

# Global Biogeochemical Cycles

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### Key Points:

- Sustainable nutrient management, a major challenge of this century, requires better quantification of nutrient budgets
- This paper reviews major challenges in defining, quantifying, and applying nutrient budgets on multiple system and spatial scales
- The improvement and limitation in nutrient budgets need to be communicated among researchers and stakeholders to enable positive changes

### Supporting Information:

- Supporting Information S1
- Table S1

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## Quantifying Nutrient Budgets for Sustainable Nutrient Management

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**Abstract** Nutrients, such as nitrogen and phosphorus, provide vital support for human life, but overloading nutrients to the Earth system leads to environmental concerns, such as water and air pollution on local scales and climate change on the global scale. With an urgent need to feed the world's growing population and the growing concern over nutrient pollution and climate change, sustainable nutrient management has become a major challenge for this century. To address this challenge, the growing body of research on nutrient budgets, namely the nutrient inputs and outputs of a given system, has provided great opportunities for improving scientific knowledge of the complex nutrient cycles in the coupled human and natural systems. This knowledge can help inform stakeholders, such as farmers, consumers, and policy makers, on their decisions related to nutrient management. This paper systematically reviews major challenges, as well as opportunities, in defining, quantifying, and applying nutrient budgets. Nutrient budgets have been defined for various systems with different research or application purposes, but the lack of consistency in the system definition and its budget terms has hindered intercomparison among studies and experience-sharing among researchers and regions. Our review synthesizes existing nutrient budgets under a framework with five systems (i.e., *Soil-Plant* system, *Animal* system, *Animal-Plant-Soil* system, *Agro-Food* system, and *Landscape* system) and four spatial scales (i.e., Plot and Farm, Watershed, National, and Global scales). We define these systems and identify issues of nitrogen and phosphorus budgets within each. Few nutrient budgets have been well balanced at any scale, due to the large uncertainties in the quantification of several major budget terms. The type and level of challenges vary across spatial scales and also differ among nutrients. Improvement in nutrient budgets will rely not only on the technological advancement of scientific observations and models but also on better bookkeeping of human activity data. While some nutrient budget terms may need decades, or even centuries, of research to be well quantified within desirable levels of uncertainties, it is imperative to effectively communicate to interested stakeholders our understanding of nutrient budgets so that scientists and a variety of stakeholders can work together to address the sustainable nutrient management challenge of this century.

**Plain Language Summary** Managing nutrients, such as nitrogen and phosphorus, is fundamental, yet challenging, for sustainable development. Nutrients are critical for plant and animal growth in agriculture and in nonagricultural ecosystems and are consequently important for food security and climate stability, as well as human health. However, historical and ongoing increases in nutrient inputs to agriculture, while increasing food production, have also contributed to severe environmental problems, ranging from local water and air pollution to global climate change. Quantifying the nutrient inputs and outputs of a farm, watershed, or any other well-defined boundary advances our knowledge of how to maintain a nutrient balance, which is an essential step toward sustainable nutrient management. So far, many research efforts have been devoted to quantifying nutrient budgets and to improving nutrient management for different systems (e.g., farms and food supply chains) and at different spatial scales (e.g., from a single farm to the entire world). However, due to the complex nature of nutrient cycles and incomplete data sets, challenges remain in quantifying and understanding nutrient budgets to inform policies and actions for sustainable nutrient management. With a systematic review of major challenges in defining, quantifying, and applying nutrient budgets, this paper calls for collective efforts by researchers, farmers, watershed managers, consumers, policy makers, and other stakeholders involved in nutrient

management to improve our ability to balance nutrient budgets and to use that understanding to grow abundant food while avoiding pollution.

## 1. Managing Nutrients in the Anthropocene

Nutrients are fundamental for the survival and growth of life on Earth. To provide a stable and abundant food supply for human society, agriculture has rapidly intensified during the last century, partly owing to our increasing capability to harness nutrients for agricultural production (e.g., the production of nitrogen fertilizer from the Haber-Bosch process and the use of phosphate rock).

The intensive use of nutrients in agricultural production, along with their use in other human activities, has largely distorted the nutrient cycles in the Earth system, especially for the macronutrients, nitrogen (N), and phosphorus (P) (Sutton et al., 2013). For example, the reactive N added to the Earth system by human activities has increased from less than 20 before 1920 to more than 180 Tg N year<sup>-1</sup> in 2015, surpassing the annual N fixed by natural processes on land or in the oceans (Fowler et al., 2013). Among all anthropogenic N and P inputs, over 90% are from agricultural production (Cordell et al., 2009; Fowler et al., 2013). This intensive use of nutrients for food production has led to severe environmental issues, ranging from local water and air pollution to global climate change. It has been reported that human disturbance to the N and P cycles have crossed the so-called “planetary boundary,” suggesting many impacts on the Earth system, including some that may be irreversible (Figure 1, Steffen et al., 2015).

In contrast to the problems of “too much” on a global scale, many regions also suffer from the problems of “too little” availability of nutrients for agriculture (Sutton et al., 2013). Regions, such as Sub-Saharan Africa, still need more macronutrient inputs (e.g., N, P, and sulfur) to reverse declining soil fertility, boost yield, and nourish the population, but fertilizers, high-quality manures, and mulch are often unavailable or unaffordable. In addition to macronutrients, the lack of micronutrients in food, such as minerals like zinc, iodine, and vitamin A, merits more attention.

Consequently, it is imperative to improve nutrient management in order to feed the population with nutritious food while minimizing unintended and undesirable environmental impacts. However, improving nutrient use requires the ability to measure and monitor the fates of nutrients that are cycling within and escaping from ecosystems, but nutrient budgets have been notoriously difficult to quantify. The objective of this review is to provide a historical overview of the nutrient budgeting efforts and identify key challenges in defining, quantifying, and applying nutrient budgets.

## 2. A Brief History of Nutrient Budgets

You cannot manage what you cannot measure (Deming, 2018). Quantifying nutrient budgets, namely the nutrients entering and leaving a given system, is the first step toward better nutrient management. This approach is based on the principle of conservation of mass, where the difference between the nutrient inputs ( $Nu_{inputs}$ ) and outputs ( $Nu_{outputs}$ , including productive outputs  $Nu_{prod\cdot outputs}$  and losses  $Nu_{losses}$ ) of a system changes the nutrient stock ( $Nu$ ) within the system (Figure 2).

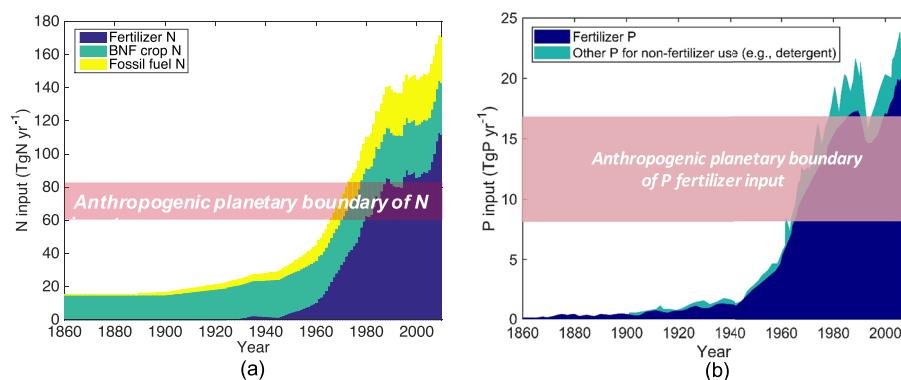
$$Nu = Nu_{inputs} - Nu_{prod\cdot outputs} - Nu_{losses}$$

Consequently, the efficiency of nutrient use ( $NuUE$ ) and the nutrient surplus ( $NuSur$ ) for a system are defined as follows:

$$NuUE = Nu_{prod\cdot outputs} / Nu_{inputs}$$

$$NuSur = Nu_{inputs} - Nu_{prod\cdot outputs} = Nu_{losses} + Nu$$

The origin of nutrient budgeting dates back to midnineteenth century, when European scientists started to quantify nitrogen in rain and drainage waters in field plots (Boussingault, 1841; Lawes, 1882). As N had just been identified as one of the critical nutrients for plant growth, nutrient budgeting started with a focus on soils and crops as a system, targeted at utilizing nitrogen for plant productivity. Since then, the budgeting

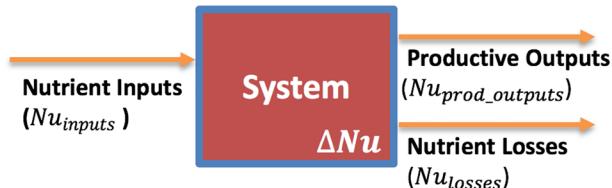


**Figure 1.** Nitrogen (a) and phosphorus (b) inputs from human activities to Earth system. The data for N and P inputs are from Holland, Lee-Taylor, et al. (2005) and Cordell et al. (2009), respectively. The anthropogenic planetary boundaries of N and P fertilizer input are from Steffen et al. (2015) and Springmann et al. (2018). BNF: Biological Nitrogen Fixation. The green area in Figure 1b denotes the P produced from phosphate rock for nonfertilizer use.

approach, also called soil nutrient balance, has been developed with more input and output items (Allison, 1955) and has been applied to other important nutrients for crop growth, such as phosphorus (P), potassium (K), calcium (Ca), magnesium (Mg), and sulfur (S).

Concerned about the depletion of soil nutrients and its consequences on food supply for the United States in early twentieth century, Lipman and Conybeare (1936) took a broader view of nutrient budgets beyond the spatial scale of a few field plots and developed nutrient budgets for all agricultural land for the United States. This development extended the application of nutrient budgeting approach from on-farm management to nutrient management on a national scale. Since the end of World War II, growing amounts of fertilizer have been applied to cropland, especially for N, P, and K, to increase crop yields. These additions have prevented nutrient depletion and, in some cases, have even built up soil nutrient stocks. However, the depletion of soil N and P is still of major concern for food security in many least developed countries, including many Sub-Saharan African countries, and the depletion of soil micronutrients is still prevalent but poorly quantified around the world (Bouwman et al., 2017; Vitousek et al., 2009).

Following the widespread reports of undesirable algal blooms in lakes and estuaries in the midtwentieth century, which has become known as eutrophication caused by excess nutrients (Galloway et al., 2013; Garnier et al., 2010; Glibert et al., 2014), growing concerns about the impacts of fertilizer runoff on water quality and human health have led to development of nutrient budgeting on watershed scales (Howarth et al., 1996; Lowrance et al., 1985; Robertson, 1986; Viets, 1971). Two decades of research have uncovered and solidified the strong linkages between riverine N output and the net anthropogenic nitrogen inputs (NANI) to the watershed (Boyer et al., 2002; Howarth et al., 1996; Jordan & Weller, 1996; Lassaletta et al., 2012; Swaney et al., 2012). The same linkage has also been found for P (Hong et al., 2012). In recent years, the impacts of climate change, especially the changes in precipitation, and the legacy effect of nutrient accumulation on riverine N export have been examined in addition to the anthropogenic N inputs (Powers et al., 2016; Sinha & Michalak, 2016).



**Figure 2.** A schematic of nutrient budgets for a system. “Productive outputs” include those nutrients harvested and removed from the system, such as crops, straw, and animal products, which are usually economically valuable products.

Although our emphasis here is on nutrient budgets in which agriculture is a major component, it should be noted that nutrient budgets are also extremely relevant for understanding the biogeochemical processes in nonagricultural systems. The classic works by Bormann and Likens (1979) on the nutrient budgets of forested watersheds of Hubbard Brook, New Hampshire, and more recent updates such as Groffman et al. (2018), revealed the impacts of forest management, natural disturbance, secondary succession, and acid deposition in the patterns and processes of forested ecosystems. Similarly, the classic whole-lake nutrient addition experiments initiated by Schindler in 1969 yielded N and P budgets and insights into their roles in freshwater eutrophication processes (Schindler et al., 2008).

The concern over increasing fertilizer use and its impacts on biogeochemical cycles also sparked the quantification of nutrient budgets on a global scale. The SCOPE report edited by Svensson and Söderlund (1976) was among the first to produce global budgets for N, P, and S (Granat et al., 1976; Pierrou, 1976; Söderlund & Svensson, 1976). Meanwhile, in the last quarter of the twentieth century, two global-scale environmental issues, namely stratospheric ozone depletion and climate change, became increasingly recognized, and the contribution of nitrous oxide ( $N_2O$ ) to these two global issues was identified (Crutzen, 1970; Ehhalt et al., 2001). Consequently, quantifying N budgets, including  $N_2O$ , on a global scale has become of interest to both scientists and policy makers (Bouwman, Beusen, et al., 2013; Crutzen et al., 2008; Davidson, 2009; IPCC, 2006; Thompson et al., 2019; Tian et al., 2019; UNEP, 2013). In the recent decade, studies on planetary boundaries further highlighted the importance of quantifying N and P budgets on a global scale, as human impacts on N and P cycles have surpassed the proposed boundaries (Rockstrom et al., 2009; Steffen et al., 2015). After the price spike of phosphate rock in 2008, there was a growing interest in long-term P rock security and the impacts of depleting P rock reserves on food security (Cordell & White, 2014). Although a significant increase in P rock reserves was reported after earlier estimations warning of an approaching peak in phosphorus reserve availability (Cordell et al., 2011; Cordell & White, 2014; USGS, 2019; Van Kauwenbergh, 2010) and researchers have also called into question the basis for a peak P hypothesis (Scholz & Hirth, 2015), there are still concerns on the depletion of rock reserves (especially high grade reserves), the geopolitical consequences of its uneven distribution, and other scarcity issues (Cooper et al., 2011; Cordell & White, 2014; Edixhoven et al., 2014; Khabarov & Obersteiner, 2017).

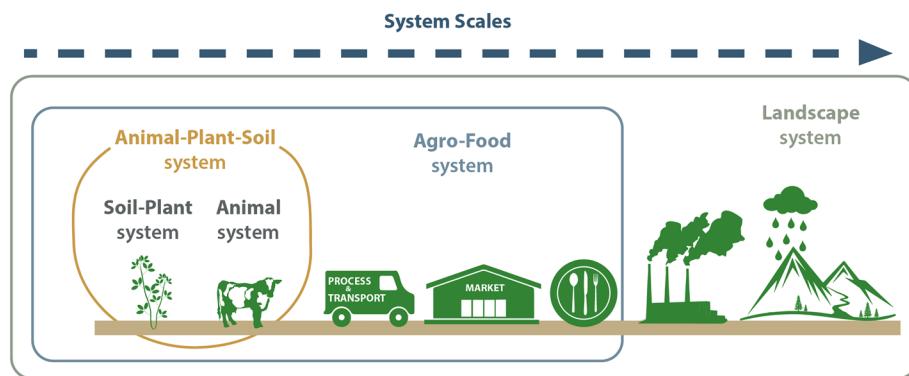
As the trade-offs between providing nutritious food to a growing population and prevalent nutrient pollution became increasingly acute at the beginning of the 21st century, improvements in nutrient management are not only needed in agricultural production but also needed in a broader system that includes consumers (Sutton et al., 2013). Consequently, nutrient budgeting approaches have extended their application from focusing on production only to considering the whole food system, including consumption and the supply chain. For example, following principles of Life Cycle Assessment, N (or P) footprint analyses quantify N (or P) pollution or resource depletion associated with consuming different food and energy goods by an individual or by an institution such as a university (Erisman et al., 2018; Hoekstra & Wiedmann, 2014; Leach et al., 2012; Leach et al., 2016; Leip et al., 2014; Uwizeye et al., 2016; Xue & Landis, 2010). Conceptual models, such as the Generic Representation of Agro-Food Systems (Billen et al., 2014), have been developed to track N flows among regions and sectors in the food system.

Overall, major motivations for quantifying nutrient budgets include the following:

1. identify and quantify missing nutrient sources or sinks (e.g., Crutzen et al., 2008; Davidson, 2009);
2. evaluate the efficiency of nutrient use in a system, such as the crop and livestock production system or the whole food system (Bai et al., 2018; Bouwman, Goldewijk, et al., 2013; EUNEP, 2015; Lu et al., 2019; MacDonald et al., 2011; Quemada et al., 2020; Zhang, Davidson, et al., 2015);
3. assess the environmental impacts of nutrient losses from a system, such as estimates of  $N_2O$  emission from agricultural production based on N inputs or N surplus (Bodirsky et al., 2012; de Vries et al., 2011; Thompson et al., 2019; Tian et al., 2019) or riverine export of N from watersheds based on the NANI methodology (Howarth et al., 2012); and
4. inform stakeholders, such as farmers and policy makers, on the performance of nutrient management and identify how the productivity of the nutrient use can be improved and nutrient pollution can be reduced (Davidson et al., 2016; McLellan et al., 2018).

### 3. Challenges in Defining Nutrient Budget Systems

Given strong motivations from multiple stakeholders involved in nutrient management, nutrient budgets have been quantified on various spatial and system scales, ranging from a single plot to the whole globe, and from a *Soil-Plant* system to the *Landscape* system (Figure 3). The flourishing application of nutrient budgets has significantly advanced our understanding of nutrient cycles, especially for N and P, but has also been accompanied with confusion, even misunderstanding, over nutrient budgets (i.e., inputs and outputs of a system) and the assessment of the system efficiency, leading to major challenges in comparing nutrient budget results among studies and across scales.



**Figure 3.** An illustration of the system scales used for quantifying nutrient budgets. The definition of the system scales is developed from Li et al. (2019). The system scales define the key elements that will be examined within a physical boundary (i.e., a spatial scale). For example, in a watershed, the assessment of the nutrient budget can focus on the crop production only (i.e., a type of Soil-Plant system), include the production of livestock (i.e., Animal-Plant-Soil System), or include the whole food supply chain (i.e., Agro-Food System), or all the other human activities that induce N inputs to the landscape, such as fossil fuel burning (i.e., Landscape System).

Nutrient budgets have been quantified for mainly five systems, including *Soil-Plant* system, *Animal* system, *Animal-Plant-Soil* system, *Agro-Food* system, and *Landscape* system (Figure 3 and Table 1). Each of these systems could be applied on most of the four spatial scales, namely Plot and Farm, Watershed, National, and Global scales. Those spatial scales define the physical boundaries for the nutrient budget accounting. Both Watershed and National scales can be considered as regional scales, but one is defined by physical characteristics of the landscape, and the other is defined by political boundaries. Given the goals of the nutrient budget study, a time scale also needs to be selected, ranging from a single growing season to several years or decades (Meisinger et al., 2008).

### 3.1. *Soil-Plant* System

The nutrient budget of a *Soil-Plant* system, also called soil surface budget (Oenema et al., 2003) or nutrient balance (McLellan et al., 2018), considers the soil and the plants growing in it as an integrated system and accounts for the inputs and outputs of the integrated system. For example, the major N inputs for the system include synthetic fertilizer, atmospheric deposition, biological fixation, and manure. The major outputs include harvested crop products (Figure 4a), which, in addition to grains and other commercial crops, could also include crop residues removed for fuel, feed or other uses off site. In this system, the dynamic nutrient exchanges within soil and between the soil and the plants are often considered as internal processes and are not accounted for in the input/output budget.

The *Soil-Plant* system approach has been increasingly adopted for assessing the efficiency of crop nutrient management, but two challenges associated with this system need to be noted: (1) the efficiency assessment is based on the assumption that the system has achieved a steady state ( $N_u$  is 0 or negligible compared to  $N_{u,prod} - N_{u,outputs}$ , see Figure 2), but long-term observations are often required to ensure the quasi steady state, especially for cropping systems with multiyear rotation practices, perennial crops, or large nutrient reserves in soil; (2) whether to include crop residue as part of the productive output of the system can lead to significant difference in  $N_{u,eff}$  assessment. For example, the N use efficiency (NUE) for the *Soil-Plant* system can increase from 43% to over 58% (depending on the harvest index) if crop residue is included as one of the productive outputs. However, the consideration of crop residue should depend on whether the residue is harvested (i.e., removed from the site) and how it is used, for which information is often limited.

Mineralization of soil organic matter provides inorganic N and P available for crop uptake, but it should not be considered as a new input to the *Soil-Plant* system defined here. Rather, it is accounted for as part of the  $N_u$  term in Figure 2 if mineralization exceeds immobilization and if the mineralized N or P is taken up by the crop and removed in the harvest. Internal cycling of soil nutrients through mineralization, immobilization, and turnover (MIT) of soil organic matter has been studied extensively for decades, as it is essential for estimating the crop demand that can be met by MIT of organically bound nutrients derived from previous years'

| Spatial\systems scales |  | Soil-Plant system  | Animal system  | Animal-Plant-Soil system   | Agro-Food system   | Landscape system                          |
|------------------------|--|--|--|--|--|---|
| Plot and Farm scale    | Several cases for cropping systems based on fertilization trials (EUNEP, 2015) | Commercial broilers; Patterson et al. (1998)                                       | The cases for mixed crop-livestock farming systems in EUNEP (2015); Powell and Rotz (2015) | The combination of animal production and crop production systems in Ma et al. (2012)   | The combination of animal and crop production and food processing in Ma et al. (2012)                      | Howarth et al. (1996); Hong et al. (2012) |
| Watershed scale        | Chen et al. (2017); Liu et al. (2008)  | Poultry layer, broiler, and turkey operations in an aquifer (ZebARTH et al., 1998) | The farm N budget for EU countries in Leip et al. (2011)                                   | The farm N budget for EU countries in Leip et al. (2011)                               | Billen et al. (2014); Ma et al. (2010); Le Noë et al. (2016); Le Noë et al. (2017); Bodirsky et al. (2014) | Han et al. (2014)                         |
| National scale         | Bouwman et al. (2017); Zhang, Davidson, et al. (2015)                          | Pig and poultry systems in Netherland (Oenema, 2006)                               | The combination of feed production and pig production systems in Uwizeye et al. (2019)     | The combination of feed production and pig production systems in Uwizeye et al. (2019) | Billen et al. (2014); Bodirsky et al. (2014)   | Smith et al. (2012)                       |
| Global scale           | Bouwman, Goldewijk, et al. (2013); Zhang, Davidson, et al. (2015)              | The pig production system in the whole pork supply chains (Uwizeye et al., 2019)   |  |  |  |   |

*Note.* Gray shaded cells note the categories with existing studies. While it is not an exhaustive list of references, it provides good coverage of the types of systems that have been studied for nutrient budgets. The cases or systems are specified in the table when more than one case or system was examined in a study.

inputs of crop residues to the soil (Lin et al., 2016; Meisinger et al., 2008; Norton & Schimel, 2011). In the context of a multicropping system, MIT imbalance under one crop can have important effects on nutrient availability for the next crop of the cropping system cycle. For example, biological N fixation inputs under soy that are retained as crop residues and incorporated into soil organic matter become important sources of N that partially meet the demand of the following corn crop. The net input/output balance is most relevant for the entire crop rotation period, whereas the internal cycling is highly relevant for varying balances of inorganic, organic, and total nutrient pools within the rotation period. As crop rotations become more complex than simple corn-soy rotations, the internal cycling and MIT estimates become more important for understanding nutrient dynamics within each phase of the cropping cycle.

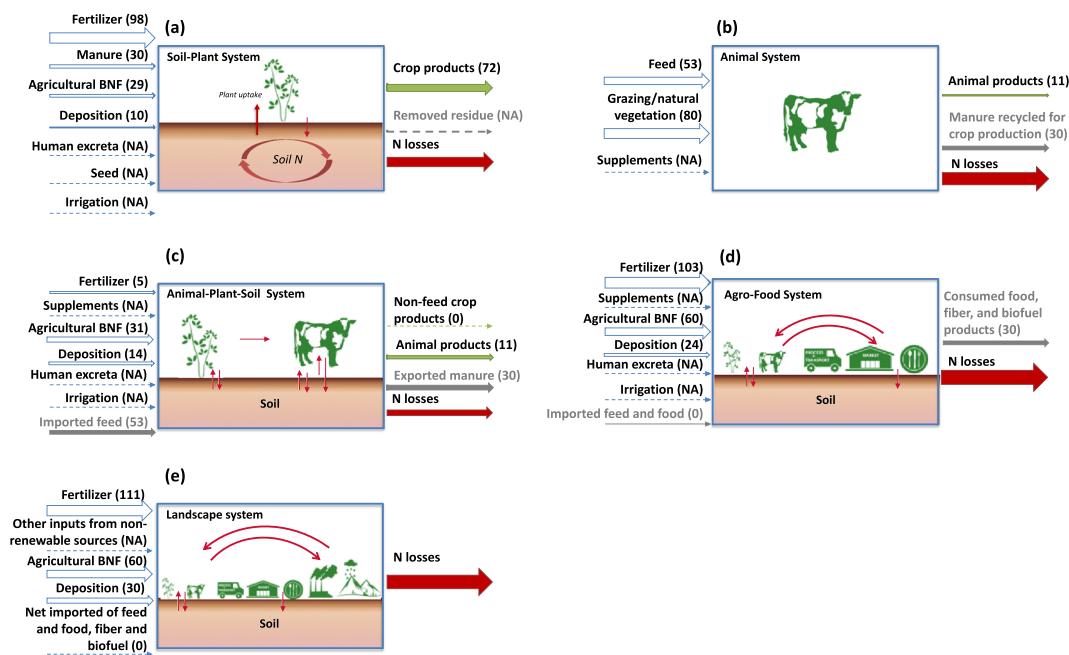
### 3.2. Animal System and Animal-Plant-Soil System

The *Animal* system concerns the animal production only and considers feed production (e.g., from pasture or crop) as a separate system providing nutrient inputs to the *Animal* system; while the *Animal-Plant-Soil* system considers animal production integrated with the crop and/or pasture production; consequently, the locally produced feed and the manure recycled to cropland are both internal processes in this system. Examples of major inputs and outputs of the two systems on a global scale are provided in Figure 4. The example shows that the NUE for the *Animal-Plant-Soil* system is 15%, in comparison to 8% for *Animal* system alone.

Extending the system boundary beyond the animal production facility or farm, the *Animal-Plant-Soil* system enables the consideration of nutrient losses occurring during the production of feed and other inputs for the livestock production. However, where to draw the spatial boundary for accounting the “imported” feed and “exported” manure affects the nutrient budget assessment and consequently challenges the intercomparisons. For example, accounting for the N inputs of imported feed increases the N inputs of the system and consequently reduces the calculated NUE. In contrast, accounting for the manure moved out of the spatial boundary (e.g., sold to another farm) as part of the productive output will increase the system outputs for the farm where it was produced and increase the calculated NUE, even though some portion of the manure N will eventually be lost in the export process (Bai et al., 2014, 2018; Godinot et al., 2015). As a result, the calculated efficiency of a system can vary depending upon how factors like feed imports or manure export are treated in the accounting system, and how recycling within a spatial boundary is considered (Quemada et al., 2020).

### 3.3. Agro-Food System

The *Agro-Food* system extends the system boundary from production to the whole food supply chain and traces nutrient use from agricultural production to marketplaces and to consumers. Many of the nutrient budget assessments for this system investigate not only the inputs and outputs of the system but also the interactions and nutrient flows between its subsystems, such as the *Soil-Plant* system and the *Animal* system. The assessment of nutrient budgets for this system is challenging for two major reasons: (1) The nutrient flows through the food supply chains (e.g., processing and retailing) are very complex, and information is often scarce (Ma et al., 2010). For example, the processing of soybean results in



**Figure 4.** Examples of major inputs and productive outputs of the five systems. The numbers in the parentheses are the estimate of each budget term for N on the global scale with the unit of  $\text{Tg N year}^{-1}$ . “NA” indicates that the estimate is not available for the budget term in this example. The estimates are based on Billen et al. (2014), with minor modifications as fully described in Table S3.

multiple co-products or by-products, such as oil and soybean meal (Dourado et al., 2011), which can be sold to customers or be recycled within the *Agro-Food* system. Most studies estimate the productive outputs of consumed food based on household surveys or daily-intake guidelines (Leach et al., 2012; Ma et al., 2010), instead of actually following the nutrient flows from the upstream source and through the supply chain. (2) With the increasingly connected interregional and international markets, the food system for a region or a country can rarely stay independent; consequently, it is challenging to draw the geographic system boundary to consider trade and recycling components across spatial boundaries while keeping the tasks of quantifying nutrient budgets manageable (Lassaletta, Billen, Grizzetti, Garnier, et al., 2014; Oita et al., 2016).

To address those challenges, two types of approaches have been taken: (1) the first approach focuses on the nutrient flows involved in producing one or a selection of food products, including the resulting nutrient inputs outside of the defined spatial boundary (e.g., the N and P fertilizer use for producing imported feed, also known as virtual N and P; Erisman et al., 2018; Huang et al., 2019; Leach et al., 2012; Mu et al., 2016; Uwizeye et al., 2019); and (2) the second approach focuses on the function of the food system and its environmental impacts within the spatial boundary; it accepts imported food or feed as nutrient inputs equivalent to others for the *Animal-Plant-Soil system* and ignores the external environmental impacts of producing food and feed imported from outside of the spatial boundary (Ma et al., 2010). This approach shares similar challenges described for the *Animal-Plant-Soil system* above. The two different approaches may result in different assessments for the N budget and the system efficiency, when the defined country/region imports or exports a large amount of feed or food. Taking China as an example, the N inputs and NUE of the *Agro-Food* system are 68.5 Tg and 6%, respectively, based on the first approach, compared to 48.8 Tg and 9% based on the second approach (based on data from Ma et al., 2010; see Table S1 for details).

The first approach, also called a N footprint for the consumed products, has been promoted to guide consumer's dietary choices. In contrast, the spatially defined second approach could be used for adjusting production and trade portfolios for a given region to meet local food demand and reduce regional nutrient pollution. However, few studies have examined the connections between two approaches and their implications in guiding the decision making by individuals and policy makers. For example, the calculated N footprint based on the first approach, which includes the virtual N inputs used outside the region to produce the imported product, may encourage more locally produced food where N inputs might be better managed, but it would,

nevertheless, add an environmental burden for the region to support more direct N inputs and their environmental consequences.

### 3.4. *Landscape* System

The *Landscape* system considers human activities and the terrestrial ecosystem as an integrated system and assesses the “new” nutrient introduced by human activities, including agricultural and industrial production, into the system via synthetic fertilizer application, atmospheric deposition, crop N fixation, and net import of livestock feed and human food (i.e., the difference between imports and exports; Howarth et al., 1996). Unless imported from another landscape region, nutrients in manure and sewage produced within the landscape as well as consumption of locally produced food are considered as internal flows of the system that are converted from the “new” nutrients and consequently are not included in the inputs of the system. The application of this system is mostly known for the NANI or net anthropogenic phosphorus input (NAPI) approaches, a positive relationship between NANI (or NAPI) and riverine nutrient export has been reasonably well established across watersheds (Hong et al., 2012; Swaney et al., 2012).

While the riverine nutrient output has been extensively examined, internal flows with positive or negative effects are less investigated, and the productive outputs of the system are generally not considered. Instead of using only productive nutrient outputs, social and economic products, such as population size or GDP, may be considered as the productive outcomes of the *Landscape* system for assessing how efficiently “new” nutrients are used for supporting human activities for a given region.

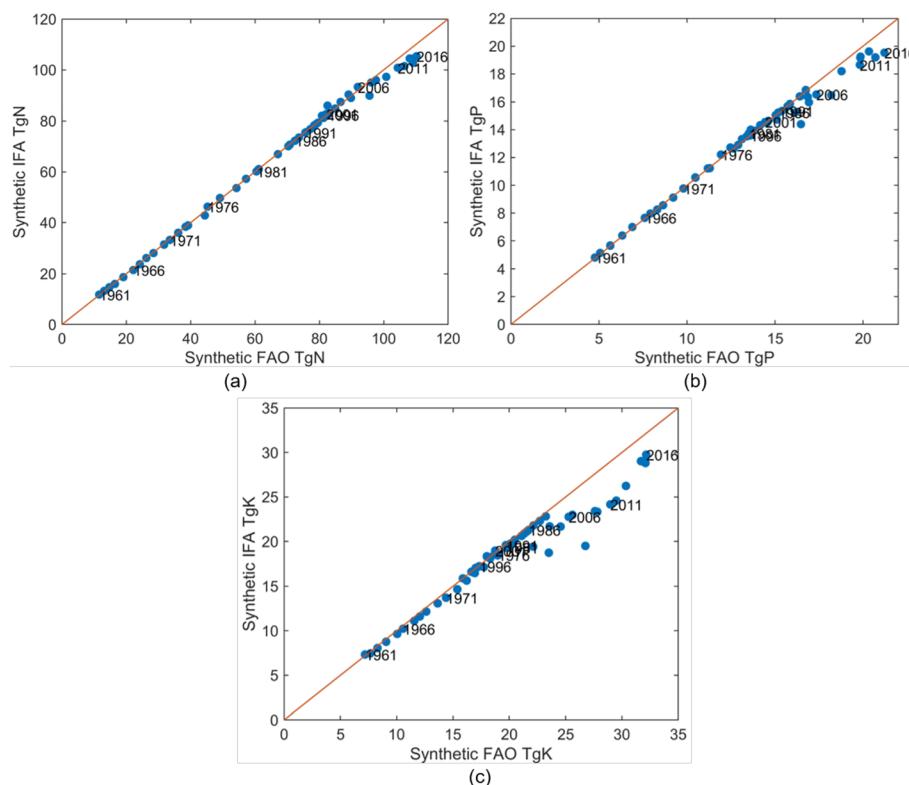
## 4. Challenges in Quantifying Nutrient Budgets

Regardless of the definition of the systems to which nutrient budgets are applied, they are usually difficult to balance, often leading to significant “missing” nutrients (Bouwman, Beusen, et al., 2013). This could be caused by the large uncertainties of many budget terms or potentially some nutrient flows have not been identified or accounted for, such as soil mineralization-sequestration, ammonia volatilization, nitrate leaching, and denitrification. The uncertainties for a nutrient budget term vary across spatial scales because available measurement approaches and data quality vary. Here we review the quantification challenges of eight major nutrient budget terms on four spatial scales, namely Plot and Farm, Watershed, National, and Global scales, and two quantitative examples are presented in Table S2 in the supporting information.

### 4.1. Nutrient Inputs From Synthetic Fertilizer

Synthetic fertilizers are the major N and P inputs for *Soil-Plant*, *Agro-Food*, and *Landscape* systems examined at the global scale, accounting for over 50% of the total inputs (Figure 4). It also dominates the nutrient inputs in many nutrient budget studies applied to various systems and at various spatial scales. For example, synthetic fertilizer accounts for about 80% of N inputs for maize production in northeast China. Consequently, a potential 10% uncertainty in the synthetic N fertilizer input will result in about 20% uncertainty in the N surplus, which is equivalent to the sum of reactive N losses in the forms of  $\text{NH}_3$ ,  $\text{N}_2\text{O}$ , and leaching and runoff (about  $21 \text{ kg N ha}^{-1}$ ) (Zhang, Ju, et al., 2019). Although 10% seems like a relatively small uncertainty for a budget term, the size of the fertilizer input is often so large that this 10% uncertainty dwarfs other budget uncertainties, even where they are a large percentage of the central estimate. Accurately quantifying the nutrient inputs from synthetic fertilizer is critical for balancing the nutrient budget. In contrast, the uncertainty of a nitrous oxide ( $\text{N}_2\text{O}$ ) emission estimate may be  $\pm 50\%$  or more due to large spatiotemporal variation of fluxes, but the flux is usually  $<10 \text{ kg N ha}^{-1} \text{ year}^{-1}$ , so this uncertainty term is not large relative to total inputs and outputs at the farm scale, despite being important for global  $\text{N}_2\text{O}$  budgets.

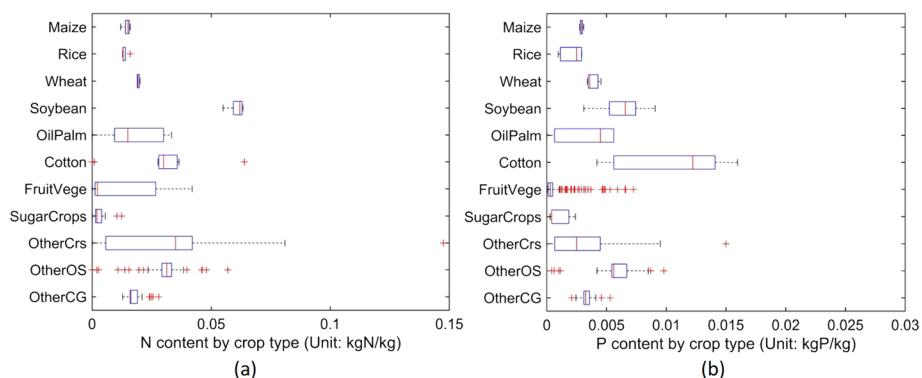
Information on fertilizer inputs is often available at two spatial scales, namely farm and national scales. Farmers (or scientists conducting field experiments) usually have good knowledge of the amount of fertilizer applied in their fields, since it is one of the major costs for their production. On the national scale, countries have been requested to report their fertilizer use for agricultural production, for which they often aggregate their census data on fertilizer use or sales from subnational scales. These data have been compiled and made available by FAOSTAT since 1961 (FAOSTAT, 2019). The International Fertilizer Industry Association (IFA) also reports fertilizer production and consumption data on a national scale based on surveys with fertilizer companies (Heffer, 2013; Heffer et al., 2017). While most countries have reported total synthetic N fertilizer consumption from the two data sources in approximate agreement, a few countries (e.g., China) show



**Figure 5.** Comparing global total synthetic N (a), P (b), and K (c) consumption reported by FAO (FAOSTAT, 2019) and IFA (IFA, 2019) from 1961 to 2016.

up to 30% underestimation by the Food and Agriculture Organization of the United Nations (FAO) data source, leading to different global fertilizer consumption estimates from the two data sources in recent decades (Figure 5). Additionally, if the N budget assessment focuses on cropland only, then the fraction of synthetic N application used for cropland versus pastures and other noncroplands may become important in determining the N inputs (Lassaletta, Billen, Grizzetti, Anglade, et al., 2014; Xu et al., 2019). This fraction ranges from 12% to 100% among countries (Lassaletta, Billen, Grizzetti, Anglade, et al., 2014) and is likely to decrease when a country becomes more developed (e.g., more fertilizer used for turf-grass) or has more pasture and agro-forestry.

In addition to some disagreement on the national scale from IFA and FAO sources, major gaps in estimating synthetic fertilizer inputs exist between the farm and national scales. Accurate spatially explicit fertilizer input data are urgently needed for nutrient budgeting on subnational or watershed scales, as well as biogeochemical modeling (Lu & Tian, 2017). To arrive at the spatially explicit fertilizer input data, one can upscale the farm-level survey or downscale the national or subnational scale statistics. However, the upscaling method requires farmers to report their fertilization rates, which concerns many farmers due to possible regulation (Osmond et al., 2015; Perez, 2015). The method may also suffer from sampling biases. On the other hand, the downscaling method relies on the distribution of cropland area and assumptions made for fertilizer distribution, both introducing biases in the estimation. For example, Lu and Tian (2017) used the HYDE 3.2 global cropland distribution map to estimate the distribution of N fertilizer inputs globally. Taking India as an example, however, the cropland area from HYDE 3.2 product is over 15% higher than a cropland distribution map from Tian et al. (2014) and about 10% lower than a new Landsat-Based cropland map (GFSAD, 2019; Oliphant et al., 2019). Some downscaling was carried out based on the assumption that fertilizers were applied in the state (or a subnational political unit) where they were sold or adjacent states (Fixen et al., 2012), while others assume a constant fertilizer application rate for all crops (Nishina et al., 2017) or all states (Houlton et al., 2019; Zhang, Davidson, et al., 2015). Only a few efforts have downscaled the fertilizer application rate considering the heterogeneity among regions and crop types (Cao et al., 2018).



**Figure 6.** N (a) and P (b) contents of 11 crop or crop groups. N contents are from five data sources (Bodirsky et al., 2012; Bouwman et al., 2017; Feedipedia, 2012; IPNI, 2014; Lassaletta, Billen, Grizzetti, Garnier, et al., 2014), and P contents are from seven data sources (AUSNUT, 2013; Bouwman et al., 2017; FAO, 2006; Feedipedia, 2012; Gourley et al., 2010; IPNI, 2014; USDA, 2013).

The gaps between the upscaling and downscaling approaches demonstrate one of the major challenges in balancing the nutrient budget on watershed scales and other subnational scales (Table S4).

#### 4.2. Nutrients in Crop or Animal Products

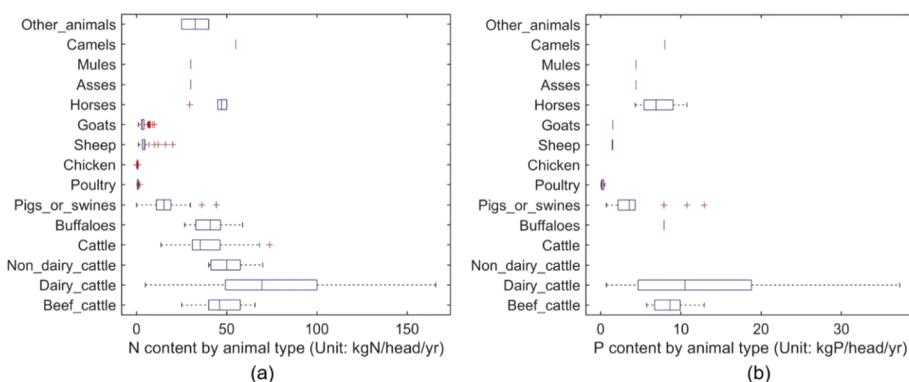
Nutrients contained in crop or animal products are essential outputs from most systems examined, are determining factors for NuUE of the system, and are important inputs for several systems (e.g., *Animal* system and *Agro-Food* system) as the feed or imported products (Figure 4).

While the nutrient contents in crop and animal products could be sampled and directly measured in studies on a farm scale, their quantification on broader spatial scales usually relies on the assumed concentrations of nutrients (kg nutrient per kg product) and the recorded product quantity converted to mass (Billen et al., 2014; Lun et al., 2018). However, those nutrient concentrations may vary among regions or even within the same region associated with different varieties and management practices (Guardia et al., 2018). Taking N and P contents in crop products as an example, Figure 6 demonstrates differences of nutrient contents for a crop type or within a crop group using several commonly used data sources (AUSNUT, 2013; Bouwman et al., 2017; FAO, 2006; Feedipedia, 2012; Gourley et al., 2010; IPNI, 2014; USDA, 2013). In addition to the spatial variation, the nutrient contents also change along time as yield changes or cultivars shift. Ciampitti and Vyn (2012) suggested that the N content in maize has decreased from  $0.0133 \text{ kg N kg}^{-1}$  grain in the period of 1940–1990 to  $0.0120 \text{ kg N kg}^{-1}$  grain in the period of 1991–2011 on average. This 10% decrease in N contents suggests that N surplus reduction in the United States estimated based on a constant N content (Zhang, Davidson, et al., 2015) may be overestimated by about  $15 \text{ kg N ha}^{-1}$ . On the other hand, Tenorio et al. (2019) estimated that yield differences explained much more variability in N surplus than variation in N concentration of harvested grain. Unfortunately, few records are available to track this critical change for most crops.

Depending on the data sources, the product quantity estimated for the same spatial boundary (e.g., a nation) may not be consistent among studies. For example, maize yield in China is estimated as  $5.5\text{--}8.9 \text{ tons ha}^{-1}$  based on intensive farm surveys but is reported as  $4.7\text{--}5.8 \text{ tons ha}^{-1}$  according to the government census (Table S4; NBSC, 2019; Zhang, Ju, et al., 2019). Using data from those different sources may help with assessing and constraining the uncertainties associated with this nutrient budget term.

#### 4.3. Nutrients in Manure

Manure excreted during animal production is one of the major pathways by which nutrients are lost to non-agricultural soils or water, causing pollution (Cordell & White, 2014). Part of the manure is recycled to the *Soil-Plant* system, accounting for about 18% and 28% of the global N and P inputs to cropland, respectively (Figure 4). Nutrients in manure could be quantified by direct laboratory analyses of manure or calculated based on animal stocks and documented nutrient excretion rates ( $\text{kg nutrient head}^{-1} \text{ year}^{-1}$ ; Billen et al., 2014; Bouwman, 1997; Bouwman et al., 2017; Hou et al., 2016; Lassaletta et al., 2019). To measure the



**Figure 7.** N (a) and P (b) contents in the manure of 15 common animal groups. N contents are from five data sources (Bouwman et al., 2017; FAO, 2018a; Lorimor et al., 2004; Mosier et al., 1998; Van der Hoek, 1998), and P contents are from two data sources (Bouwman et al., 2017; Lorimor et al., 2004). The small variations among data sources for several animal types are likely caused by lack of measurement.

nutrient excretion rate in a lab, MidWest Plan Service provides detailed information on laboratory selection, sampling, and testing for N, P, and K (Lorimor et al., 2004). N excretion rate can also be estimated using the difference between the animal's total N intake and total N needed for growth and milk production (Dong et al., 2006) or assumed to be proportional to the weights of slaughtered animals (Sheldrick et al., 2003; Lassaletta, Billen, Grizzetti, Anglade, et al., 2014).

Similar to nutrients in crops and animal products, quantifying nutrients in manure depends on knowledge of nutrient concentrations and manure mass, which vary by factors such as year, region, weather, animal type, diet, animal age, treatment, storage, and calculation method (Figure 7). However, it is usually assumed that nutrient contents do not change over time and constant nutrient excretion factors per animal are applied at the large region scale (Van der Hoek, 1998). Those assumptions and parameters used for the estimation lead to uncertainties in the quantification. For example, the default N excretion rates provided by Intergovernmental Panel on Climate Change have uncertainty about  $\pm 50\%$  (Dong et al., 2006).

Recent efforts have been devoted to developing country- and livestock-specific manure N parameters (e.g., the development of the Global Livestock Environmental Assessment Model; FAO, 2018a, MacLeod et al., 2017). Regularly measuring and reporting manure nutrient concentrations, excretion rates, and the fate of manure by animal type and region will help to improve the quantification of nutrient budgets in the form of manure.

#### 4.4. Atmospheric Deposition

Atmospheric deposition is a relatively small source of N inputs for most agronomic systems examined by the nutrient budget approach, accounting for 6–14% of the total inputs for agricultural landscape globally (Figure 4), but could be important for the N budget of *Landscape* systems in regions with little agricultural activity. For example, N deposition accounts for about 35% of net anthropogenic N inputs to watersheds in the northeast region of the United States. (Hong et al., 2013). Inputs of P through atmospheric deposition are usually minimal and are often neglected in budgets (Mogollón et al., 2018).

The quantification of atmospheric N deposition only accounted for N in rainfall in the early twentieth century (Lipman & Conybeare, 1936) and has evolved to include both wet (e.g.,  $\text{NO}_3^-$  or  $\text{NH}_4^+$  in rain or snow) and dry (e.g., reactive N gas or particles) deposition in recent decades (Fenn et al., 2008; Hember, 2018; Schwede & Lear, 2014; Yu et al., 2019). The wet deposition is often determined by the amount of precipitation and the N concentration in precipitation, which can be directly measured in the field. The measurement techniques have been long established, but Erisman et al. (2005) found that the widely used bulk sampler reported higher deposition flux than wet-only sampler by up to 40%. In addition to the influence of dry deposition, the uncertainties in the measurement could be caused by factors such as the placement of samplers and sample handling and analysis. On a spatial scale larger than an experimental site, the quantification of wet deposition requires the interpolation and/or modeling of existing observations at discrete locations, where the availability of observations and the selection of interpolation methods (e.g., kriging)

affect the uncertainties in the estimated deposition. Models such as the Community Multiscale Air Quality have been used to simulate atmospheric transport, chemistry, aerosol physics, and deposition at a 12-km horizontal resolution as inputs to N budgets (Pinder et al., 2012). Countries in North America, Europe, and East Asia have established national or regional monitoring networks for deposition (Hember, 2018; Xu et al., 2015), but the observation is still fairly scarce in regions such as Africa and South America.

The quantification of dry N deposition is even more challenging and uncertain than the wet deposition, even though its quantity is comparable to wet deposition (Schwede & Lear, 2014; Xu et al., 2015; Yu et al., 2019). Dry deposition can be directly measured with flux towers or chambers for spatial scales of a plot to a farm, but the measurement is limited by (1) available sensors that can provide robust, high-frequency, and high-accuracy measurement of gas concentrations (Erisman et al., 2005; Zhang, Lee, et al., 2015); and (2) logistical resources needed for maintaining a long-term observation. So far, systematic monitoring of dry deposition, which did not start until the end of the twentieth century, is still limited, constraining our understanding of the dynamic processes of gas exchange at the land surface.

In contrast to wet deposition, dry deposition on a regional scale cannot be simply interpolated from point observations, because it is strongly affected by factors such as land surface properties (e.g., surface roughness) and climate (Erisman et al., 2005; Xu et al., 2015). Instead, the regional dry deposition has been estimated as the product of atmospheric N concentration and deposition velocities at the land surface (Flechard et al., 2011; Xu et al., 2015; Yu et al., 2019), where the N concentration can be interpolated or modeled from point observations, and the deposition velocities can be estimated based on modeled meteorological field and land cover map (Holland, Braswell, et al., 2005). However, different models and parameters used in estimating the two variables result in differences in the estimated dry deposition by a factor of 2–3 (Flechard et al., 2011). To improve the quantification of regional dry N deposition, the tropospheric column concentrations of  $\text{NO}_2$  and  $\text{NH}_3$  observed from satellites (e.g., Ozone Monitoring Instrument) have recently been used, in combination with ground observations, to infer  $\text{NO}_2$  and  $\text{NH}_3$  concentration at the land surface (Jia et al., 2016; Yu et al., 2019). While this advancement is promising to improve the spatial and temporal coverage of dry N deposition monitoring, more direct measurements of gaseous and particulate N fluxes over different land surfaces and in different climates are still needed to improve the understanding of the dynamic dry N deposition processes.

#### 4.5. Nitrogen Fixation

N fixation is an important N budget term, estimated to account for about 17% of N inputs in the *Soil-Plant* system and about 32% in the *Agro-Food* system globally (Figure 4). Like denitrification, biological N fixation (BNF) is one of the most uncertain terms in N budgets, owing to the difficulty in measuring it. Several methods have been developed to attempt to measure BNF in a variety of croplands and natural ecosystems (Unkovich et al., 2008). Most plot-level estimates of N fixation by legume crops are based on a method that compares the isotopic signature of shoot tissue of the legume crop with a reference plant (often a nonleguminous “weed”) growing in the same soil (Högberg, 1997). The  $^{15}\text{N}$  content of the reference plant reflects the isotopic signature of the available soil N pool, whereas N fixed from the atmosphere is assumed to have a delta  $^{15}\text{N}$  signature of zero per mil or, in some cases, with a small fractionation correction factor of about  $-1$  per mil (Alves et al., 2006). The percentage of N derived from the atmosphere (%Ndfa) is calculated from measurements of  $^{15}\text{N}$  in the crop shoot tissue and a mixture model using the atmospheric inputs and the soil inputs as  $^{15}\text{N}$  end-members. Estimates can be variable, but they frequently fall in the range of 60–80% of the N in shoots derived from the atmosphere for most leguminous crops (Anglade et al., 2015; Figueira et al., 2016; Peoples et al., 2009). The rate of N fixation is then calculated by multiplying this percentage by the total N stock of the shoots. A multiplier is then used to account for belowground N mass, which often adds another 30–70% to the BNF estimate and is another source of great uncertainty (Anglade et al., 2015).

The %Ndfa method and other methods have also been used to estimate associative N fixation by organisms living in the rhizosphere of some Gramineae crop plants, such as sugar cane, tropical pasture grasses, and rice (Unkovich et al., 2008). The estimated associative N fixation rates are generally lower than leguminous symbiotic BNF and remain somewhat controversial (James, 2000). A mass balance approach has been used to estimate nonsymbiotic BNF in maize, rice, and wheat production systems, where BNF was calculated from the difference of all of the other inputs and outputs (Ladha et al., 2016). The error term of this BNF

estimate includes the errors of all of the other estimated inputs and output of the budget, including denitrification, which is also highly uncertain.

Labeling by incubating nodules in an atmosphere with  $^{15}\text{N}_2$  is a more direct but costly method that is usually performed in the laboratory and then extrapolated to field rates based on estimates of nodule biomass and with assumptions relating laboratory and field conditions. Similarly, the activity of the nitrogenase enzyme can be estimated in laboratory or field incubations using acetylene reduction, adjusted by laboratory-calibrated conversion factors to rates of  $\text{N}_2$  reduction, and then extrapolated to the plot scale (Cleveland et al., 1999).

Symbiotic BNF has been estimated in nonagricultural ecosystems by measuring N fixation rates with the acetylene reduction method and extrapolating those rates using estimates of plot-level nodule biomass or metrics on the abundance of leguminous tree species known to nodulate (Batterman et al., 2013; Sullivan et al., 2014). The %Ndfa method has also been applied to nonagricultural soils (Davidson et al., 2018). Fixation by free-living microorganisms in soil is usually a small but sometimes important input to nonagricultural ecosystems. It has usually been measured by the acetylene reduction method and scaled to ecosystem or even biome levels based on estimates of net primary productivity or evapotranspiration (Cleveland et al., 1999).

Regional and global bottom-up estimates of N fixation are usually based on assumed average rates of BNF by each leguminous crop and by scaling BNF in nonagricultural ecosystems using relationships with net primary productivity or evapotranspiration (e.g., Howarth et al., 2012). Mechanistic models of N fixation that include other environmental controls, such as nutrient limitations of N and P and temperature responses, have been applied at biome and global scales (Houlton et al., 2008). Mass balance and isotopic discrimination have been used to estimate BNF on a global scale (Vitousek et al., 2013). Despite these multiple approaches developed over decades, N fixation estimates remain poorly constrained in most N budgets.

#### 4.6. Leaching and Runoff

Runoff and leaching are the major pathways that nutrients escape the agricultural land and contaminate water bodies. N and P in runoff and leaching can be measured directly in the field. To study nutrients in soil solution at different depths and its transport, there are sampling tools such as suction cups, lysimeters, ion-exchange resins, and groundwater wells (Lehmann & Schroth, 2003; Snyder, 1996). The nutrient loss in soil could also be measured using tracers, such as  $^{32}\text{P}$ ,  $^{33}\text{P}$ , and  $^{15}\text{N}$  (Lehmann & Schroth, 2003). To estimate N and P loss from cropland to water bodies for a larger scale (e.g., watershed scale), researchers can measure water flow (discharge) and nutrient concentration to determine the nutrient load, which includes discharge measurement and water sampling processes (Deelstra & Øygarden, 1998). When there are no discharge measurements, the annual rate can be estimated using climatological data (Deelstra et al., 1998). The choice of methods depends on factors such as project scale, project purpose, soil conditions, and cost (Lehmann & Schroth, 2003; Snyder, 1996).

Another way to estimate leaching and runoff is through modeling, especially for large-scale studies (e.g., global scale, Bouwman et al., 2017). The model can also be used to study how effective different best management practices (BMPs) are on reducing the nutrient loss (Borah & Bera, 2003). Different methodologies and different temporal and spatial scales have been adopted by different models, making it difficult to compare results across models and to evaluate uncertainties. For example, Mekonnen and Hoekstra (2018) estimate P load as the product of the P surplus of the *Soil-Plant* system and a spatially explicit erosion-runoff-leaching fraction; while the Integrated Model to Assess the Global Environment-Global Nutrient Model (Beusen et al., 2015; Bouwman et al., 2017) not only considers the P inputs as fertilizer and manure for the current season but also considers the P reserves in soil accumulated in previous years or decades. Comparing these and other modeling studies, the loads of global P in runoff and leaching to freshwater range from 0.16 to 5 Tg P year $^{-1}$  (Bouwman et al., 2009; Bouwman, Goldewijk, et al., 2013; Mekonnen & Hoekstra, 2018; Penuelas et al., 2013; Seitzinger et al., 2005).

Where direct measurements or models are not available, nutrient loss in leaching and runoff has been estimated with simple assumptions. For example, P in leaching is sometimes assumed as a fixed fraction of P budget terms, such as 12.5% of P inputs (Bouwman, Goldewijk, et al., 2013; Lun et al., 2018) or 20% of P surplus (Springmann et al., 2018). These simple assumptions introduce large uncertainties into results. For

example, global P leaching and runoff from cropland estimated by Lun et al. (2018) is 20% lower than that by Bouwman, Goldewijk, et al. (2013), whereas the estimates for pasture by Lun et al. (2018) is 60% higher than Bouwman et al. (2009).

#### 4.7. Gaseous Emission

While virtually no P exists in gaseous phase, N emission in the gaseous phase is an important part of the N budget for most systems. One of the major gaseous phase losses of N from terrestrial and aquatic ecosystems is in the form of N<sub>2</sub> gas as the end product of denitrification. Although N<sub>2</sub> emission does not have direct negative impacts on the environment, it is critical to quantify it for balancing the budget, and it represents N that might have been retained and recycled within the system for productive outputs. However, direct measurement of N<sub>2</sub> emission to the atmosphere from soils and water bodies has been challenging due to the high background concentrations in the atmosphere and high spatial and temporal variation in the emission (Davidson & Seitzinger, 2006). Attempts have been made to estimate N<sub>2</sub>O:N<sub>2</sub> emission ratios in the laboratory and then apply an average ratio to field-based N<sub>2</sub>O emission measurements to estimate N<sub>2</sub> emissions (Schlesinger, 2009), but the reported ratios are highly variable and the uncertainties using this approach are large due to several soil environmental factors (Houlton et al., 2013).

Ammonia (NH<sub>3</sub>), nitric oxide (NO), and nitrous oxide (N<sub>2</sub>O) are three other major gaseous emissions that negatively affect the environment (Davidson et al., 2011), including climate change and stratospheric ozone depletion (N<sub>2</sub>O), downwind soil acidification (NO), formation of fine particulate matter that poses respiratory health risks (NH<sub>3</sub> and NO), and a precursor (NO) to formation of tropospheric ozone and nitrogen dioxide, which are also human respiratory health risks. With advances in gas analyzers and micrometeorological methods (e.g., soil chambers and micrometeorological towers), the emissions of all three gases have been measured on plot scales. Based on the plot-scale measurement, emission factors (e.g., IPCC, 2006), inventories (e.g., Davidson & Kanter, 2014; Houlton et al., 2013), and biogeochemical models (e.g., Del Grosso et al., 2005; Tian et al., 2019) have been developed to estimate emissions from agricultural and nonagricultural soils and from other sectors (e.g., industry and biomass burning) at regional to global scales. However, given large heterogeneity of emissions among locations, time periods, and management practices (Eagle et al., 2017), this upscaling, bottom-up approach inherently includes considerable uncertainties.

To evaluate and constrain those uncertainties, observations of NH<sub>3</sub> and N<sub>2</sub>O emissions on regional and global scales have been developed. The global N<sub>2</sub>O emission from anthropogenic activities has been derived from inverse modeling of temporal and spatial variation in atmospheric N<sub>2</sub>O concentrations (top-down approach, Thompson et al., 2019; Prather et al., 2015). At broad global scales, the bottom-up and top-down approaches are in reasonable agreement, suggesting that agricultural sources account for two thirds or about 4.1 Tg N-N<sub>2</sub>O year<sup>-1</sup> of total anthropogenic emissions (Davidson & Kanter, 2014). However, the agreement between the two approaches has not always been reached on regional scales. For example, Griffis et al. (2013) and Zhang et al. (2014) found that the N<sub>2</sub>O emission estimate from top-down atmospheric observation was about 2 times that of the estimate from bottom-up Intergovernmental Panel on Climate Change emission factors for United States corn belt region, suggesting underestimated emission factors for the region or unaccounted sources in this agriculture-dominated landscape. Top-down inverse modeling approaches are now being applied to continental and subcontinental scales (Thompson et al., 2019).

In addition to atmospheric measurement of gas concentration, advances in satellite observations provide another opportunity to validate and constrain NH<sub>3</sub> and NO<sub>x</sub> emission on regional to global scale (Van Damme et al., 2018; Zhang et al., 2018). For example, a gridded NH<sub>3</sub> emission map was derived from the Infrared Atmospheric Sounding Interferometer satellite observation, and early results suggest that the bottom-up inventory (Emission Database for Global Atmospheric Research) may underestimate NH<sub>3</sub> emission by over 1 order of magnitude for many agricultural emission hot spots. Miyazaki et al. (2017) estimated global surface NO<sub>x</sub> emissions from the assimilation of multiple satellite data sets, including tropospheric NO<sub>2</sub> columns, and profiles of O<sub>3</sub> and HNO<sub>3</sub>.

#### 4.8. Nutrient Storage or Depletion in Soil

Soil is one of the major nutrient stocks of a system that can increase or become depleted. Part of the P applied to crops has been shown to be retained in agricultural soils during multiple years of P application (Le Noë et

al., 2018; Rowe et al., 2016; Sattari et al., 2014). This increased soil-P stock can become a source of P for future years, which is called the P legacy. This is the case of some European countries where decades of P overfertilization built up a large soil-P stock. Today, yields are often maintained even after a total elimination of P fertilization (Bouwman et al., 2017). Some soils, particularly those in tropical areas with highly weathered soils, are P-fixing because phosphate binds to iron and aluminum oxides (Roy et al., 2016). In the areas with high proportions of P-fixing soils, P recovery by crops can be low and P surpluses can be high, with most of the P surplus retained in the soil. The release of P fixed on iron and aluminum oxides may be slower than the release of P accumulating in less highly weathered soils in temperate regions. Therefore, the consideration of the P legacy effect is different when compared with soils that are “non-P-fixing.” P legacies can also play a role after agricultural activities cease, influencing P budgets in the postagricultural ecosystems (MacDonald et al., 2012).

The legacy effect is probably less common for N than for P, but N can also accumulate in soils. When soil N is at or near steady state, mineralization and immobilization of soil N are roughly in equilibrium. However, where soil C sequestration is occurring (e.g., conservation tillage), part of the N surplus will be retained with the C sequestration, promoting the stabilization of soil N through microbial turnover (Lin et al., 2016). van Groenigen et al. (2017) suggested that significant C sequestration at global scales (e.g., if the 4% initiative is achieved; Francaviglia et al., 2019) would consume a large fraction of N surplus. However, the N can also be lost if soil organic matter is later lost through erosion or net decomposition due to changing management practices. Van Meter et al. (2016) have introduced another N legacy concept, suggesting that N accumulated in agricultural soils during many years' fertilization can continue to affect stream and coastal water quality for several years to follow. This could be a contributing reason why the water quality in the Gulf of Mexico has not improved in the short term despite considerable mitigation efforts (Van Meter et al., 2018). A negative legacy effect results when N is mined from soils where inputs are insufficient (David et al., 2001) or where organic soils are drained (Van der Pol, 1992); soil fertility and productivity may decline as the result.

Changes of soil nutrients could be quantified by measuring nutrient contents in surface soil layers or soil profiles (Chen et al., 2017; Van Meter et al., 2016; Yan et al., 2014). However, the direct measurement approach faces major challenges to accurately quantify changes in soil nutrient stocks. Both nutrient concentrations (e.g., mg N g<sup>-1</sup> dry soil) and bulk density (g dry soil cm<sup>-3</sup> soil) must be accurately measured (Davidson & Ackerman, 1993). Unfortunately, bulk density measurements are commonly missing. Spatial heterogeneity of soil nutrient content is significant in both horizontal and vertical directions and can be affected by soil management practices such as tillage and fertilization (Christianson et al., 2012; Van Meter et al., 2016). Furthermore, short-term changes in nutrient stocks are usually small compared to the total stock (Yan et al., 2014). Therefore, the sampling error may be much larger than the actual change of nutrient stock in the soil. To quantify soil nutrient changes on plot to regional scales, more long-term site experiments and large-area soil sampling are needed, but both require significant human resources and financial support.

In addition to direct measurement of soil nutrient contents, isotope methods and modeling methods have been developed to estimate changes in soil nutrients. For example, Figueira et al. (2016) determined changes in inputs, outputs, and soil N stock using natural <sup>15</sup>N enrichment in the soils sampled from a chronosequence of soybean fields in Brazil. A simplified two-pool (labile and stable pools) P model, the Dynamic Phosphorus Pool Simulator (DPPS), has been developed to estimate P legacies from regional to global scales (Lun et al., 2018; Mogollón et al., 2018; Zhang et al., 2017). Other researchers used soil P dynamics models with more than two P pools (Goll et al., 2012; Ringeval et al., 2017; Wang et al., 2009; Yang et al., 2013) to study the distribution of soil P. Due to lack of data and understanding, it is difficult to compare and validate soil P pool estimates from different models.

## 5. Challenges in Informing Nutrient Management

The quantification of nutrient budgets has provided invaluable information for nutrient management. For example, the NUE and nutrient surplus for a *Soil-Plant* system on a farm scale can help farmers understand changes in soil nutrient stocks and estimate the fertilization needs for the next season. The quantification of nutrient footprints for the *Agro-Food* system informs individual consumers or institutions (e.g., a university) about the impacts of their day-to-day choices (e.g., dietary and energy choices) on nutrient pollution (Leach

et al., 2016). However, using nutrient budgets to inform sustainable nutrient management faces multiple major challenges.

### **One of the major challenges is to define and assess “sustainable” nutrient management with nutrient budgets**

Nutrient management affects multiple sustainability targets, involving complex trade-offs among stakeholders and across spatial scales. Nitrogen-efficient technologies on a farm scale do not necessarily lead to the reduction in nitrogen fertilizer use or nitrogen pollution for the region, given the complex feedbacks in the market and other socioeconomic factors (Zhang, Mauzerall, et al., 2015). Importing food from regions with higher NUE relieves nitrogen pollution in the importing countries and the world but adds the burden of more pollution to the regions exporting food (Huang et al., Oita et al., 2016; Zhang, 2017). Therefore, whether a nutrient management practice or food policy should be deemed as sustainable needs to be assessed on multiple spatial scales and is affected by the regional priorities in pursuing sustainability.

The quantification of nutrient budgets of various systems and spatial scales provides great opportunities for assessing impacts of nutrient management on multiple scales. Quantifying and connecting nutrient budgets across scales enables systematic evaluation and monitoring of nutrient management effectiveness. The importance of cross-scale assessments was clearly shown by Bai et al. (2014, 2018) for Chinese livestock systems. Increases in NUE were reported when estimated at the farm scale (herd scale) but when including the whole agro-food system NUE decreases were observed. Multi-system and multi-spatial scale approaches involving structural changes for alternative sustainable pathways at the system level are needed to improve sustainable use of manure by promoting reintegration of crop and livestock production systems (Garnier et al., 2016, Zhang, Liu, et al., 2019), to improve crop NUE through an optimal spatial allocation of fertilizers (Mueller et al., 2014, 2017; Zhang et al., 2018), to analyze the effect of dietary changes (Westhoek et al., 2014), or to combine supply- and demand-side actions (Bodirsky et al., 2014; Desmit et al., 2018; Lassaletta et al., 2016).

In addition to the cross-scale assessment, it is important to evaluate the impacts of nutrient management on other sustainability targets, such as food security and land use change. Integrating the efficiencies of nitrogen use (i.e., NUE) and land use (i.e., yield) on a national scale, a Sustainable Nitrogen Management Index has been developed and applied to measure the sustainability of agricultural production on the national scale (Sachs et al., 2016; Schmidt-Traub et al., 2017; Zhang & Davidson, 2019). However, this index is still limited in the scope of sustainability targets included in the assessment. To address this limitation, many ongoing efforts are devoted to develop indicators for assessing agriculture sustainability from environmental and socioeconomic perspectives and on various spatial scales (FAO, 2018b; Zhang, Liu, et al., 2019).

### **The second challenge is to connect actions by stakeholders with their impacts on the environment**

Actions by stakeholders, such as consumers' dietary choices and farmers' choice of fertilizer management practices, are usually linked to the nutrient inputs and/or the efficiencies of the system directly; while the environmental impacts are best assessed by the damage to ecosystem and human health caused by nutrient loss in the form of pollutants. The environmental impacts are not only affected by the amount of nutrient loss quantified in a nutrient budget but also the timing and location of the loss, as well as many ecological factors, such as climate and soil conditions. Therefore, many environmental models have attempted to connect the stakeholder activities with the environmental impacts by simulating ecological processes involved in the emission, transformation, and transportation of the nutrient pollutants (Beusen et al., 2016; Desmit et al., 2018, Malagó et al., 2019, Schroeck et al., 2019). However, the wide adoption of environmental modeling approaches by stakeholders is limited by the regions or conditions where the models can be calibrated with data and by the transparency of the modeling inputs and processes (McLellan et al., 2018).

In contrast to the attempt in modeling the complex environmental impacts from nutrient budgets, several relatively simple indicators have been developed based on nutrient budgets. For example, N surplus (also called N balance) of the *Soil-Plant* system is directly affected by farmers' management practices and is proven to be significantly related to yield-scaled N losses when aggregated over multiple sites and years (McLellan et al., 2018; Sela et al., 2019). However, it should be recognized that the N surplus is not always the best predictor of the actual environmental impacts of an individual farm for a given year, which is also largely

affected by factors such as year-to-year weather variations and the legacy effects of N in the soil. Similarly, N and P footprints are designed to connect consumers' dietary choices with broad-scale estimates of N and P pollution; however, most footprints are calculated with parameters generalized for a country and consequently limited in reflecting the different environmental impacts caused by different producers' choices in nutrient management practices. Where identifying the specific, near-term local impacts of the farmer or consumer decisions is desired, then more measurements and models tailored to those specific circumstances are needed.

#### **The third major challenge is to effectively communicate nutrient budgets to engage actions**

Simple and transparent messaging is critical for effective communication with stakeholders, especially for consumers. However, nutrient budgets, especially for N and P, are complicated with varying uncertainties, and the complex trade-offs among various sustainability goals and across scales further complicate calls for action. Consequently, a delicate balance needs to be reached to enable effective public communication with scientific rigor, and sometimes compromises need to be made considering the regional or global priorities.

Effective engagement of stakeholders for sustainable nutrient management should also go beyond the traditional "one-way" communication, namely from researchers to stakeholders. Involving stakeholders in the design and development of nutrient budgeting measurements and estimates and its communication products, such as indicators or decision-support models, is an important process for building trust and collaborative working relationships between researchers and stakeholders. It may also improve the accuracy of the nutrient budget terms and is a critical step toward engaging more stakeholders to incorporate science-based nutrient budget research in their decision making (Wall et al., 2017). However, few researchers and students have been trained to work effectively at the interface of science and communication (Safford et al., 2017), and the efforts in translating science to practices need to be better valued (Chapin, 2017).

Improving the accuracy of nutrient budgets is a worthy scientific pursuit, but communicating the meaning and significance of nutrient budgets for ecosystem and human health and their economic implications is paramount. Describing changes as accurately as possible in terms of teragrams (or any other units) of N or P per year is necessary for scientific integrity and to form the basis of a case for concern, but explaining the impact of changes in nutrient budgets goes beyond quantification of stocks and fluxes. Human alteration of the N budget has been reported by scientists for several decades (Crutzen, 1970; Davidson et al., 2011; Fowler et al., 2013; Galloway et al., 2003; Vitousek et al., 1997), but widespread appreciation within the policy community for the seriousness of the topic accelerated after the issue was described in terms of nitrogen excess exceeding a proposed planetary boundary of a "safe operating space for humanity" (Rockstrom et al., 2009). A crucial remaining challenge will require the convergence of biogeochemistry and agronomy with the social sciences in order to translate documented biophysical changes of nutrient budgets to quantitative or qualitative explanations of their impacts on economics, human health, and ecosystem functions that humans value.

## **6. Concluding Remarks**

Managing nutrients is fundamental to sustainable development in this century, and it requires good information about nutrient budgets. The growing efforts in quantifying nutrient budgets for various systems and across different scales have enabled researchers to understand complex nutrient cycles and their interactions with human and Earth systems. Those efforts have also provided useful information for many stakeholders, including farmers, retailers, consumers, and policy makers, to assist their decisions that affect nutrient management. However, the expanding application still requires consistent and structural definitions of systems and their budget terms to enable comparisons and interlinkages among studies; large uncertainties in multiple nutrient budget terms need to be constrained; and the scientific findings need to be better communicated to engage positive changes.

By reviewing nutrient budget studies in five systems, namely *Soil-Plant* system, *Animal* system, *Animal-Plant-Soil* system, *Agro-Food* system, and *Landscape* system, this paper provides an initial attempt toward a consistent framework for nutrient budget analyses. The review of the quantification challenges reveals the varying uncertainties in major nutrient budget terms across spatial scales. The levels of challenge in

constraining those uncertainties also vary largely among budget terms and scales: some already have well-established methods for measuring and validating, while others still rely on very crude parameters developed from few studies or observations and require the development of basic observation approaches. To address the quantification challenges, advances in monitoring both ecological and socioeconomic changes are needed. Recent developments in remote sensing, sensor technologies, and isotope methods provide opportunities to overcome difficulties in monitoring nutrient flows on a variety of spatial and temporal scales. Increasing interest in quantifying nutrient budgets by governments and farmers could potentially motivate more resources toward improving the collection of human activity data related to nutrient budgets (e.g., fertilization rate and food trade). To take advantage of those opportunities, interdisciplinary modeling and data synthesis approaches need to be developed.

The current limitations in nutrient budgets and the need for their improvement should be better communicated not only among researchers but also stakeholders engaged in nutrient management. It will take decades, even centuries, for all nutrient budget terms to be accurately accounted for, but this should not prevent scientists from communicating the importance of nutrient budgets and the insights they provide for sustainable management.

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