that guide fertilizer use following "4R" principles (i.e., right rate, right time, right place, right source) (Li et al., 2019; Zhang, Mauzerall, et al., 2015); technological innovation with enhanced efficiency fertilizers that better synchronize N supply with dynamic N uptake or delay N transformation processes by using inhibitors (Akiyama et al., 2010; Li et al., 2018); and various policy interventions that cap fertilizer application rates (Li et al., 2020; Van Grinsven et al., 2012) or incentivize organic-based nutrient recycling (Chadwick et al., 2015). Recent progress has been made in developing new indicators and credit systems to manage N in a more systematic manner involving stakeholders beyond farmers (Gu et al., 2019, 2021; Leach et al., 2012; Liang et al., 2018, 2021). For example, N footprint analyses, which quantify N pollution associated with producing a food product, have been proposed as part of food labeling to inform consumer behavior (Leach et al., 2012).

Despite tremendous efforts devoted to N management, overall progress has been limited globally and N-related pollution problems still prevail in developed as well as developing countries (Billen et al., 2014; Reay et al., 2012; Steffen et al., 2015). This unsatisfactory progress could be attributed to three major challenges in N management:

1. N inefficiencies and N losses permeate throughout the food chain from farm to fork and beyond (e.g., waste management) (Erisman et al., 2018; Kanter et al., 2019), but current N management efforts have been mostly concentrated on improving on-farm practices (Kanter et al., 2019). Compared to the disappointing global average of 43% NUE for crop production, only 16% of the N added to agricultural production was consumed by human as food products (Zhang et al., 2020), highlighting the importance of N management beyond farms. However, few approaches are available to unpack the loss of efficiency, understand the N dynamics, identify the drivers for N loss along the supply chain, and enable coordinated N management across systems. The N budget approach, which quantifies the N inputs and outputs of a defined system, has offered promising options (Zhang et al., 2020), as it is relatively simple and transparent and provides indicators (e.g., N use efficiency and harvested N) that have been widely recognized by stakeholders. However, although the N budget approach has been applied to various systems and spatial scales (Billen et al., 2014; Erisman et al., 2018; Howarth et al., 2012; McLellan et al., 2018; Oenema, 2006), few have applied the approach consistently across different systems and scales to investigate the interconnections among systems (Zhang et al., 2020).
2. Most of the current policies and approaches focus on one or a few specific aspects of N challenges without systematically considering complex tradeoffs among various N management goals and across different spatial and system scales (Kanter, Chodos, et al., 2020). These fragmented and uncoordinated approaches may result in many unintended consequences. For example, over the past two decades, China encouraged the development of Concentrated Animal Feeding Operations (CAFO) to meet the growing domestic demand for animal-source foods. However, the increasing CAFO production has de-coupled the N cycle between

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crop and animal production, resulting in the discharge of manure N to streams as well as increasing chemical fertilizer application to croplands. Recent efforts have been devoted to regulating manure discharge and incentivizing manure recycling, however, due to the lack of guidance and coordinated efforts, environmental challenges remain, such as over application of manure in some regions and vulnerability of the food supply chain to shocks such as African Swine Fever. In addition to the tradeoffs among N management goals, the increasing reliance on feed import in China has led to tradeoffs among world regions. For example, the feed (primarily soy) import helps to relieve resource constraints and N pollution pressure in China, but it has also led to deforestation and N pollution in exporting countries such as Brazil and the U.S. (Huang et al., 2019). Identifying these complex tradeoffs in countries at different development stages and with different natural endowments is crucial for sustainable N management.

3. Managing N involves many stakeholders, including multiple economic sectors and governmental ministries, with differing or even competing interests across multiple spatial scales (Kanter, Chodos, et al., 2020). However, the responsibility for N management has been mostly borne by farmers with limited accountability from other stakeholders. In fact, stringent measures that impose N pollution mitigation targets squarely on farmers without properly addressing the financial health of farming operations are neither sustainable nor equitable, as implicated by farmer protests in Europe in recent years (Kanter et al., 2019; Oenema, 2004; Poore & Nemecek, 2018). The lack of support and cooperation from stakeholders other than farmers could be attributed in part to the absence of institutional innovation to establish platforms and opportunities for multi-agent participation, making it difficult for other stakeholders to participate effectively in N management. The scientific communities, together with policy-makers and the private sector, must go beyond the realm of agriculture to explore new pathways and find more comprehensive and broadly-reaching solutions (Kanter et al., 2019; Kanter, Chodos, et al., 2020).

To address these challenges in N management, we designed a hierarchical framework encompassing the complex cycles of N in human activities and natural systems and connecting the primary interest of various stakeholders along the supply chain, yet simple enough to guide policies and day-to-day actions: the *C*ropping system is nested within *A*nimal-crop system, which is nested within the *F*ood system, and finally nested within landscape *E*cosystem (*CAFE*). We demonstrate the utility of the framework by applying it on a national scale for 13 countries representing different agricultural, socioeconomic, and geopolitical conditions. These countries encompass different economic development stages (i.e., developed countries and developing countries) and account for 66% of world N fixation (synthetic fertilizer and crop N-fixation) (Zhang, Davidson, et al., 2015) and nearly 40% of cropland (Food and Agriculture Organization of the United Nations, 2010). With the support of the framework, we unpack the challenges and opportunities for integrated N management in these countries, and discuss policy implications for N management in these countries and the world.

2. A Novel Framework for a Wicked Problem

To unpack the "wicked problem" of N management, the new framework, *CAFE*, defines four N management systems in a hierarchical manner (Figure 1), of which the basic functions are meaningful for all stakeholders and for which data are often available (Table S1 in Supporting Information S3). The definition of each system is based on an extensive review of N budget approaches and other N management tools (Zhang et al., 2020). Each system is defined by virtual or physical boundaries and characterized by indicators such as N inputs, N outputs (in products), N surplus (inputs minus outputs), and NUE (Figure S1 and Table S1 in Supporting Information S3, detailed information for definitions see Section 8). Moving up along the *CAFE* system hierarchy, new N dynamics are included (Table S1 in Supporting Information S3). Improving N management for a given system relies not only on the improvement for subsystems, but also the N dynamics among subsystems (Figure 1). Stakeholders are connected to the *CAFE* framework and other stakeholders through the management indicators that are of their primary interest and through interconnections of those indicators across systems (Figure S2 in Supporting Information S3).

The *CAFE* framework is different from existing frameworks such as N footprint analyses (Leach et al., 2012) and the proposed analytical framework for national N assessments (Gu et al., 2019). While these three frameworks are all based on the quantification of various N flows, their scope and function are different. The N footprint approach focuses on N pollution associated with the whole supply chain of a given food product and does not usually distinguish where the pollution takes place. Therefore, the N footprint indicator is useful to guide food

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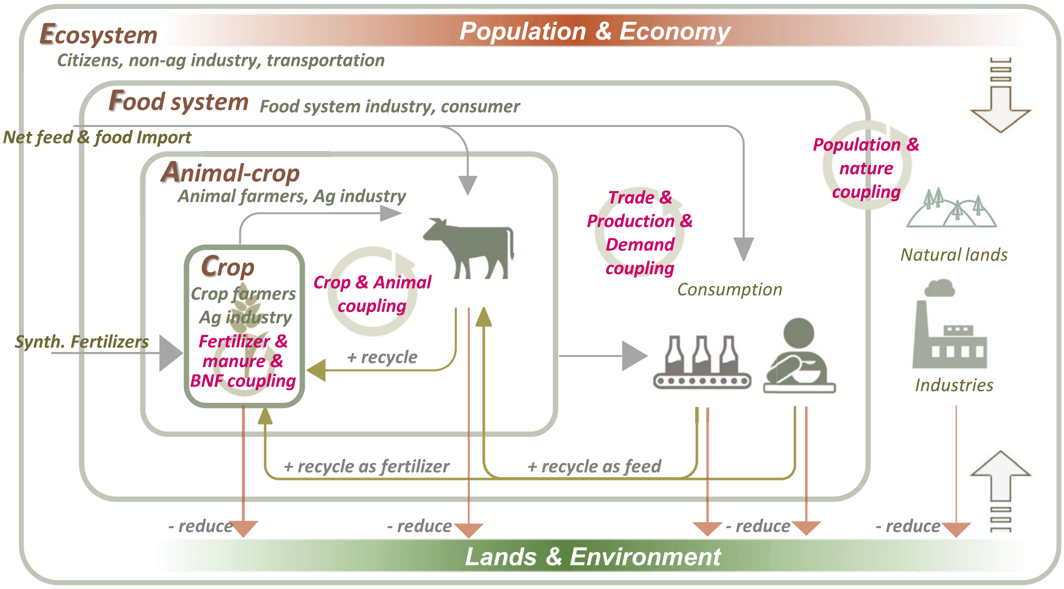

Figure 1. The *C*ropping, *A*nimal-crop, *F*ood, and *E*cosystem (*CAFE*) framework for regional N governance. The improvement in the bigger system relies on improvements in sub-systems as well as the new N dynamics involved in the bigger system. In this framework, N-rich food is the dominant target. For graphical simplicity, nonproductive products, such as air and water pollution, are not shown. Arrows represent N flows across different systems; the tawny arrows indicate N recycling processes that could be strengthened.

consumption toward low-pollution products in general, but it does not necessarily lead to reduction in N pollution for a particular region of interest. In contrast, the application of the *CAFE* framework requires a clearly defined spatial boundary, and the N surplus indicators measure the pollution stress added to the region by a given system (e.g., cropping system and food system). Given *CAFE*'s focus within a spatial boundary, importing food and feed could be considered as one of the strategies to alleviate the local N pollution stress within that bounded region.

In addition to the footprint approach, several models have been developed to provide detailed accounting of N flows in the agro-food system and to estimate reactive N loss (e.g., Billen et al., 2014; Gu et al., 2019). Based on one of the models, a framework was proposed to guide the development of management strategies for reactive N (Gu et al., 2019). While it is valuable to have a detailed accounting of N flows, especially reactive N loss in different forms, the application of this framework is limited by data availability and highly uncertain model parameterizations. The rather complex interconnections among the 14 subsystems identified by the model may also make it challenging for broader adoption by different countries and stakeholders. In contrast to the complex model and framework, *CAFE* identifies four systems that are of major concern for stakeholders and with minimal burden for data collection. Rather than focusing on various forms of reactive N losses, *CAFE* focuses on major N inputs and productive N output for each of four systems, and aims to assess the efficiency of these systems and the loss of efficiencies across systems.

CAFE can be applied at any operational or spatial scale, be it a farm, a community, a nation or multiple nations, or the globe, as long as the spatial boundary is clearly defined. As the first application of such a framework, we focus on demonstrating its utility at the national scale, which is of particular interest for policy makers and other stakeholders beyond farms.

3. N Surplus at the Ecosystem Level and Contribution of Subsystems

Applying the *CAFE* framework to developed and developing countries, totaling 13 countries around the world using the data from 2000 to 2010 (Data are shown as arithmetic average of this period, Figure 2, Figure S3 in Supporting Information S3), we found that N surplus at the Ecosystem level varies a great deal, from 23 kg N ha^{-1} in Uganda to 383 kg N ha^{-1} in China. The positive N surplus for Ecosystem across countries indicates that human activities have inevitably added pressure to the ecosystem, despite the development stage or geographic

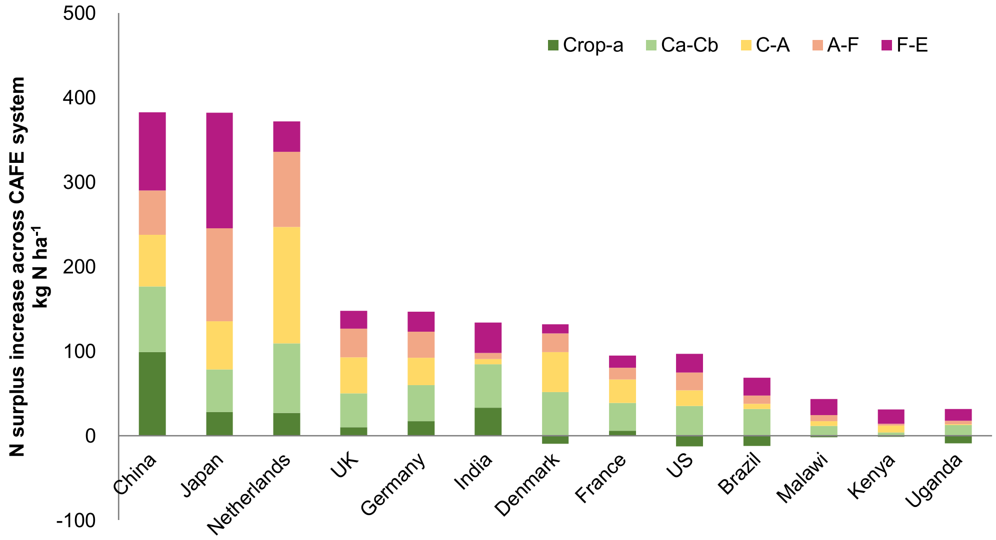

Figure 2. N surplus per agricultural area in a hierarchical Cropping, Animal-crop, Food, and Ecosystem (*CAFE*) N governance framework, using 13 countries as examples (averaged for the period of 2000–2010). Each bar denotes N surplus of the total Ecosystem for a given country divided by agricultural area (including cropland and managed pasture), with the different colors representing N surplus increase between adjacent systems. Crop-a means the Cropping system only considering mineral fertilizer as input, while Crop-b considers all N sources besides mineral fertilizers, as well as manure, biological N fixation, atmospheric N deposition. C-A, A-F, and F-E indicate the Cropping system to Animal-Crop system, Animal-Crop system to Food system, and Food system to landscape Ecosystem, respectively.

location of a country. But the level of pressure, indicated by the N surplus for Ecosystem, varies among countries: some countries (e.g., China and Japan) have exceeded the planetary boundary by a factor of 4 or 5, considering 80 kg N ha^{-1} as the planetary boundary for N surplus level based on previous study (EU Nitrogen Expert Panel, 2015; Zhang, Davidson, et al., 2015). In contrast, other countries (e.g., Kenya and Uganda) may still be able to increase their N use intensity without irreversibly degrading the environment. This N surplus in Ecosystem does not have a significant relationship with a country's income level (measured by gross domestic product (GDP) per capita), but is positively related to population density (Figure 3a, $p < 0.01$). The N surplus assessment at the Ecosystem level identifies priority regions for reducing N pollution pressure.

Surprisingly, the Cropping system level contributes less than half of the N surplus for most countries (Figure 2). This highlights the importance of N management beyond croplands. Across the 13 countries, N surplus within Cropping system follows the classic Environmental Kuznets Curve (EKC) as income level increases (Zhang, Davidson, et al., 2015) (Figure 3b), suggesting that it increases with income growth at early stages to boost crop yield and then levels off or even decreases as income increases further. Beyond the Cropping system (i.e., the difference in N surplus between Ecosystem and Cropping system), however, N surplus increases linearly with GDP (Figure 3c), indicating the lack of regulations on N losses from activities beyond croplands even in developed countries. In addition, the increase in income level is often associated with more animal products and diversified food demands, as well as higher volume of imported feed and food (new N input beyond the Cropping system), all potentially leading to increased N surplus beyond the Cropping system. Overall, even though many developed countries have made remarkable achievements in improving cropland management, N surplus beyond the cropping system becomes more prominent. It is critical for countries, especially developed countries, to develop strategies (e.g., policies and technologies) for decoupling the linear relationship between income and beyond-farm N surplus and demonstrating the possibility of achieving EKC for N surplus beyond the farm gate.

4. N Dynamics Across the System Hierarchy

Moving up the *CAFE* hierarchy, N surplus increased by 72% (7%–341%; median, min and max of all 13 countries) from the Cropping to the Animal-crop system, 126% (12%–395%) to the Food system, and 194% (58%–1,130%) to the Ecosystem, respectively (Figure 2). Conversely, NUE decreases from 52% (31%–71%) in Cropping system to 40% (24%–67%) and 29% (11%–51%) in Animal-crop system and Food system, respectively (Figure S4 in

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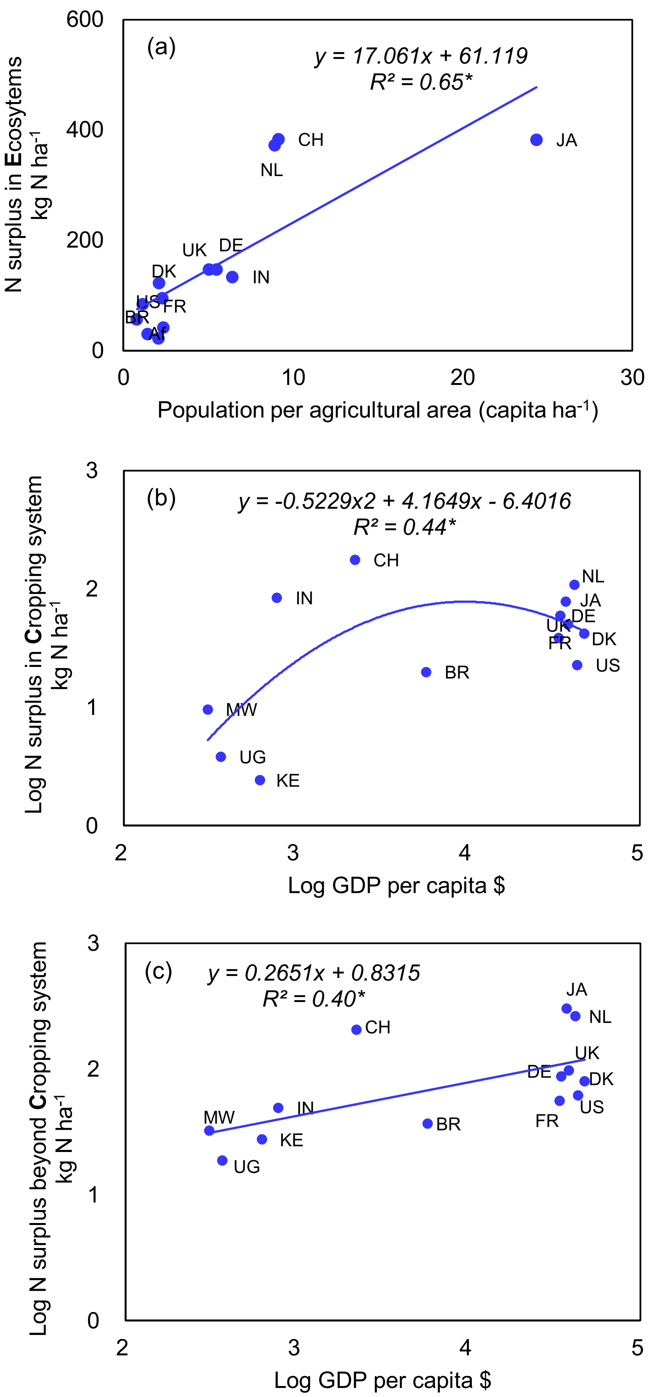

Figure 3. The influence of socio-economic factors on N surplus across the Cropping, Animal-crop, Food, and Ecosystem (*CAFE*) system. (a) The population density with total N surplus at the Ecosystem level, (b) the gross domestic product (GDP) per capita with N surplus in the Cropping system, and (c) the GDP per capita with N surplus beyond the Cropping system. Names of countries abbreviated as: BR = Brazil, CH = China, DE = Germany, DK = Denmark, FR = France, IN = India, JA = Japan, KE = Kenya, NL = Netherlands, MW = Malawi, UG = Uganda, UK = United Kingdom, US = United States of America.

Supporting Information S3). In other words, N surplus increases while NUE substantially decreases when moving across the hierarchy, from agricultural production through consumer food production. However, the degree of increase at each system level is very different among countries, indicating priority areas for improving N management for each country. Consequently, we examined N surplus in the Cropping system and the changes between systems along the system hierarchy (Figures 4 and 5, Figure S7 in Supporting Information S3), and identified major N management gaps in each country, as well as their potential causes.

The Cropping system still presents fundamental challenges for N management (especially in developing countries). These challenges are often in forms of two extremes: some fast-developing countries, such as China and India, have N surplus for Cropping system at the rate of 84–176 kg N ha^{-1}, with low NUE around 30%, whereas many others, such as Kenya and Uganda, have N surplus close to 0 or even negative, indicating likely mining of soil N and potential reduction in the already-low yield. In contrast, all of the developed countries examined in this study have made progress in reducing their N surplus and increasing their NUE, while also increasing or maintaining high crop yield. Such achievements have mainly benefited from implementing regulations for fertilizer application (including manure; e.g., Denmark, France, and Germany) or voluntary adoption of N-efficient and low-emission technologies (e.g., U.S.; see Table S3 in Supporting Information S3 for more examples). But considering all major N input sources such as manure, biological N fixation (BNF), and atmospheric deposition in addition to chemical fertilizer, the N surplus for the Cropping system (Crop-b as defined in Figure 2) in some developed countries (e.g., The Netherlands, 109 kg N ha^{-1}) still needs to be reduced to curb the N surplus within the planetary boundary (80 kg ha^{-1}).

Among all countries examined in the study, N surplus for the Cropping system is closely related to the sources (or makeup) of N inputs. Increasing manure and BNF (as a proportion of total N input) is associated with lower N surplus and higher NUE; the beneficial impact levels off when manure and BNF inputs reach about 50% (Figure 4). This is an important insight, highlighting the critical importance of increasing manure and BNF as the sources of N inputs in crop production. Crop rotation is one of the approaches for balancing different sources of N inputs, and has been widely adopted in countries such as Denmark, France, and the U.S. where higher NUE values have been achieved, that is, >60% as well as lower N surplus (Table S3 in Supporting Information S3). The diverse multispecies systems, including annual and perennial crops, help to improve N uptake by crops from multiple sources and bring about many co-benefits, such as higher overall productivity and nutrient retention, better control of pests and diseases, greater biodiversity, enhanced soil health, and higher productivity (Malézieux et al., 2009).

Moving from the Cropping system to the Animal-crop system, N surplus increased by 35 kg N ha^{-1} on average for the 13 countries (52 kg N ha^{-1} for developed countries and 15 kg N ha^{-1} for developing economies), mainly due to N loss generated from livestock housing and manure management processes. The Netherlands presented the highest increase in N surplus, even though the NUE for livestock system (38%) and area-based recycled N from manure (74 kg N ha^{-1}) are among the highest (Figure 5). Such apparent paradox of high efficiency and high N surplus is also evident in many other countries (e.g., Denmark), and is mainly driven by increasing livestock density (measures the stock of animals, e.g., cattle, pigs, poultry, converted

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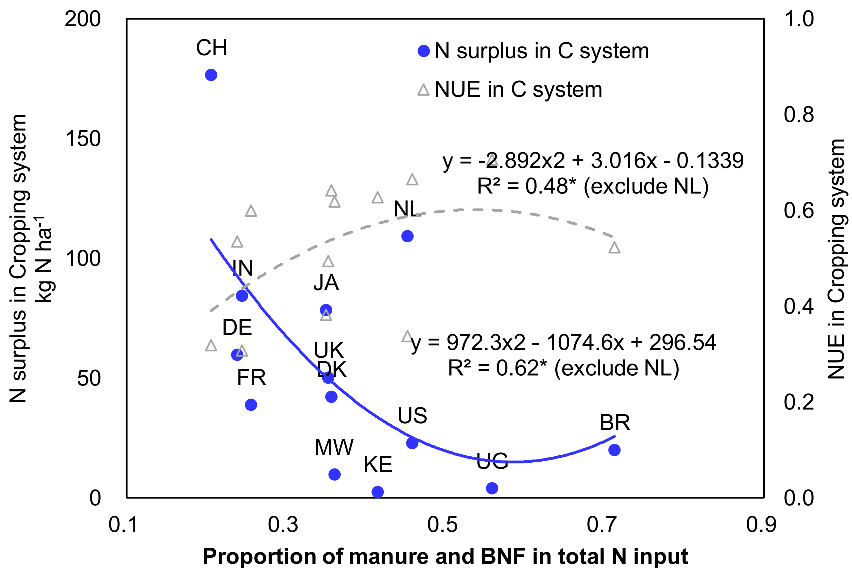

Figure 4. The relationship between N sources (or makeup, percentage of manure and biological N fixation (BNF) in total N input) and N surplus and N use efficiency (NUE) in the Cropping system.

in livestock units (LU) per hectare of managed agricultural area using specific coefficients) (Eurostat, 2009; Lesschen et al., 2011), which reflects the decoupling of crop and animal systems. In fact, the N surplus increase from Cropping system to Animal-crop system has a strong positive relationship with livestock density among countries (Figure 5). In contrast to Cropping system, where the productivity is still constrained by the physical limits of photosynthesis per unit of land surface, intensification of livestock systems has relaxed the constraints of land on its productivity by employing CAFOs and importing feed across regional or national boundaries. While being effective in boosting economic return per unit of land for the livestock producers, the increase of these quasi-landless livestock production systems has driven the production of animal and feed further apart, decoupled the N cycles between livestock and cropland (or pasture land), and left almost no physical limitation for productivity, as well as N inputs in forms of feed, per unit of land surface. For example, the livestock productivity in The Netherlands is as high as 102 kg N ha^{-1}, which is almost two times of crop N yield, and 90% of the feed is imported (Figures S3b and S5 in Supporting Information S3). Among all countries examined in this study, lower

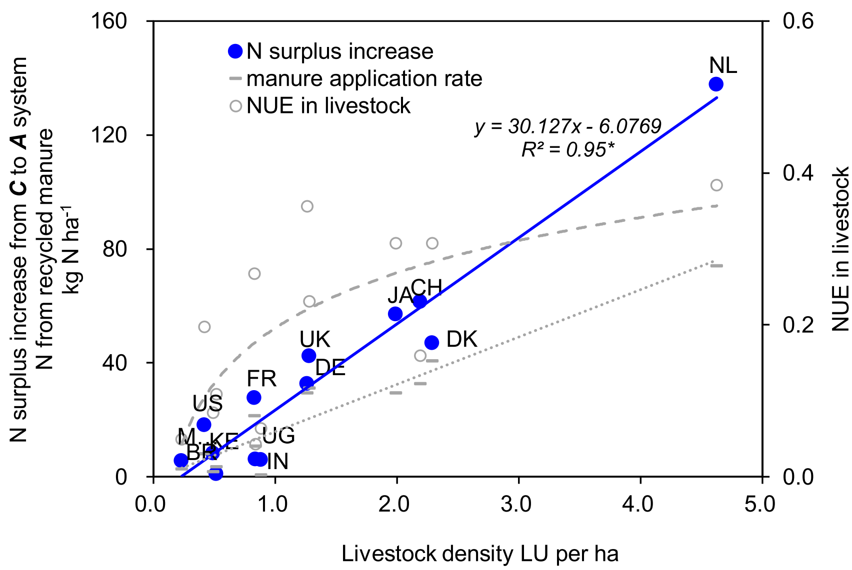

Figure 5. The relationships between livestock density and the N surplus increase from the Cropping system to the Animal-Crop system (left axis) and N use efficiency (NUE) in livestock systems (right axis).

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self-sufficiency in animal feed is associated with higher livestock density and higher proportion of domestic animal product as of total food product (Figures S5 and S6 in Supporting Information S3).

Improving NUE in Animal systems and recycling more manure from livestock to Cropping systems can relieve the N surplus pressure. For example, with similar livestock density, the N surplus increase from C to A in Denmark is 14 kg N ha^{-1} lower than China, given the higher NUE and recycled manure (Figure 5). In fact, the manure management is stricter and more effective in Denmark and it is the only country that requires manure to be applied mechanically (Table S4 in Supporting Information S3), which greatly increased N fertilizer equivalence of manure to 60%–75%, compared to only 20%–50% in the UK (Van Grinsven et al., 2012).

However, the relief brought by improvement in NUE and manure recycling has been limited so far. For example, even with the manure management being most strict and effective among countries, Denmark still has N surplus around 89 kg N ha^{-1}, higher than the planetary boundary, and further reduction is needed for relieving the regional environment burden. The NUE for livestock systems increases as livestock density increases, but levels out at around 40%, indicating a potential limit for improvement with existing technologies and management practices (Figure 5). Similarly, the highest manure application rate in cropland is 74 kg N ha^{-1} in The Netherlands, indicating a potential limit for recycling manure back to adjacent croplands. The distance between livestock and crop production adds technological and financial challenges for increasing manure recycling. Consequently, providing guidance for or regulating livestock density is needed for reducing N surplus generated in livestock. Based on our analysis, when the livestock density is below 1.2 LU per ha, the total N surplus in A-C system is mostly less than 80 kg N ha^{-1} (the estimated planetary boundary) and the feed self-sufficiency is higher than 50% (Figure S6 in Supporting Information S3). Currently, only Denmark has implemented regulations on livestock density at the farm level (Table S4 in Supporting Information S3), but the allowable level is still much higher than 1.2 LU per ha (1.7 LU/ha for cattle and 1.4 LU/ha for pigs) (Ministry of Environment and Food of Denmark, 2017).

Moving from the Animal-crop system to the Food system, N surplus increased by 31 kg N ha^{-1} on average (42 kg N ha^{-1} for developed countries and 12 kg N ha^{-1} for developing economies), mainly due to food loss and waste along the food supply chain as well as overconsumption of food. We define the food demand as the N needed for universal healthy diets, calculated as the population multiplied by the recommended per capita dietary intake (see Section 8). Overconsumption of N, which is the intake beyond the dietary requirement, is therefore included in the calculated N surplus of the food system, and can be influenced by dietary choices. Most of the increased surplus goes to incineration, landfill, or sewage, and threatens the environment, with very limited recovered food recycled for use as compost or animal feed (Galloway et al., 2014; Griffin et al., 2009). Taking the U.S. as an example, while the N surplus increase per hectare from the Animal-crop to the Food system is quite low due to large agricultural areas, the N from domestic food supply is more than seven times the human dietary need (3 kg N per capita per year (de Vries et al., 2013; Galloway et al., 2014)) and the difference is as high as 18 kg N per capita per year. While part of the surplus N is in the non-edible portion of food, some of which is used as animal feed or in non-food end uses (e.g., 25% of maize goes to ethanol industry, which is equivalent to 3.6 kg N per capita per year), most (70%–80%, 14 kg N per capita per year) is embedded in food waste generated in consumer-facing businesses (e.g., supermarkets, restaurants) as well as homes in developed countries. As a result, the NUE in the Food system in the U.S. decreased by 51% compared to the relatively high NUE in Animal-crop system, with only 24% of the N invested in food production eventually providing for the human demand. For many Americans, food represents only a small portion of family budgets (e.g., 6.6% in U.S. compared to 26.9% in China, USDA, 2012 data), the financial cost of wasting food is too low to outweigh the convenience, and the culture of high consumption behavior leads to food waste (Gunders, 2012).

The N surplus increases from the Animal-crop system to the Food system is associated with population density normalized by agricultural area (Figure S7 in Supporting Information S3). Japan shows the highest N surplus increase (by 98 kg N ha^{-1}) with the population density at 25 people ha^{-1} agricultural land (including crop and pasture land). Tremendous efforts have been made in balancing food supply with demand in Japan, recycling byproducts in commercial stages at the rate of 80% (mostly as animal feed), and enacting mandatory regulations such as the Food Waste Recycling Law in 2001 (Liu et al., 2016) (Table S5 in Supporting Information S3). Those efforts have successfully brought the N surplus increase down to the level close to that of The Netherlands (85 kg N ha^{-1}), which has a population density around 10 people ha^{-1} agricultural land, but further potential for improvement in recycling rates in Japan is very limited due to declining agricultural production and limited agricultural land (Liu et al., 2016).

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From the Food system to Ecosystem, N surplus increased by 38 kg N ha^{-1} on average (41 kg N ha^{-1} for developed countries and 35 kg N ha^{-1} for the developing economies), mainly contributed by consumed N in food and other agricultural products, as well as emissions from non-agricultural activities (e.g., industry and transportation). Population density (people per hectare agricultural land) drives the generation of household waste/sewage that ends up in the ecosystem, which is strongly associated with the N surplus increase from Food system to Ecosystem (Figure S7 in Supporting Information S3). The densely inhabited nations, such as The Netherlands, Japan, and China, showed similarly high N surplus per agricultural area (around 380 kg N ha^{-1}) at the Ecosystem level, creating a significant environmental burden. Meanwhile, deposition of N from the atmosphere to the natural land including water bodies, largely from the non-agricultural activities, is another component of the N surplus increase from Food system to Ecosystem, and it accounts for about half of the increase. The deposition rate is highly related to the development stage. For China and India, with their fast-developing pace and less regulation of air pollution, the N deposition rates exceeded 15 kg N ha^{-1} (Liu et al., 2013; Vet et al., 2014), which is 2–3 times of that in the EU and the U.S. where more strict regulations to control NOx and NH_3 emission and deposition exist, such as the "National emission ceiling" (Table S6 in Supporting Information S3).

Overall, population density plays a major role in determining the N surplus pressure on the environment (measured by N surplus for the Ecosystem), but the dominating contributors of N surplus increase vary among countries, and efforts in improving N recycling and system efficiencies can potentially relieve part of the pressure. The main contributor to the high surplus is the Cropping system in China, the increase from Cropping system to Animal-crop system in The Netherlands, and the N surplus beyond Animal-crop system in the U.S. and Japan, indicating different priorities for mitigation.

5. Managing N Surplus at Each Level of the *CAFE* Hierarchy

To address N management priorities identified by the *CAFE* framework in each nation or community, actions from key stakeholders, such as farmers and consumers (noted for each system in Figure 1), are essential. Meanwhile, leadership from local and national governments, industry, academia, and NGOs is also indispensable. Therefore, for each system in the *CAFE* framework, we provide a partial list of examples of policies and actions that could improve system performance while contributing to the overall sustainability of N management collectively (Table 1) and examine the application of these measures in selected countries (Tables S3–S6 in Supporting Information S3). We also consider how actions taken in one system could have ripple effects throughout other systems in the *CAFE* framework.

5.1. Cropping System

The ultimate aim of any measure or policy in this system is to better synchronize soil N supply with plant N demand. One of the most widely proposed policy ideas is to enable and incentivize the development and adoption of best management practices (BMPs) that improve productivities and NUE for crop production (Gu et al., 2021; Zhang, Mauzerall, et al., 2015). Potential policy interventions include designing monetary incentives or regulatory measures to encourage farmers to use chemical fertilizer more prudently and to recycle nutrients from animal and household wastes. Examples of regulatory measures include mandatory restrictions on when and where fertilizers may be applied, using N quotas and promoting rotation and cover crops via mandatory and voluntary programs. Investments are needed not only in research and technology development (e.g., establishing regional trial networks (Yu et al., 2019)), but also in socioeconomic studies on the barriers for BMPs adoption, as well as social services for farmers (e.g., keeping records of commercial farms and cooperatives, and providing soil analysis). The active involvement by the private sector including the fertilizer industry is essential for providing innovative products and improving quality of services especially in developing countries. Consequently, policies that focus on a smaller number of non-farmer actors in the food system who can influence farm-level N management behavior, from multinational corporations to city food procurement offices, may be more effective and easier to implement (Kanter et al., 2019; Kanter & Searchinger, 2018).

5.2. Animal-Crop System

The aim of measures and policies in this system is to reverse the global trend of increasingly separated crop and animal production systems. For example, integrating animal and crop production systems could encourage

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Table 1
A List of Practical and Promising Measures Across CAFE Systems on Improving Nitrogen Management

CAFE	Practical and promising measures					
Cropping system	Keeping records of farms	Soil analysis	Regulated fertilization periods/areas	Low emission application[a]	N application quota	Rotation and catch crops
Animal-crop system	Max. animal stock density	Regulated housing facilities[b]	Low protein feeding	Regulated manure storage capacity[c]	Regulated manure discharge	Manure application recommendation[d]
Food system	Post-harvest facility investment[e]	Food date label simplification[f]	Policies for supporting food donation[g]	Consumer education[h]	Policies for using pre-consumer food waste for feed	Policies for supporting post-consumer food waste for composting
Landscape Ecosystem	Water quality monitoring network[i]	Nitrate or ammonia vulnerable areas define, and activities restricted	Policies for wetlands and riparian protection[j]	Point discharge control system[k]	Policies for supporting sewage sludge for fertilizers	National NO_x emission ceiling[l]

[a]Low emission application indicates "4R" fertilization management principles, recommendation on rate, source, application methods, and timing. [b]Regulated housing facilities indicate requirements for reducing nitrate leaching and ammonia emission, such as floor materials. [c]Regulated manure storage capacity indicates requirements of construction materials of manure storage vessels to ensure groundwater and surface water is not polluted and being long-lasting. [d]Manure application recommendation indicates nutrient test to recommend appropriate rate and low emission application method such as injection, and other treatments to reduce loss. [e]Post-harvest facilities investment such as construction of warehouse and/or a concrete drying pavement to improve the quality and value of grains of small farmers and reduce food loss during post-harvest. [f]Food date label simplification means implementing a standard date labeling which carry simpler, easier-to-understand quality and safety information, leading to fewer items being thrown in the trash by consumers. [g]Policies for supporting food donation, providing liability protection for both food donors and the nonprofits accepting donations. [h]Consumer education on what they can do to reduce the food waste such as buying less during a weekly food shop and less meat dietary. [i]Water quality monitoring network comprises all sampling activities to collect and process data on water quality for the purpose of obtaining information about the physical, biological, and chemical properties of water. [j]Wetlands and riparian protection includes actions like construction of wetland treatment cells, reforestation and stream bank and wetland restoration. [k]Point discharge control system aim to control any pollutant to navigable waters from any point source, such as regulatory of wastewater discharge, permitting to meet a total maximum daily load. [l]National NO_x emission ceiling indicates regulations to control the total NO_x emission volume from fossil fuel source, such as vehicles and power plants.

effective recycling of manure and innovative manure management alternatives, in addition to optimizing the whole-farm production portfolio and spatial allocation of crops and livestock, and enhancing livestock nutrition and dietary efficiencies (De Klein et al., 2017; Snyder et al., 2014). To enable such integration, innovative policies and incentives are needed to foster the development of advanced manure treatment technologies that can recover and concentrate N for long-distance transport and more efficient usage, such as limiting the animal density, strengthening the regulation of manure discharge, and supporting the development of manure treatment industry. Moreover, the improvement of NUE in Animal-crop systems can be achieved by promoting low protein feeding, forbidden manure discharge, providing manure application recommendation services to recommend appropriate rates and low emission application methods such as injection, and other treatments to reduce loss such as composting and acidification (Hou et al., 2015). But, as demonstrated in our results, a focus on improving NUE is not sufficient and may lead to the "high-efficiency-high-surplus" paradox, especially for countries with high population density and intensive livestock production. Consequently, policies that mandate or highly incentivize maximum stocking density, improved housing facilities, and manure storage capacity are potentially necessary to ensure positive outcomes for both agricultural productivity and the environment.

5.3. Food System

The aim of policies and measures in this system is to direct the shift toward healthy and less N-intensive diets and to minimize food loss and waste from farm to fork. Reducing food loss and waste at the source and re-purposing food materials that are undesirable for humans to use as animal feed via treatment technologies (Dou et al., 2018) should be top priorities. All countries are facing the challenges of reducing food loss and waste, but the focuses for improvement are different (Aschemann-Witzel, 2016): infrastructure for food storage and transportation needs to be improved in some developing countries, whereas consumer behavior interventions are crucial in developed countries and fast developing economies (such as China and Brazil). Such interventions require collective efforts from both private and public sectors. Private sector commodity suppliers and retailers can link consumer demand for sustainably-produced food to supply chains and farming practices through product labeling (e.g., N footprint labels) and promotion, and through requirements placed upon suppliers (Leach et al., 2012). Improving the clarity of the sell-by date guidelines can also reduce unnecessary discarding of edible food (Kanter et al., 2019). Meanwhile, public education on reducing food waste and healthy eating through information campaigns (e.g., promoting SDG 12.3, which calls for halving global per capita food waste at retail and consumer level by 2030) and practical actions (e.g., changing the size or type of servings in catering activities in public schools) is critical for engaging consumers toward sustainable food behaviors. This education is especially important for school children for long-lasting impacts (Reynolds et al., 2019). Policies fostering food donations are also helpful for preventing pre-consumer food from being wasted (Van Zuiden, 2012). Other technical and political solutions to help re-purpose pre-consumer food discards via animal feeding and post-consumer food waste for fertilizers should also be involved (Dou et al., 2018).

International trade for food or feed is also an important component in the N equation (Xu et al., 2019), affecting N surplus in both the exporting and importing regions (Huang et al., 2019; Sun et al., 2018). Consequently, decisions on food and feed trade should consider the effect of embedded N on the domestic N cycle. A balance of N between food/feed import and domestic production and consumption needs to be achieved to avoid oversupply and waste. Such policies may include border adjustment taxes to account for surplus N in imported food products, as proposed for carbon in the European Union (EU Green Deal, 2020).

5.4. Ecosystem

This landscape level of the *CAFE* framework bears the consequences of the inefficiencies at all other levels and as such requires a broader policy approach that includes looking beyond the agricultural sector to other N polluting sectors, including industry, transportation, and wastewater treatment. The ultimate aim in this system is to balance agricultural and industrial productivity with Ecosystem functions at landscape scales, and reduce the negative impact of N surplus on human and ecosystem health. Potential measures include treating and potentially recycling human sewage, using natural or constructed wetlands and riparian vegetation as nutrient filters receiving farm runoff (McLellan et al., 2015), regulating reactive N emissions (e.g., $NO_{x,}$ N_2O) from industry and sewage treatment plants (Eshleman et al., 2013), and limiting agricultural activities in protected areas. Despite the potential for improvements in the C, A, and F systems, human activities, including agricultural and industrial production, inevitably add new reactive N to the Ecosystem and cause disturbance to the natural environment and N cycles. To facilitate and inform decision-making, a monitoring network of the use and impacts of reactive N needs to be established, and *CAFE* provides a framework for designing such network systematically.

6. Institutional Innovation for Managing N With the CAFE Framework

Taken together, this selection of policies and actions reflects the necessity of systemic solutions that improve the coupling between different system components under the framework of metacoupling (human-environment interactions within as well as between adjacent and distant systems) (Liu, 2017). They include coupling chemical fertilizer with other N sources like manure and BNF, coupling animal production with crop production to reduce animal density and increase manure recycling, coupling domestic demand and production with food importation, and coupling human activities with environmental capacity (Figure 1). Consequently, the development and implementation of these policies and actions require more coordination and collaboration across traditionally defined boundaries among disciplines, sectors, government agencies, and other stakeholders.

6.1. Jointing Efforts Cross Sectoral, Institutional and Disciplinary Boundaries

Interdisciplinary scholarship underlies the solutions to practical challenge of environmental impact and societal welfare (Chapin, 2017). Fragmented, discipline-specific research is not well-equipped to address the environmental problems embedded in the whole complex agro-ecosystem. Integrated research programs are greatly needed and can be conducted at various spatially targeted areas, such as watersheds, to address regional or national N pollution by engaging relevant stakeholders and making plans (Chopra et al., 2005; Reimold, 1998; Sims et al., 1999). In the U.S., there are many specific watershed scale projects such as the Gulf of Mexico, Chesapeake Bay, and Great Lakes Programs (Russo et al., 2008). These projects usually adopt a management structure, such

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as stakeholder involvement, geographic management units, coordinated management activities, or management schedules. These projects are good examples for integrating natural and social sciences, identifying individual and collective responsibilities among different stakeholders, and playing significant roles on improving watershed ecological functions, particularly water quality (Russo et al., 2008). Furthermore, the types of N-relevant research questions that social scientists can ask and address are very different from the natural sciences (e.g., how are cultural attitudes toward meat evolving? What are the most effective policy levers to stimulate behavioral change?) and are more attuned to the realities of policymaking (Kanter, Chodos et al., 2020, Kanter, Del Grosso et al., 2020).

6.2. Optimizing Governance Coordination for Better Consistency Between Agricultural Development and Environmental Sustainability Objectives

Complex tradeoffs exist among various sustainable N management goals and across regions. For example, the Chinese government has made ecological protection a top priority, however, large changes in production modes can lead to unintended costs if enacted blindly without carefully considering multiple impacts, such as farmers' interests and food demand. During the pork crisis in 2019 in China, due to the ban on livestock production in the Yangtze River basin, the benefit to the environment was enhanced, but farmers' profits decreased and consumers' costs increased. Complex tradeoffs also exist among regions, and they are most relevant to the N flows between regions in forms of food and feed trade. For example, our results suggest that importing feed enabled intensification of livestock production, but led to challenges in recycling manure back to cropland. The production of feed in other countries, such as Brazil, has led to adverse environmental impacts such as deforestation and potential N pollution (Huang et al., 2019; Xu et al., 2019), although economically profitable for that country. It appears that reducing a region's reliance on the import of food and feed may trigger more direct feedback of human activities to their environmental consequences, motivating better coupling of N flows within the region, and consequently improving N efficiencies for the Food system and reducing N surplus in the Ecosystem. But it should be recognized that a certain level of import-dependency may be necessary, especially for those countries with limited land resources, and may help to alleviate domestic production shocks due to extreme weather or other events. Ultimately addressing those complex tradeoffs requires vertical (international to local) and horizontal (across ministries and agencies) coordination across government, given N's unique dual role as an essential nutrient and a major pollutant. This is a particularly challenging task given the lack of incentives that many impact-focused (i.e., water, climate, air) policymakers have to develop integrated approaches, which is especially important for N.

6.3. Mandatory and Voluntary Policies Should Be Combined to Increase Management Efficiency

Mandatory policies such as establishment of protection zones together with regulations are necessary in many cases. However, for wider landscape N management, there is increasing evidence that voluntary programs are becoming more effective and flexible to link different stakeholders together and to complement compulsory rules. For instance, the co-operative agreements between farmers, local industries, and authorities aiming at reducing on-farm pollution diffusion to water bodies can be a win-win-win solution as shown in some EU countries such as France and Germany (Heinz, 2007). Farmers can get free advisory programs, monitoring services, and compensation payments from water companies if agro-environmental targets can be achieved (e.g., topsoil nitrate content is less than 80 kg N ha^{-1} in Germany). Water companies can save the costs of water treatment, closing wells, and conveyance of water from remote sources when N surplus is low in local farmlands. France and Germany, which show advanced performance in our *CAFE* analysis, have many such voluntary agreements that advise farmers with different financial sources, resulting in greater capacity to maintain better water quality than permitted standards (Heinz, 2007). In Denmark, more voluntary marketed-based policies include subsides to low-N application in sensitive areas, better manure handing and animal housing, organic farming, etc., which are complementary to the mandatory regulations (e.g., N quota, manure application with machinery, cover crop and rotation). Considerable progress has been made during the past 30 years in N sustainable management, and Denmark has become a good example globally (Dalgaard et al., 2014; Van Grinsven et al., 2012). It is also important for policymakers to consider creative policy options that go beyond traditional mandatory or voluntary policies. For example, in 2015 the Indian government introduced a policy requiring fertilizer manufacturers to coat all domestically sold urea with neem, a natural coating substance that delays N release over the course of the

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growing season. This policy targets the 30 urea-producing production facilities in India, instead of its 120 million farmers, making it an easier task for implementation and enforcement.

6.4. Strengthening Science-to-Policy Interfaces to Develop Science-Based Management Strategies and Tools

For both mandatory and voluntary programs, implementing N management should use science- and evidence-based tools, indicators, and methodologies, which may require targeting the most important science to the policies and practices that need to be strengthened (Lu et al., 2015). Many EU member states (e.g., Denmark, France, and The Netherlands) regulate farmers' N application by setting specific N quotas in different cropping systems and soil types (Van Grinsven et al., 2012). Germany monitors N surplus to regulate farmers' fertilizer input (Van Grinsven et al., 2012). These standards of regulatory indexes are mainly generated and updated by research based on long-term monitoring work, scientific calculation, and modeling. For example, the Knowledge Center for Agriculture and the Danish Pig Research Center in cooperation with Aarhus University provide results on annual N quota adjustments (Ministry of Environment and Food of Denmark, 2017). In the U.S., the best N management practices for guiding local farmers are mainly provided by a loose nationwide system of cooperative extension, which crosses the boundaries between universities, farmers, and local managers. The system consists of agricultural experiment stations that are affiliated with a public university in each state, providing science-based recommendations and training to both private consultants and farmers (Safford et al., 2017). These guidelines customize the optimal fertilizer management practices following "4R" stewardship principles within local contexts and can be very specific to farming conditions, flexible to adopt, and effective for improvement (Gardner, 1990; Mcdowell, 2003; Randall et al., 2008). However, such interfaces remain under-resourced regarding nutrient management, few researchers have the opportunity to work effectively at the interface of the natural and social sciences and management, and training for this work is scare in developing countries. More efforts, both financial and intellectual, in translating science to climatic, agronomic, and culturally-specific practices need to be devoted and highlighted (Chapin, 2017).

7. Conclusion

Application of the *CAFE* framework at the national scale highlights the importance of including the multiple systems that contribute to and are impacted by N management practices and losses, from crop production to entire ecosystems. This analysis also reveals the critical roles of population density and economic factors in determining the pattern of N surplus in the *CAFE* hierarchy: Cropping system is the major contributor to the Ecosystem N surplus for most developing countries, whereas contribution from systems beyond Cropping system dominate in developed countries, and densely populated countries like Japan, The Netherlands, and China, are among the highest in N surplus for the landscape Ecosystem. With the projected increase of population and economic prosperity in developing countries, N surplus generated both within and outside croplands will continue to grow in the absence of effective regulations and incentives, which may aggravate the already-bad situation. In addition to optimizing each subsystem, improvement of the coupling between systems is critical to reducing losses and increasing efficiency: that is, improving the coupling of different system components, such as coupling chemical fertilizer with other N source like manure and BNF, coupling animal production with crop production to reduce animal density to a reasonable level and increase feed self-sufficiency to above 50%, coupling domestic demand and production with the assistance of trade, and coupling human activities with ecosystem capacity. The quantification of N surplus and NUE for each subsystem in the *CAFE* framework assists the identification of N management priorities for each country, such as the need to reduce excess N fertilizer application in China, adjusting animal-crop production portfolio in The Netherlands, reducing food wastes in the U.S., and lowering crop storage wastage and increasing overall productivity in Africa countries. The cross-country/cross-region comparison under a consistent *CAFE* framework facilitates more effective experience-sharing and ultimately better policy. Linking key stakeholders and establishing institutional innovations are key solutions for sustainable N management.

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8. Materials and Methods

8.1. A Detailed Description of the *CAFE* Framework

1. Cropping system. The Cropping system considers cropland as its system boundary, which receives N inputs from chemical fertilizer (N_{fer}), manure (N_{man}), biological fixation (N_{fix}), and deposition ($N_{dep,\mathrm{CrLand}}$). The system's productive output is N in harvest crops for human consumption ($N_{yield,non-feed}$; e.g., grain, fiber and biofuel) and animal production ($N_{yield,feed}$) (Figure 1 and Figure S1 in Supporting Information S3). Data for those variables are often available on the farm scale, and can be aggregated to subnational and national scale crop statistics to assess the regional performance of N management in crop production systems. The N surplus for this system (NS_c) is defined in two ways: as the difference between N fertilizers and productive N outputs (Crop-a, NS_{c1}), or all N inputs and productive N outputs (Crop-b, NS_{c2}), NUE in cropping system is defined as the sum of productive N outputs divided by the sum of N inputs (NUE_C):

$$NS_{c1} = N_{fer} - N_{yield,feed} - N_{yield,non-feed}$$

$$NS_{c2} = N_{fer} + N_{fix} + N_{dep,\mathrm{CrLand}} + N_{man} - N_{yield,feed} - N_{yield,non-feed}$$

$$NUE_C = (N_{yield,feed} + N_{yield,non-feed}) / (N_{fer} + N_{fix} + N_{dep,\mathrm{CrLand}} + N_{man})$$

   The N Surplus may be accumulated in soil to increase soil fertility or be lost to the environment in various forms, such as dissolved organic-N, NH_3, NO_x, NO_3^-, N_2O, and N_2, leading to environmental issues such as air and water pollution. But over a longer time period, such as a decade, the average annual accumulation or loss of N in soil is usually negligible compared to the annual total N inputs, therefore, a positive N surplus could be considered as an indicator of likely N pollution. In contrast, a negative N surplus suggests the cropping system is mining the soil N, which potentially leads to a decline of soil fertility.

2. Animal-crop system. Integrating livestock with crop production, the Animal-crop system imports N from the Cropping system plus other N inputs, includes N input to pastures ($N_{pasture}$), imported feed ($N_{import,feed}$), and recycled byproducts as feed ($N_{recycled,feed}$). Manure application to cropland is N recycled within the system. The Animal-crop system implies the virtual combination and coupling of two systems on a regional scale for evaluation and management and does not necessarily indicate that all farms have both crop and animal production. The N embedded in non-feed crop ($N_{yield,non-feed}$) and animal products ($N_{yield,livestock}$) are the productive output of the integrated Animal-crop system. The N surplus (NS_A) and NUE (NUE_A) are defined as:

$$NS_A = N_{fer} + N_{fix} + N_{dep,\mathrm{GrLand}} + N_{import,feed} + N_{pasture} + N_{recycled,feed} - N_{yield,livestock} - N_{yield,non-feed}$$

$$NUE_A = (N_{yield,livestock} + N_{yield,non-feed}) / (N_{fer} + N_{fix} + N_{dep,\mathrm{GrLand}} + N_{import,feed} + N_{pasture} + N_{recycled,feed})$$

   Similar to N surplus for the Cropping system, this N surplus may be accumulated in soil or be lost to the environment during crop and pasture production or livestock housing and manure management. N accumulation in soils and livestock bodies in the system has been considered as negligible over the long run (Oenema, 2006), therefore, N surplus indicates likely N loss from the system to the environment. Success criteria for NUE and surplus in integrated Animal-crop system have not been reported for any countries.

3. The Food (Food, Fiber and crop-based bioFuel) system traces N from the Animal-crop system to marketplaces and to consumers, and its productive output is N imbedded in the effective uptake of products by consumers ($N_{demand,domestic}$). Fiber and crop-based biofuel can be regionally important, but food is the dominant N product in national scale examples presented in this study. We use "Food system" below to keep the name short. Food imports provide an alternative for supplying food with domestic natural resources, and introduces new N to the system, therefore, N imbedded in food imports is considered as an N input to the Food system in addition to all inputs for the Animal-crop system. Considering the socioeconomic benefits brought by exporting agricultural products, N imbedded in exported products (N_{export}) is also considered as a productive outcome. The import and export of fiber and crop-based biofuel products are treated the same as food products. Crop-based biofuel products are mainly bio-ethanol, like corn ethanol and other ethanol produced by lignocellulosic crops with little N content, leaving N-rich residues that can be recycled as animal feed. The N surplus (NS_F) and NUE (NUE_F) for this system are defined as:

$$NS_F = N_{fer} + N_{fix} + N_{dep,\mathrm{CrLand}} + N_{pasture} + N_{import} - N_{demand,domestic} - N_{export}$$

$$NUE_F = (N_{demand,domestic} + N_{export}) / (N_{fer} + N_{fix} + N_{dep,\mathrm{CrLand}} + N_{pasture} + N_{import})$$

In addition to the N surplus incurred in the Animal-crop system, N surplus for the Food system also includes food lost and food wastage at any point along the food supply chain, from loss to pests during on-farm storage, to losses during processing and distribution, and at processing factories, markets, restaurants, households, even includes overeating beyond a recommended healthy diet. Some losses are inevitable from food processing, for example, wheat middling, tofu cake, brewer grain, etc., although those losses can be reduced through greater efficiencies and through capturing and recycling the losses. In addition, unutilized human waste/excrement is also a part of surplus. Food wastage reduces the efficiency of the supply chain to meet its objective and requires that the crop and animal production systems produce additional harvest that would not otherwise be needed. Success criteria for NUE and surplus in Food system have not been reported globally.

4. The landscape Ecosystem receives N inputs to the entire defined geographic boundary, such as N deposition (from fossil fuel for industry and transportation), and natural N fixation. In addition to N inputs to the Food system, new inputs to this system include atmospheric N deposition from sources such as industry and automobiles to the natural habitat ($N_{dep,NaLand}$). Only products exported from the landscape constitute productive outputs, since the N in products consumed within the landscape is either retained or carried away as pollutants downwind and downstream. Following this definition, the surplus for a given landscape is equal to the net anthropogenic nitrogen import, so called NANI, which has been widely adopted for watershed nitrogen assessment (Howarth et al., 2012). The N surplus (NS_E) in this system are defined as:

$$NS_E = N_{fer} + N_{fix} + N_{pasture} + N_{dep,CrLand} + N_{dep,NaLand} + N_{import} - N_{export}$$

Besides being emitted as water or air pollutants to the environment, the N surplus in this system can be retained in soils and sediments or forest biomass, as well as constructed filters (e.g., wetlands, riparian, water treatment plants) intended to filter nutrients. Some of the N may also be transformed to innocuous N_2 gas in soils, wetlands, and sewage treatment plants, which is a gaseous, non-productive, but also non-polluting export. The NANI is usually correlated with N pollutants and can be seen as an effective indicator to assess the N environmental burden which may leads to eutrophication and causes loss of ecological functions of aquatic ecosystems.

8.2. The N Surplus Increase Between Adjacent *CAFE* Systems

Following the definition of the N surplus across *CAFE* systems, increases in the N surplus between two adjacent systems were identified to help upstand the N dynamics and to explore the driving factors for these N surplus increases.

8.2.1. From Cropping System to Animal-Crop System

$$\begin{aligned} NS_A - NSc &= N_{pasture} + N_{import,feed} + N_{recycled,feed} - N_{yield,livestock} + N_{yield,feed} - N_{man} \\ &= NS_{pasture} + (N_{pasture,feed} + N_{import,feed} + N_{recycled,feed} + N_{yield,feed} - N_{yield,livestock}) - N_{man} \\ &= NS_{pasture} + NS_{livestock} - N_{man} \end{aligned}$$

8.2.2. From Animal-Crop System to Food System

$$\begin{aligned} NS_F - NS_A &= N_{import} - N_{demand,domestic} - N_{export} - N_{import,feed} - N_{recycled,feed} + N_{yield,livestock} + N_{yield,non-feed} \\ &= N_{import,food} - N_{export} + N_{yield,livestock} + N_{yield,non-feed} - N_{demand,domestic} - N_{recycled,feed} \\ &= N_{domestic,food\ supply} - N_{demand,domestic} - N_{recycled,feed} \end{aligned}$$

8.2.3. From Food System to Landscape Ecosystem

$$NS_E - NS_F = N_{dep,NaLand} + N_{demand,domestic}$$

where $NS_{pasture}$ and $NS_{livestock}$ indicates the N surplus generated in pasture production and livestock production, $N_{pasture,feed}$ and $N_{import,feed}$ are harvest pasture for animal feed and imported forage crops, byproducts, and parted

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cereals crops used for animal feed, respectively, $N_{domestic,food\ supply}$ is the domestic N supply as food products, and $N_{dep,NaLand}$ is the atmospheric N deposition to natural lands.

8.3. Data Sources

For each system in the *CAFE* framework, we accounted for the total quantity of N (measured by kg N yr^{-1}) entering or leaving a country for each N input and productive output term based on public databases or literature values. We choose the time period from year of 2000–2010 for the data availability, consistency, and quality. The data after 2010 still needs to be refined and processed, such as distinguishing the N fertilizer use for cropland and grassland from the dataset of "N fertilizer agricultural use" provided in FAOSTAT. The mathematic results are shown as arithmetic averages of this period. In addition, we divided the total N quantity by the agricultural land area (cropland and managed pasture land) reported by FAOSTAT (2010) in order to assess the pressure of N surplus loading to the agricultural land and to enable comparisons among countries with largely different land areas (Figure 2 and Figure S3 in Supporting Information S3).

The following sections introduce how each N input or output term is estimated for each country.

1. Cropping system. All N inputs and productive outputs for this system have been reported on a national scale for all major crop types in the Global Database of N Budget in Crop Production (GDNBCP) (Zhang, Davidson, et al., 2015). We used this N budget database and the harvested area data for each crop from FAOSTAT to aggregate the quantity of N inputs and outputs for each of the 13 countries. Annual atmospheric N deposition in selected countries are referred to various studies shown in Table S2 in Supporting Information S3.
2. Animal-crop system. We calculated the total N fertilizer inputs to pasture according to the N fertilizer application rate and pasture land area in GDNBCP (Zhang, Davidson, et al., 2015), which are derived from FAOSTAT. The total N fixation by pasture was derived based on the reported N fixation rate of 5 kg N ha^{-1} yr^{-1} (Herridge et al., 2008). Following the methods used in the development of GDNBCP (Zhang, Davidson, et al., 2015), we derived an average deposition rate for the selected countries as the deposition rate to the pasture.

In addition to N inputs to cropland and pasture, imported feed is a new N input component for the Animal-crop system. The imported feed includes two major categories: (a) imported forage crops (i.e., silage, grass, soybeans, and green maize): and by-products or co-products from food processing industry (i.e., bran, wheat mids, dried brewer's grains, etc.) and (b) imported cereal crops (e.g., maize, wheat), used for animal production. The N inputs in the first category were derived from FAOSTAT trade sheet (www.fao.org/). To calculate the N inputs in the second category, we estimated the imported cereal crops based on the FAOSTAT trade sheet, and the fraction of imported cereal crops used for animal production, assuming that the fraction of cereal crops used for animal production is the same for imported and domestically produced cereal crops. The fraction of cereal crops used for animal production is estimated by the overall domestic consumption of cereal crops (estimated from the FAOSTAT trade sheet) and the amount of cereal crops used for human consumption (Billen et al., 2014).

The outputs in Animal-crop system consist of two components, animal products and non-feed crop products. Animal products ($N_{yield,livestock}$) included livestock primary and processed products, commodities items listed in Data S1 in Supporting Information S1; data were derived from the FAOSTAT, livestock production sheet (2000–2010). Non-feed crop products ($N_{yield,non-feed}$) is the rest of domestic crops for feed from total crops harvest. Harvest pasture for animal feed ($N_{pasture,feed}$) refers to the agro-food N Budget in Billen et al. (2014).

3. Food system. Additional N input in this system is from imported food, fiber, and crop-based biofuel, and outputs include food, fiber, and crop-based biofuel for domestic consumption and export. The data of food and fiber import and export for each country (N_{import}, N_{export}) were from FAOSTAT, Trade sheet, 2000–2010. Domestic food demand ($N_{demand,domestic}$) was calculated by the population multiplied by the recommended dietary intake by human (3 kg N yr^{-1} per capita (de Vries et al., 2013)) in each country. N imbedded in consumed fiber and biofuel products was ignored because N contents in biofuel products is negligible compared to N in food and products (Huo et al., 2012; Nocentini et al., 2018).
4. Landscape Ecosystem. In addition to N inputs to the "Food system," new N inputs to this system include atmospheric N deposition to natural lands. Information on N deposition for the whole landscape in 13 selected countries was obtained from the studies shown in Table S2 in Supporting Information S3.

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The productive ecosystem outputs include only part of the food system outputs, namely the exported food, fiber and crop-based biofuel products (N_{export}). For the 13 countries examined in this study, the N loads from upstream are negligible compared to other N inputs, and were consequently ignored.

8.4. Livestock Density

Livestock density was expressed as LU per ha of agricultural area, in which the relative weight of a mature dairy cow is set at 1 and the other livestock categories at 0.5 for beef cattle, 0.35 for pigs, 0.012 for laying hens, 0.018 for other poultry, and 0.1 for sheep and goats (Eurostat, 2009). The feed demand for 1 LU roughly ranges between 4,000 and 7,000 kg dry matter per year, depending on milk yield quality of the feed, and production system. The data of livestock numbers of different animal types is from FAOSTAT livestock sheet.

8.5. Sensitivity Analysis

To test the robustness of the results we did the sensitivity analysis using different time period data including 1990–2000, 2000–2010, and 1990–2010 and got consistent trends. Figures S8 and S9 in Supporting Information S3 show part of the examples in Cropping system and Animal-Crop system.

Conflict of Interest

The authors declare no conflicts of interest relevant to this study.

Data Availability Statement

The data sets used in this study were reported by FAOSTAT (2010) and Global Database of N Budget in Crop Production (GDNBCP) (Zhang, Davidson, et al., 2015). Data of nitrogen contents and the classification of different crop and animal products were provided in Dataset S1" in Supporting Information S1 and data of nitrogen budgets across CAFE system during the defined periods for the selected countries can be accessed from Dataset S2 in Supporting Information S2. These two datasets are also available on the repository at https://datadryad.org/stash/share/TwLv7bMJBxi4GRF3Lx68jIGeDSm2B0GIvFQJc4qAmKo.

Acknowledgments

We thank H. Charles J. Godfray for his comments on an earlier version of the manuscript and assistance in manuscript revision. This work was financially supported by National Key Technologies R&D Program (Grant 2016YFD0201303), Major Science and Technology Program of Hainan Province (Grant ZDKJ2021008), Innovative Group Grant of the Natural Science Foundation of China (Grant 31421092), National Science Foundation CNS-1739823, and National Socio-Environmental Synthesis Center (SESYNC) under funding received from the National Science Foundation DBI-1639145.

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