Half-century history of crop nitrogen budget in the conterminous United States: Variations over time, space and crop types

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Key Points:

- We synthesized county-level crop nitrogen (N) budget data of eight major crops in the United States during 1970-2019
- The national crop NUE increased from 0.55 kg N kg^{-1} N in the 1970s to 0.65 kg N kg^{-1} N in the 2010s, with an average NUE of 0.59 kg N kg^{-1} N
- N surplus accounts for 41% (27%-55%) of the national total N input, with a significant decline detected during the recent decade

Abstract

Spatiotemporal patterns of crop nitrogen (N) budget have important implications for agricultural N management and environmental policy. Previous studies examined crop N budget in different countries but often overlooked cross-crop differences at sub-national scales. In this study, we synthesize multiple databases to examine the N budget of eight major crops in the United States at the county scale during 1970-2019. Our analyses show that national crop NUE increased from 0.55 kg N kg^{-1} N in the 1970s to 0.65 kg N kg^{-1} N in the 2010s. Four out of eight crops such as corn, rice, cotton, and sorghum demonstrated an increasing NUE trend during the study period, whereas the other crops overall presented a declining NUE trend. Nationwide, about 41% of the total N input was not used by these crops (N surplus) over the study period, of which temporal variation was mainly driven by corn due to its large planting area and high N input. The national N surplus first increased in the 1970s and remained relatively stable till the 2000s. Since the early 2010s, however, N surplus began to decline and approached the levels in the early 1970s—an encouraging development that may lead to decreased N pollution to the environment. The hotspots of national N surplus coincided with corn- and rice-producing counties. The sub-national variations and temporal dynamics in crop N budget revealed in this study highlight the urgent need to understand the farm-level crop N balance and the dominant factors controlling crop NUE for mitigating N pollution.

1 Introduction

The world agriculture system has been altered substantially to meet the rapid increase in global food demand (Lassaletta et al., 2016). Since the 1960s, global nitrogen (N) fertilizer use has increased nine-fold (Lu & Tian, 2017). During the period from 1984 to 2016, global inorganic N deposition increased by 8% from 86.6 to 93.6 Tg yr^{-1}, with a significant increase in East Asia and

South America and a large decline in Europe (Ackerman et al., 2019). An overall estimate of 50–70 Tg N yr^{-1} is fixed by legume plants in global agricultural systems (Herridge et al., 2008). Over two-thirds of the total anthropogenic N inputs (e.g. N fertilizer, atmospheric deposition, and fixed N by legume crops) on the global land surface were reported to return to the atmosphere, percolate to the surface and subsurface water bodies, or transport to the ocean (Cao et al., 2020; Cassman et al., 2002; Schlesinger, 2009; Tilman et al., 2002). The proportion of N that is not utilized by crops (i.e., the difference between N inputs and N harvested in crops), usually defined as N surplus, was found to account for nearly 40% of global total N inputs (Liu et al., 2010). Tremendous N surplus induced by excessive N input can be potentially lost to the environment. It can be emitted into the atmosphere, causing green-house effect (i.e. N_2O) and air pollution (i.e. NH_3 as precursors to PM2.5), re-deposited onto sensitive ecological communities, or delivered to groundwater and surface water, imposing detrimental impacts on the biodiversity and functioning of recipient ecosystems (Barak et al., 1997; Bouwman et al., 2002; Kumazawa, 2002; Van Meter et al., 2017).

Crop nitrogen use efficiency (NUE)—the harvested N in crop production per unit N input—has been proposed as an important indicator for agricultural sustainability in many countries (Norton et al., 2015). Improving crop NUE has been perceived as one of the most effective means for alleviating environmental degradation while increasing or maintaining crop productivity (Davidson & Kanter, 2014; Giller et al., 2004; Howarth et al., 2002). Therefore, it is of critical importance to understand crop NUE from both agronomic and socio-economic perspectives, addressing the challenges of food security, environmental degradation, and climate change (Zhang et al., 2015).

A wide variety of studies analyzed crop NUE trends at various spatial and temporal scales, revealing large disparities of crop NUE among countries (Lassaletta et al., 2014, 2016). Crop NUE was found to level off or even decline in developing countries as N input increases (Ciampitti & Vyn, 2014; Lassaletta et al., 2014; Zhang et al., 2015). This implies that increasing N input can hardly further stimulate crop yield gain, but instead leads to a greater N surplus. For example in China, the grain was only enhanced by 28% with an increase of N fertilizer consumption by 54% (Meng et al., 2016). However, the crop NUE in North America was reported to be higher than that in other regions around the globe and maintained an increasing trend as N input was raised (Lassaletta et al., 2016; Zhang et al., 2015). As of 2007, the United States national crop NUE was reported close to 55% with all intentional and unintentional N inputs entering food, livestock feed, biofuel, and industrial products counted (Houlton et al., 2013). Although previous studies have advanced our understanding of crop NUE, sub-national patterns and dynamics of crop-specific NUE are generally overlooked due to limited data availability. Besides, crop-specific tendencies in using N have not been explicitly examined because N input data for different crop types are either missing or discontinuous in the majority of countries. Using publicly available data sets, Sabo et al., (2019) compiled the inventories of N inputs and outputs as well as terrestrial N surpluses for all sub-basins of the contiguous U.S. and found that the national-scale N budget experienced little changes (±6%) between 2002 and 2012. By using county-level data, a recent study developed an 88-year (1930–2017) data set of N mass balance across the contiguous U.S. (Byrnes et al., 2020), but cross-crop variations were not examined. As prior work to this study, Lu et al. (2019) investigated state-level crop-specific NUE, reporting that the NUE of corn and winter wheat had a large spatial heterogeneity across the U.S. and their responses to N enrichment slowed down in the recent two decades. Therefore, it is important to understand the spatiotemporal patterns of N budget and NUE across various crop types for developing better crop nutrient management practices, creating a nutrient trading system, and reducing nutrient loss to the environment.

In this study, we compiled a multi-source database to characterize the crop-specific N budget for eight major crops in the U.S. from 1970 to 2019. Major crops of interest include barley, corn, cotton, durum wheat, rice, spring wheat, sorghum, and winter wheat. We extracted county-level data of major N input sources and crop yield to calculate crop-specific NUE and N surplus. We assessed how the crop NUE and N surplus changed along with crop yields and N input rates. We then examined the temporal changes of crop-specific NUE and N budget at the national scale. The spatial heterogeneity of crop-specific NUE and N surplus was also quantified at a county level over the past five decades. Specifically, we aimed at answering the following questions in this study: (i) How have crop yields and NUE responded to N input dynamics in the U.S. since 1970? (ii) How have the crop-specific NUE and N surplus changed over space and time? (iii) What are the relationships between the N surplus, crop yield, and N input across these eight crops?

2 Data and Methods

We obtained historical county-level crop yield data of barley (*Hordeum vulgare* L.), corn (*Zea mays* L.), cotton (*Gossypium hirsutum* L.), durum wheat (*Triticum durum* Desf.), rice (*Oryza sativa* L.), spring wheat (*Triticum aestivum* L.), sorghum (*Sorghum bicolor* L.), and winter wheat (*Triticum aestivum* L.) from the USDA National Agricultural Statistics Service (NASS) crop databases (http://www.nass.usda.gov/index.asp). Our published crop-specific fertilizer management database (Cao et al., 2018) has been updated and further refined from the state level to the county level. We also considered other anthropogenic N sources in this study, including manure N application, atmospheric N deposition, and residual N fixation from previous-year soybean rotation for corn only. We analyzed the responses of yield and NUE to N input as well as the input-driven (i.e. N input-driven) and output-driven (i.e. crop yield-driven) N surplus trends from 1970 to 2019. Based on the county-level crop-specific N budget data sets, we further examined their spatial patterns and temporal dynamics.

2.1 Crop yield and anthropogenic N input data sources

Cropland planting area: We obtained the county-level planting areas of eight major crop types from 1970 to 2019 from the USDA-NASS crop database (https://www.nass.usda.gov/). Only the counties with tracked crop areas were included in this study. We gap-filled the planting area of the major crops by assuming the missing county-year samples follow the inter-annual variation in the state-level planting area of each crop (USDA-NASS, https://www.nass.usda.gov/). Except for the eight major crops plus soybean, the rest crop types (e.g., fruits, vegetables, and small grains) were grouped as “Others” and added up to keep the state-level cropland acreage consistent with the USDA-NASS survey.

Crop yield: The annual yield for each crop in each county from 1970 to 2019 was obtained from the USDA-NASS (https://www.nass.usda.gov/). Specifically, we used the year 1970 as the starting year to limit our analyses to a period when private sectors in the U.S. have heavily invested in advanced crop hybrids, agricultural management practices, and crop technologies to improve crop production (Evenson & Gollin, 2003; Tilman, 1998).

Chemical N fertilizer use: We developed annual county-level N fertilizer use data of these eight crops from 1970 to 2019 by combining the state-level crop-specific N fertilizer use rate (Cao et al., 2018; Lu et al., 2019) with the county-level N fertilizer consumption amount obtained from the Nutrient Use Geographic Information System (NuGIS, https://nugis.tfi.org/). Our newly developed dataset differs from the NuGIS data by particularly characterizing the cross-crop variations in N fertilizer use rates at the county level. Specifically, based on the most recent surveys conducted by USDA-NASS, we updated our published state-level crop-specific N fertilizer data to 2019 using the same approach adopted in Cao et al. (2018) and Lu et al. (2019). Since the NuGIS county-level total N fertilizer consumption is only available from 1987 to 2014, we used the inter-annual variations of state-level total N fertilizer consumption (Cao et al., 2018) to gap-fill the periods of 1970-1986 and 2015-2019. The crop-specific N use rate generated above was downscaled from the state- to county-level as Eq. 1:

$$N\ rate_i^{ct} = \frac{N\ cons_{ct}}{\sum_{i=1}^{10} N\ rate_i^{st} \times Area_i^{ct}} \times N\ rate_i^{st} \qquad (1)$$

where $N\ rate_i^{ct}$ is N fertilizer use rate or manure N application rate of crop type *i* in county *ct*, $N\ cons_{ct}$ is annual county N fertilizer consumption or manure N consumption, $N\ rate_i^{st}$ is N fertilizer use rate or manure N rate of crop type *i* in state *st*, and $Area_i^{ct}$ is county-level planting area for crop type *i*. Crops include the abovementioned eight crops, soybean, and "Others".

Manure N application: We reconstructed the county-level crop-specific manure N use rate in the U.S. from 1970 to 2019. We obtained total manure consumption in each county during 1987-2014 from the NuGIS (https://nugis.tfi.org/) and state-level crop-specific manure use rate from the USDA-ERS (https://data.ers.usda.gov/reports.aspx?ID=17883). Assuming that county-level missing data (1970-1986 and 2015-2019) follow the same inter-annual variations as the national manure N use developed by the FAO (http://www.fao.org/faostat/en/#data/EMN), we gap-filled the missing years in the county-level recovered manure data (i.e. the proportion that is applied in fields).

We converted the state-level crop-specific manure use rate to manure N use rate from 1996 to 2010 based on animal manure types and the manure moisture status from the USDA-ERS census data (https://data.ers.usda.gov/reports.aspx?ID=17883) (see manure N content conversion coefficients in Table S1). We downscaled the crop-specific manure N use rate generated above from state- to county-level by using Eq. 1. Unlike fertilizer data, we used the multi-year average of the crop-specific manure N use rate in each state instead of annual values owing to limited data.

Atmospheric N deposition: We obtained the total N deposition data at a 4-km resolution for the period 2000 to 2015 from the National Atmospheric Deposition Program (NADP) total deposition maps (TDEP, http://nadp.slh.wisc.edu/committees/tdep/tdepmaps/, (Schwede & Lear, 2014)). To get the crop-specific N deposition rate for each county, we first resampled the TDEP N deposition into 5-arc-minute maps in ArcMap 10.7 (ESRI, 2018). We then extracted the N deposition data from 2000 to 2015 specifically for the eight crops using time-series crop distribution maps that have the same spatial resolution. The crop distribution maps were aggregated from 1 km land-use maps that were obtained from a recent study (Yu & Lu, 2018). The land-use maps were developed by keeping the county-level harvested area of each crop type in each year consistent with the county-level census records provided by NASS, USDA (http://www.nass.usda.gov/index.asp). Using the TDEP N deposition in 2000 as the endpoint, we back-calculated the gridded N deposition for the period 1970-1999 by taking the annual percentage change rate of the North American gridded N deposition data (Wei et al., 2014). The N deposition from 2016 to 2019 was assumed to remain at the same level as of 2015. Finally, we calculated the average N deposition rate of all the grids in which the same crop type was planted within each county in each year.

N residues from previous-year soybean fixation: Using the Crop Data Layer (CDL, https://www.nass.usda.gov/Research_and_Science/Cropland/Release/index.php), we first identified the pixels in which soybean rotated with any of these eight studied crops in three consecutive years from 2008 to 2017. We included the rotation sequences such as soybean-other crop-soybean, soybean-other crop-other crop, or soybean-soybean-other crop ("other crop" here refers to one of the eight studied crops). We found that 74.4% of the national soybean area was used for rotation with the eight studied crops in 2017. Among them, 66.4% of soybean was rotated with corn, 4.3% with spring wheat, and 2% each with winter wheat and rice. The percentages of soybean rotating with the rest four crop types were all less than 1%, which together accounted for 1.7% of the national soybean used for rotation purposes. Therefore, in our study, we only considered the residual benefit of N fixation from the previous year's soybean cultivation for corn N budget. In addition, there are a few data points available to indicate the national acreage percentages of soybean rotated with corn, for example, 93% in 1993 (United States Department of Agriculture, 1994), 91% in 1997 (Padgitt et al., 2000). Soybean used in corn rotation became firmly established in the U.S. around 1940 (North Carolina Soybean Producers Association, 2019). Hence, we assumed that 100% soybean was used in corn rotation in 1940. Based on these available historical data points, we used linear interpolation to gap-fill the area percentages of soybean rotating with corn in the missing years from 1970 to 2008. Then the county-level soybean percentages and yields were used to calculate the residual N for corn in each county each year. The details of the residual N from previous-year soybean fixation can be found in Lu et al., (2019).

2.2 Calculation of county-level crop-specific NUE and N surplus

The NUE was calculated as a ratio of crop-recovered N to total N input in units of kg N kg^{-1} N (also referring to a percentage (%) of total N input). The crop-recovered N, indicating how much N is retained per unit crop yield, was calculated as crop yield multiplied by a crop N recovery coefficient. We adopted the N recovery coefficients from the NuGIS, which is 0.012 kg N kg^{-1} grain for corn and sorghum, 0.0194 kg N kg^{-1} grain for winter wheat, spring wheat, and durum wheat, 0.011 kg N kg^{-1} grain for rice, 0.02 kg N kg^{-1} grain for barley, and 0.067 kg N kg^{-1} harvest for cotton.

N surplus, defined as the portion of total N input that is not recovered by crops, was used to represent N that will be eventually lost to the air, soil and water systems. N surplus is widely used to assess the potential agricultural N loss (Sabo et al., 2019; Swaney et al., 2018; Wachendorf et al., 2006; P. Xu et al., 2006). We estimated N surplus (in units of kg N ha^{-1} yr^{-1}) as total N input received by each crop minus the crop-recovered N.

2.3 Spatial patterns of county-level NUE and N surplus

We used metrics that summarize the decadal average crop NUE and N surplus to represent their spatial patterns. The unit of spatial NUE and N surplus is kg N kg^{-1} N and Gg N per county yr^{-1}, respectively. Besides, we also applied Sen's slope to obtain the change magnitude of annual total N input and N surplus of the eight studied crops during two periods, 1970-1989 and 1990-2019, across space (Zhang et al., 2017). Sen's slope estimates monotonic trends and has low sensitivity to outliers (Sen, 1968). The statistical significance of the estimated Sen's slope was evaluated at a 5% level by using a non-parametric Mann-Kendall test (Kendall, 1948).

3 Results

3.1 Crop-recovered N responses to N input of eight major crops

Since we adopted constant crop-specific N recovery coefficients, we used crop-recovered N to represent crop yield when studying the trajectories of NUE and N surplus against crop yields. We calculated the planting area-weighted average crop-recovered N versus N input rates across the U.S. (Figure 1a). The national average crop-recovered N shows a distinct curvilinear relationship with the N input trajectory, indicating a strong N fertilization effect. However, it is noteworthy that the declines in crop-specific recovered N response are hidden by the national average. Specifically, the N inputs of corn and rice fall in the high end of the spectrum, while the rest of the crops cluster at the lower end. The recovered N of most crops, such as barley, corn, cotton, and winter wheat, first increases and then declines or levels off as N input rate increases. Spring wheat and durum wheat are consistent in showing an increasing response, followed by a slowed-down trend of recovered N after reaching the maximum. Rice recovered N near-linearly increases with N input rate. Among the eight crops, sorghum is the only crop showing a "V-shaped" response in recovered N with N input increases.

3.2 Nitrogen use efficiency response to N input of eight crops

The trajectories of crop NUE responses to N input (Figure 1b) are generally contrary to the above-described crop-recovered N trajectories. It is noteworthy that all crops except corn show a declining NUE trend as N input increases. Specifically, barley, cotton, spring wheat, winter wheat, and durum wheat show a similar decreasing trajectory. The decreases in sorghum NUE presents a smaller range, compared with other crops. Rice NUE shows a near-linear decreasing course with N input ranging from 100 to 250 kg N ha^{-1}. Corn NUE is relatively stable along lower N input gradient (e.g., 100 to 150 kg N ha^{-1}). When N input rises to a higher end (e.g., 175 to 225 kg N ha^{-1}), corn NUE begins to slightly increase. The national crop NUE overall displays a slightly increasing trend (e.g., from 0.55 to 0.65 kg N kg^{-1} N) as N input increases, which is dominated by corn NUE.

We also compared the temporal changes in NUE of the eight studied crops over the past 50 years (Figure 2). Specifically, the NUE of barley and winter wheat experienced a large decline from 1970 to the mid-1990s and started increasing afterward. The NUE of spring and durum wheat declined from over 1.0 kg N kg^{-1} N in the 1970s to 0.6 kg N kg^{-1} N in the early 2000s and leveled off thereafter. The NUE of cotton and sorghum was relatively stable (e.g., 0.6 kg N kg^{-1} N) from 1970 to the mid-1990s, followed by an increase thereafter. Corn NUE consistently increased from 0.45 kg N kg^{-1} N in the 1970s to 0.6 kg N kg^{-1} N in the 2010s. Rice had a lower NUE (0.4 kg N kg^{-1} N) than other crop types and showed a marginal variation over the study period. During the first half of the study period (e.g., from 1970 to the mid-1990s), national crop NUE remained stable at ~0.5 kg N kg^{-1} N. It slowly increased to ~0.65 kg N kg^{-1} N by the end of the 2010s, with an average of 0.59 kg N kg^{-1} N over the entire study period (Figure 2).

3.3 Relationship between crop-specific N Surplus, recovered N, and N input

We examined the dynamics of annual N surplus along the gradient of crop-recovered N and N input for the eight crops (Figure 3). Generally, N surplus shows a curvilinear relationship with N input rates for all crops except that corn N surplus displays a "V-shape" when its N input ranges from 170 to 220 kg N ha^{-1} (Figure 3a). Among the eight crops, corn and rice show the highest N surplus as well as the highest N input rates. Along with the N input gradient, the national average N surplus displays a three-stage trajectory: a first increasing stage (e.g., N surplus changes from 25 to 50 kg N ha^{-1}) and then a leveled-off stage (e.g., ~50 kg N ha^{-1}), followed by a further rising stage (50-62 kg N ha^{-1}), the latter of which is likely driven by the trend of corn N surplus due to its large planting acreage.

By contrast, the crop-specific N surplus shows a non-linear relationship with crop-recovered N (Figure 3b). Unlike other crop types, rice demonstrates a consistently increasing N surplus trajectory as recovered N increases, indicating rice N input increase is more rapid than its recovered N increase. A few crops, such as corn and winter wheat, show a shift from a positive to a negative relationship between their recovered N and N surplus, indicating N surplus in these two crops shifts from an input-driven to an output-driven pattern. Durum wheat and spring wheat experience a first-declining-and-then-increasing trend, while barley, cotton, and sorghum show a consistent non-linear declining trend along the recovered N gradient. At the national scale, N surplus first increases from 30 to 55 kg N ha^{-1} as crop-recovered N rises from 40 to 90 kg N ha^{-1}, and it starts decreasing when crop-recovered N is above 90 kg N ha^{-1}. Such results at the national scale suggest that increases in recovered N of output-driven crops likely helped reduce the national N surplus, among which corn was the dominant contributor due to its large planting area.

3.4 Spatiotemporal patterns of NUE and N surplus of eight major crops across the U.S.

3.4.1 Corn

The cross-county corn NUE showed a distinct spatial pattern over decades (Figures 4a-e). During the 1970s and 1980s, corn NUE in most counties was less than 0.4 kg N kg^{-1} N, except for the Great Plains and the Midwest regions (Figures 4a and 4b). Since the 1990s, the NUE in most counties started to increase, including major corn-producing states (e.g. Iowa, Nebraska, Wisconsin) (Figure 4c). During the 2000s, the area with corn NUE > 0.5 kg N kg^{-1} N further extended to other Midwestern states, such as Illinois, Indiana, and Michigan, and the NUE in the southern and eastern U.S. increased from 0.2 to 0.5 kg N kg^{-1} N (Figure 4d). During the 2010s, corn NUE in most counties switched to a higher level (> 0.5 kg N kg^{-1} N), with peaks at 0.9 kg N kg^{-1} N yr^{-1} in some northeastern states like New York (Figure 4e). However, the NUE in North and South Dakota were as high as 0.8 kg N kg^{-1} N in the 1970s and the 1980s, which possibly signals soil mining at low levels of N input, and NUE gradually declined to ~0.5 kg N kg^{-1} N in the most recent decades. In the most recent decades, our study shows many corn-producing counties in the western U.S. “disappeared”. It is likely related to the fact that USDA NASS decided to stop reporting crop yield in these areas if cropland areas declined to a lower-bound threshold. Some marginal corn-producing counties remained at a relatively high NUE level in the Central Valley in California, Washington, and Idaho(Figures 4a-e).

Corn N surplus experienced a consistent spatiotemporal pattern in the U.S. from the 1970s to the 2010s (Figures 4f-j). The hot spots of the largest N surplus (> 5.0 Gg N yr^{-1}) were found in the Midwestern counties, with the peaks in both Illinois and Iowa (Figure 4b). Over the past five decades, high N surplus intruded westward to a larger spatial extent, including the Dakotas, Nebraska, and the southern parts of Minnesota. It showed a stable trend in the remaining regions over the study period, except that increasing corn N surplus was found in the Central Valley in California after the 2000s.

3.4.2 Rice

Compared with corn, rice NUE showed an overall lower value (< 0.5 kg N kg^{-1} N), but a relatively stable trend over decades (Figures S1a-e). The number of rice-growing counties also decreased over time, concentrating on the northern and central counties within the Central Valley and the Lower Mississippi Alluvia Valley (LMAV). In the LMAV region, the NUE in the 1970s was the highest among the five decades, implying the possibility of soil mining when the anthropogenic N input rate was low. In the Central Valley, the lowest NUE occurred in the 1970s and reached a peak in the 1990s, while it gradually decreased during the post-1990 period (Figures S1a-e).

3.4.3 Wheat

Winter wheat NUE overall showed a decreasing trend over the past five decades (Figure S2), reflecting the low levels of N input and possible soil mining at the beginning of the study period. Specifically, several states, including Arkansas, New Mexico, and Texas, were characterized by increasing NUE since the 2000s. The winter wheat NUE in a few states, including North Dakota, Oklahoma, and Missouri, decreased from ~1.0 kg N kg^{-1} N in the 1970s to < 0.5 kg N kg^{-1} N in the 1990s and then increased to ~0.6 kg N kg^{-1} N by the 2010s.

In contrast to winter wheat, the NUE of spring wheat and durum wheat showed a distinct spatial pattern over the five decades (Figures S3 and S4). For spring wheat, NUE reached 1.0 kg N kg^{-1} N in most areas in the 1970s and largely decreased in Minnesota and the western area of North and South Dakota (Figure S3). The remaining states, including Idaho, Montana, Oregon, and Washington, demonstrated a slightly decreasing trend over the study period, although this decreasing trend became smaller or insignificant after the 2000s. The NUE dynamics of durum wheat over the study period was similar to that of spring wheat, decreasing from ~1 kg N kg^{-1} N in the 1970s to 0.4 kg N kg^{-1} N in the 2010s. However, durum NUE kept relatively stable in California and Arizona over decades (Figure S4).

The largest N surplus (above 5.0 Gg N yr^{-1}) of winter wheat was found in the counties in Kansas, Oklahoma, and Northern Texas, and it kept relatively stable in the rest counties (Figures S2f-j). The N surplus of durum wheat and spring wheat also remained stable over decades, with the largest N surplus (6.5 Gg N yr^{-1}) found in the border between Montana and North Dakota (Figures S3 and S4).

3.4.4 Barley, Cotton, and Sorghum

In terms of crop spatial distribution, not surprisingly, the number of counties where barley, cotton, and sorghum were planted was found to decrease over the past five decades (Figures S5-S7). The major producing areas of these three crops were found to respectively shrink into the northwest, the southern coastal regions, and the Great Plains. Barley NUE remained high (~1 kg N kg^{-1} N) over the entire study period (Figure S5). For cotton, the highest increases in NUE were found in the southeastern coasts, and it remained stable (~0.7 kg N kg^{-1} N) in the southwestern coasts of the U.S. (Figure S6). Sorghum NUE was relatively low (~0.4 kg N kg^{-1} N) in most of its planting regions in the 1970s and the 1980s and peaked in the central Great Plains in the 1990s (~0.8 kg N kg^{-1} N) (Figure S7). Since the 2000s and the 2010s, however, sorghum NUE has shown large variations across space.

The N surplus of these three crops was relatively homogeneous across the nation (Figures S5-S7), with less than 2.5 Gg N yr^{-1} N surplus found in most counties. The highest N surplus of them all occurred in the 1970s and the 1980s.

3.5 Crop-specific nitrogen budget across the U.S.

We examined the national-level N budget of the eight crops from 1970 to 2019 (Figure 5). N fertilizer was the dominant N input for the eight crops and thus the dominant driver of N surplus dynamics. Among them, corn showed a consistently increasing trend in total N input amount and crop recovered N from 1970 to 2019 (Figure 5b). Corn N surplus first increased from 2.5 Tg N yr^{-1} in 1970 to 3.8 Tg N yr^{-1} in the late 1980s and then slightly decreased till the 2000s, and it overall experienced large variations in the 2010s and eventually decreased to a level observed in the 1970s (Figure 5b). The total N input and crop recovered N of barley and sorghum showed a similar decreasing trend from the early 1970s to the end of the 2010s (Figures 5a and 5g). Barley

N surplus was around zero in the 1970s and sharply increased to ~0.2 Tg N yr^{-1} in the 1980s; it decreased to less than 0.1 Tg N yr^{-1} in the 2000s and to around zero in the 2010s (Figure 5a). The maximum N surplus of barley reached up to 62% of its total N input in 1985. Sorghum N surplus slowly decreased from 0.3 Tg N yr^{-1} in the 1970s to close to zero in the 2010s, accounting for 34% to 50% of its total N input over the study period (Figure 5h). Mainly driven by the increasing N fertilizer input, the N surplus of cotton, rice, durum wheat, spring wheat, and winter wheat all slowly increased from the 1970s to the 2000s and then gradually decreased till the end of the 2010s (Figures 5c,d,e,f,h). Specifically, cotton N surplus ranged from 0.1 to 0.4 Tg N yr^{-1}, accounting for 17% to 48% of the total N input (Figure 5c). Rice N surplus accounted for 53% of its total N input and remained relatively stable over time, with an average of 0.1Tg N yr^{-1} (Figure 5e). The maximum N surplus of durum wheat and spring wheat accounted for 60% (0.05 Tg N yr^{-1}) and 52% (0.26 Tg N yr^{-1}) of their total N inputs, respectively, while their negative N surplus in the 1970s was likely due to the small N fertilizer input and soil mining (Figures 5d and 5f). Winter wheat N surplus ranged from 8% to 26% of total N input and reached a maximum value of 0.58 Tg N yr^{-1} in 1989 (Figure 5h).

We also aggregated the N budget of the eight studied crops to represent the national crop N budget (Figure 6). Overall, the national total N input slowly increased from 3.3 Tg N yr^{-1} in 1970 to 8.2 Tg N yr^{-1} in 2019. Over 90% of the national total N input was from fertilizer use, followed by the residual N of soybean rotation and atmospheric N deposition. Crop-harvested N increased by 120%, with a minimum value of 2.6 Tg N yr^{-1} found in 1970 and a maximum value of 5.8 Tg N yr^{-1} in 2016. Crop N surplus on average accounted for 41% (27% to 55%) of the total N input during the study period. A minimum crop N surplus of 1.2 Tg N yr^{-1} was found in 1971, and a maximum of 5.0 Tg N yr^{-1} in 2012 (Figure 6). The 2012 peak of crop N surplus might be related to the widespread drought in the U.S. this year. Additionally, the national N surplus showed a near-parabolic trend, with an annual increasing rate of 0.09 Tg N yr^{-1} (p<0.05) over the period from 1970 to 1989 and an annual decreasing rate of 0.02 Tg N yr^{-1} (p=0.1) from 1990 onward. The temporal dynamics of the national total N surplus were dominated by corn due to its large planting areas and high N input rates per unit area.

3.6 Spatiotemporal trends of total N input and N surplus of the eight studied crops

3.6.1 Corn

During the period from 1970 to 1989, corn total N input in the Midwest U.S. showed a significantly increasing trend, with a maximum annual increasing rate > 0.2 Gg N yr^{-2}, while it experienced a decreasing trend in the states in the eastern coast and the central Great Plains (Figure 7a). Since 1990, the significantly increasing trend in corn total N input has extended to a much larger area covering the entire Midwest U.S. and the northern Great Plains, with a peak increasing rate greater than 0.25 Gg N yr^{-2} found in Dakotas (Figure 7b).

The trend of corn N surplus from 1970 to 1989 demonstrated a similar spatial pattern but a smaller extent (p<0.05), compared with its total N input (Figure 7c). The maximum increasing rate (~0.2 Gg N yr^{-1}) was found in the Corn-Belt region, including Illinois, Iowa, Indiana, Ohio, Michigan, and the eastern parts of Dakotas, while the N surplus in the remaining regions showed either a significantly decreasing or a flat trend during this period (Figure 7c). In the post-1990 period, corn N surplus in most of the Corn-Belt region switched to a decreasing or flat trend; however, it significantly increased in the northern Great Plains (e.g., Dakotas and Kansas) due to the significant increases in its total N input (Figure 7d).

3.6.2 Wheat

The majority of the winter wheat growing areas experienced significant increases in total N input during the pre-1990 period (Figure S8a). From 1990 to 2019, however, it reversed to a significantly decreasing trend in most areas, except in Dakotas and Wyoming (Figure S8b). Winter wheat N surplus during these two periods followed a similar spatial pattern as its total N input (Figures S8c and S8d). Similar to winter wheat, spring wheat also experienced a positive increase in total N input over the past five decades, with a hotspot found in the northern Great Plains (Figure S9a). Spring wheat total N input in the eastern part of Dakotas had a significant decrease during the post-1990 period (Figure S9b). The spatial patterns of spring wheat N surplus trend during these two studied periods were largely driven by its total N input, with a peak (> 0.25 Gg N yr^{-2}) found in the northern Great Plains (Figures S9c and S9d). Durum wheat showed insignificant trends in total N input and N surplus during the two studied periods, and only some marginal counties in the northern states and Arizona demonstrated a significant trend over the past five decades (Figure S10).

3.6.2 Barley, Cotton, Rice, and Sorghum

The total N input of barley had an increasing trend in most counties from 1970 to 1989 (Figure S11). The largest total N input increase rate (e.g., > 0.2 Gg N yr^{-2}) was found in North Dakota. However, most of the counties have been characterized by decreasing total N input since 1990 (Figures 11a and 11b). The trends of barley N surplus mostly followed the spatial patterns of its total N input in the pre- and post-1990 periods (Figures S11c and S11d).

Cotton total N input showed a significantly decreasing trend in most counties (Figures S12a and S12b). Over the entire study period, cotton N surplus showed a decreasing trend, with the peak annual decreasing rate (e.g., -0.2 Gg N yr^{-2}) detected in central Texas since 1990 (Figures S12c and S12d).

Rice total N input in the Rice-Belt demonstrated an overall increasing trend during the entire study period (Figures S13a and S13b). In other main rice-producing regions, such as California, the total N input experienced an insignificant decreasing trend from 1970 to 1989, and this decreasing trend became smaller or turned into an increasing trend after 1990. Not surprisingly, rice N surplus followed the spatial patterns of its total N input (Figures S13c and S13d), indicating an "input-driven" trajectory of N surplus.

Sorghum total N input displayed decreasing trends in most counties over the entire study period (Figures S14a and S14b). Counties in South Dakota and southeastern Missouri were identified as hotspots of increasing total N input from 1970 to 1989, whereas the centers of increasing N input moved to counties in eastern Kansas from 1990 to 2019. Similar to other above-mentioned crops, sorghum N surplus trend was also largely driven by its total N input over the entire study period (Figures S14c and S14d), demonstrating a smaller area with a significant changing trend after 1990 than N input ($p<0.05$).

4 Discussion

Our analysis quantified yield responses, represented by crop-recovered N, to the N input of eight major crops in the U.S. using the long-term data. The largest yield response to N addition was found in corn nationally, while yield responses in the remaining seven crop types shifted to a lower rate as N input increases (Figure 1a). It likely indicates N saturation or limiting factor shifted from N to other resources (such as water, phosphorous, etc.) for these seven crops. We also examined the county-level crop-specific patterns of NUE and N surplus. A consistent pattern that emerged for six out of the eight crops showed a decreasing NUE along the N input gradient, whereas the NUE of corn and rice barely changed (Figure 1b). Our analyses illustrated that the N surplus of corn and winter wheat was likely reduced by their yield increases, whereas the N surplus of the other studied crops was likely to be mitigated by a lower N input (Figure 3).

4.1 NUE responses to N inputs

National crop NUE demonstrates a non-linear relationship with N input increases (Figure 1b). The non-linear NUE has been previously reported across the entire U.S., with a rapid yield response to medium-level N fertilizer use rates (125-175 kg N ha^{-1} yr^{-1}) (Lassaletta et al., 2014; Zhang et al., 2015). However, these previous studies did not provide information on crop-specific NUE patterns and yield responses to N input increase. Using state-level survey data, Lu et al., (2019) demonstrated that crop yield of corn and winter wheat in the U.S. plateaued with N fertilizer increases, and NUE declined when N input exceeded the threshold for these two crops. This finding generally agrees with our county-level analyses, suggesting that crop NUE declined along with N input increases (Figure 1b).

Within the U.S., increasing crop yields, resulting from advanced crop varieties, improved management practices and other technological improvements, largely increased crop NUE since the 1990s (van Grinsven et al., 2015; Xu et al., 2013). Additionally, N fertilizer input has remained steady in the U.S. since the 1990s (Cao et al., 2018). Among the eight studied crops, our analysis identified two crop groups showing different NUE dynamics along the N input gradients. The NUE of six crop types (e.g., barley, cotton, durum wheat, sorghum, spring wheat, and winter wheat) experienced an overall decreasing trajectory with a lower range of N input rates (e.g., 10-100 kg N ha^{-1}, Figure 1b), while corn and rice NUE barely varied with N input rates that distributed in a higher range (e.g., 100-220 kg N ha^{-1}). Hence, the flat national relationship between crop NUE and N inputs should be used with caution for the nutrient management of specific crops at local scales because spatiotemporal variations in crop yield and NUE are likely to be smoothed when aggregated to the national scale. Our results also reflect that it is critical to developing sub-national analyses for understanding the impacts of N addition on yield gain of specific crops, especially in counties with a large amount of N input and large cropland areas like the U.S. Midwest.

4.2 Dynamics of U.S. crop NUE over space and time

Our estimates showed that the national crop NUE ranged from 45% to 73% with an average of 59% in the U.S over the past 50 years (Figure 2). A previous study reported the national crop NUE of 2007 was close to 55% in the U.S. (Houlton et al., 2013), which is slightly lower than our estimate. Using national-level data, Lassaletta et al., (2014) reported that U.S. crop NUE was in between 47% and 73% from 1961 to 2010, with a mean value of ~60% over those 50 years, agreeing well with our estimates. Nevertheless, we revealed more variations over space, time, and crops.

Among these crops, the spatiotemporal patterns of barley and three wheat types largely differ from other crops (Figures 2 and S2-S5). The initial higher NUE values of barley and three wheat types were likely due to the low levels of initial N input, and wheat might access extra N through soil mining at this stage, reducing soil fertility (De Klein et al., 2017). The NUE decreases in these crops were possibly caused by increases in N input that was used to alleviate soil degradation (De Klein et al., 2017). According to a recent study, N fertilizer input into winter wheat sharply increased from the 1970s to the 2000s (Cao et al., 2018). However, winter wheat yield increases were slower compared with its N input rates (Lu et al., 2019), leading to the decreasing NUE trend. The latter increasing trend of its NUE since the 2000s (Figure 2) was possibly induced by the recent increases in yield and stagnated N fertilizer inputs. For example, winter wheat yields were reported to approach their maximal potential levels in the U.S. Great Plains, with less genetic advancement compared with corn (Ray et al., 2012). Cao et al. (2018) demonstrated that the N fertilizer input rates for winter wheat have leveled off since the 2000s. Another study shows that, for winter wheat from 1960 to 2008, a change point of its NUE (transitioning from an increase to a level-off) was observed in the western U.S. in 1993 and the central U.S. in 2003 (Lin & Huybers, 2012). Besides, the winter wheat-growing areas presented a broad range of agroclimatic zones, which might drive the spatial variability of its NUE.

Despite the increases in N fertilizer input rates for durum wheat and spring wheat in the U.S. from the 1960s to the most recent decade (Cao et al., 2018), the NUE of these two wheat types largely decreased from the 1970s to the 2000s and remained leveled-off since then. It is likely caused by the higher N input than the N demand for these two types of wheat. Besides, durum wheat and spring wheat were mainly distributed in the northwestern states, in which arid and semi-arid conditions might be one of the limiting factors for crop yield and NUE.

For corn, cotton, and sorghum, although NUE declines occurred in some counties (e.g. corn NUE declines in Dakotas and sorghum NUE declines in northern Texas), it overall increased over the past five decades (Figure 4, S1, S6, and S7). Even though an increasing trend was widely observed, corn in southern Great Plains and the eastern U.S. still showed a consistently low NUE (e.g., less than 0.5 kg N kg^{-1} N) over the past five decades (Figure 4). A previous study reported that the lower corn yield in Texas might be due to the common fall N application practice that likely increased soil N loss during the winter fallow period (Torbert et al., 2001). Here we used corn as an example to compare its annual N surplus against fall and spring N fertilizer application rates since these two application timings account for the majority of annual total N input. For example, fall N fertilizer application accounted for 20% of annual total N fertilizer use in Texas and 31% in Iowa and Minnesota, and spring N application accounted for 34% in Texas, 43% in Iowa, and 59% in Minnesota in 2015. Across the U.S., corn fall N application from 2010 to 2019, ranging from 0-60 kg N ha^{-1}, positively correlated with its N surplus ($R=0.57$, $p<0.01$, Figure 8a). On the other hand, a positive correlation was found between the spring N fertilizer application and N surplus with a higher statistical significance (Figure 8b). These findings suggested that fall N fertilizer application to corn could significantly contribute to its annual total N surplus, despite its lower input rates.

The increases in cotton NUE in the southeastern U.S. over the past five decades were more likely due to its yield increases because its total N input remained relatively stable (Figure 4c). However, cotton NUE in the southern Great Plains (e.g., Texas) did not show significant changes although it remained relatively high (Figures S6 and S12). A recent study reported there was a difference in the adoption of conservation tillage between the southern Great Plains cotton systems (30%) and the southeastern U.S. (70%) (DeLaune et al., 2020). The residue retaining benefits of reduced till during the cotton rotation phase could significantly increase cotton lint yields in the southern Great Plains (Baumhardt et al., 2012), which might contribute to the different cotton yield responses to N input rates (Boquet et al., 2004). The 1990s-onward high sorghum NUE (> 0.6 kg N kg^{-1} N) in the Great Plains was likely due to the reducing total N input (Figures 5g, S14a, and S14b). Rice NUE displayed a consistent spatiotemporal pattern over the past five decades (Figures 2 and S1), reflecting the linear relationship we found between its yield and N input over the entire study period (Figure 1a). More specific studies are needed to investigate whether other agricultural management practices can help reduce the N surplus of these three crops across regions.

4.3 Regional hotspots of high NUE: a future increase in crop NUE?

Regional variations in patterns of major crop NUE versus total N inputs from 1987 to 2012 were reported by a previous study that investigated ~17 crop species in the U.S. (Swaney et al., 2018). This study showed that crop NUE likely decreased with growing N fertilizer input in the majority of regions in the U.S. but did not report crop-specific crop NUE. Our results identified the critical areas with high crop NUE, well echoing the findings of a recent study (Sabo et al., 2021). High NUE in corn was found on the most fertile Midwest soils (e.g., Iowa, Illinois, Minnesota, and Nebraska), which received plentiful rainfall, and advanced management practices (e.g., rotation, tile drainage, and tillage). Furthermore, previous studies showed that earlier corn planting in some of these regions (e.g., Minnesota, Nebraska, and Iowa) contributed significantly to yield increases (Kucharik, 2006, 2008), which likely boosted the NUE increases. High crop NUE was also found in the Great Plains, where irrigation practice is widely applied to winter wheat and sorghum, as well as the south coasts, where advanced management practices (e.g. pest and disease control) are utilized for cotton production.

However, these high-NUE counties and regions were found to be the hotspots of N surplus for each crop type owing to the large planting area and high N input rates. For instance, the Midwest regions showed the largest corn N surplus (on average 5-8 Gg N yr^{-1}). Nevertheless, we still found that the national total N surplus approximately declined at an annual rate of 0.8 Tg yr^{-1} during the 2010s (Figure 6), which was significantly driven by corn N surplus decline (Figure 5b and S15b). Particularly during 2015-2019, we found that corn N surplus in the U.S. Midwest decreased to a level close to the 1970s (Figure 3f and Figure S15b). The national total N input for corn during 2015-2019 was averaged ~2 Tg N yr^{-1} higher than that in the 1970s, while N surplus levels were close between these two periods (Figure 6). This indicates that crop harvested N was much higher during the most recent years, likely driven by crop technology improvement and enhanced agricultural management practices. Corn yields were found to increase over the past decades (Kucharik et al., 2020), and better N management practices (e.g., using cover crops and bioreactors) adopted by farmers might also help enhance crop yield and retain soil N (Christianson et al., 2018; Stuart et al., 2018). The most notable effort is the 4Rs of Nutrient Stewardship strategy (i.e., right N fertilizer application place, right application time, right N fertilizer use amount, and right N fertilizer use rate), which promotes proper nutrient management strategies to improve crop N use efficiency (IFA, 2009). In addition, policy-based efforts, such as the "avoid, control, and trap" strategy proposed by the USDA (Osmond et al., 2015), and state-based efforts, such as Iowa's Nutrient Reduction Strategy, have set nutrient reduction goals and provided voluntary guidance to farmers to meet those goals (Lawrence, 2013). On the other hand, we also realized that the 2012 drought in the U.S. was likely the main reason for the high average N surplus in the 2010s (Figure 5 and 6), causing a major increase in N surplus in Dakotas, Missouri, and Kansas (Figure 7). This indicates that extreme climatic events like drought could largely undermine the higher-input-efficiency strategy's effectiveness in improving crop yield, which was proven in our follow-up analysis by using this database (Zhang, et al., 2021).

Our work also showed that among the eight crops, the counties with the lowest NUE generally corresponded to a much smaller total N surplus because the total N input amount in these counties was not as high as that in the hotspot regions. This finding indicates that future increases in N input in the low-NUE counties should be cautious because it requires much larger crop yield increases to take up excessive N in these areas, despite the low N surplus revealed. Boosting crop yield was shown to be difficult in these regions due to the low soil fertility or climatic stresses (Hou et al., 2019). As climatic changes will continue, the low crop NUE regions may be at a higher risk than other regions. Compared with other key planting areas, the climate conditions in the U.S. Great Plains, southwestern U.S., and the western U.S. have a higher likelihood of extreme heat (i.e., days above 30 ºC or extreme degree days) and drought combined, both of which have been documented as causes of crop yield loss (Basso & Ritchie, 2014; Lobell et al., 2013, 2014). The historical trends in climate and current low-NUE may be a precursor of continued N surplus increases in the next few decades in these counties.

Even in the counties with the highest crop NUE (e.g. the Midwest), extreme rainfall and flooding events likely drove a larger magnitude of N loss via leaching to surface water and groundwater bodies (Lu et al., 2020). Climatic warming and intensive land surface management (e.g. tillage) could largely increase N_2O emission given the large input of N fertilizer (Gregorich et al., 2008). Additionally, N fertilizer shifting to the Urea-base fertilizer could enhance NH_3 emission, especially in areas with alkaline soils like in the northern Great Plains (Cao et al., 2020). Using county-level crop yield data and climate datasets, a recent study demonstrated that the magnitude of corn yield loss driven by excessive rainfall was comparable to that driven by extreme drought in the U.S. (Li et al., 2019). Therefore, farmers in these counties should be cautious when increasing N fertilizer use because the yield gap in these regions is likely the smallest nationwide. It is even more difficult to avoid potential N loss in these areas given the smaller yield gap and the higher likelihood of frequent extreme rainfall events and longer drought.

4.4 N loss risk and recommendations for future N management

Achieving food security, sustainability, and ecosystem services at the regional and global scales is the ultimate goal of modern agriculture (Foley et al., 2011). Previous studies estimated that global cereal grain demand will not be met without at least a 60% increase in global N use if crop NUE remains the same as the current level (Dobermann & Cassman, 2005; Ladha et al., 2016). A recent study developed spatially explicit typologies for components of crop N mass balance and indicated a large range of crop N trajectories across the U.S. (Byrnes et al., 2020), however, crop-specific N trajectories were not reported in this study. Using crop-specific N budget data, our analyses suggested that the N surplus of corn and winter wheat was likely output-driven and could be potentially reduced by enhancing the crop yield (Figure 3b). Besides, the national crop N surplus was largely driven by corn N surplus (Figures 5b and 6). Therefore, it will be more challenging to decrease the U.S. crop N surplus without further increasing corn yield that was detected linearly increasing over time (Kucharik et al., 2020). A recent study demonstrated that corn yield response likely leveled off when N fertilizer exceeded its threshold level at approximately 150 kg N ha^{-1} (Lu et al., 2019). Furthermore, past trends may not necessarily indicate a continued future increasing trend of crop yield. Based on the above-mentioned evidence, it is still unclear whether further increases in N inputs can keep enhancing yields, and the yield response can be much more complex when the spatial heterogeneity of other environmental facts is considered. Hence, without a comprehensive understanding of crop yield performance under different management practices given various physical conditions, it is challenging to predict the future risk of N loss from cornfields in the U.S.

Among the remaining seven crop types, a few of them, such as barley and winter wheat, demonstrated a shift in N surplus from input-driven to output-driven as yield increases (Figure 3). Interestingly, in the major winter wheat-producing states (e.g. Oklahoma and Kansas) and barley-producing states (e.g. North Dakota and California), we found that the N surplus showed an overall decreasing trend over decades, which might be due to the consistently increased NUE observed in these states. Therefore, for the remaining counties where wheat and barley are widely planted, increasing crop yield could help to reduce N loss. Additionally, efficient N management of these specific crops is of particular importance for policy- and decision-making that aims at reducing N-related environmental pollution (Mitsch et al., 2001; Rabalais et al., 2002).

Compared with the crops discussed above, sorghum and durum wheat showed distinct relationships among N surplus, N input, and crop yield. Recent studies pointed out that the optimum N application rates might be much less if sorghum was grown under limited irrigation and dryland areas in the Great Plains (Hao et al., 2014; Tamang et al., 2011). Our analyses showed that N surplus decreased along with the sorghum yield gradient (Figure 3). Therefore, to maintain a lower N surplus, N input to sorghum should be cautiously estimated with the considerations of local environmental conditions (e.g., climate and soil wetness) and management practices (e.g., irrigation availability) that jointly affect its yield. For durum wheat, our analyses presented an overall positive correlation between its N surplus and N input rate as well as its yield. This indicates that N input stimulated durum wheat yield, but its yield increase was slower than the N input growth. An experimental study showed that, as the N fertilizer rate increased from 73 to 403 kg N ha^{-1}, although grain yield and total N uptake of durum wheat increased, its NUE linearly decreased (Liang et al., 2014). Besides, N fertilizer effects were much more significant than cultivar effects on its NUE, and so growers' decisions on N input rate would be more efficient in enhancing the NUE than their cultivar selection (Liang et al., 2014). Liang et al., (2014) also pointed out that grain yield and total N uptake would plateau when its N fertilizer input reached a threshold. Therefore, the loose coupling between elevated N fertilizer input and yield gain implies that durum wheat N surplus is more regulated by its N fertilizer input.

4.5 Uncertainty

The data and methods adopted in our study have several limitations. First, we used the NuGIS N fertilizer consumption ratio of the county to state in 1987 and 2014 to gap-fill the county total N consumption for the periods of 1970-1986 and 2015-2019, respectively. The gap-filled county-specific N fertilizer consumption had little impact on the state- and national-level analyses. However, it might lead to over- or under-estimation of N fertilizer use rates and N surplus amount in some counties for these periods, if significant changes occurred in county-level crop acreage or crop-specific fertilizer use rate. This uncertainty can be reduced if the county-level total N fertilizer consumption data become available in the entire study period. Second, due to the lack of time-series data of county-level crop-specific manure application rate, we assumed that manure N applied per acre of each crop type was consistent with the multi-year crop-specific average values reported in each state. Such an assumption likely led to a less variable manure application rate for a given crop over time and space. Third, we used harmonized cropland distribution database to extract the N deposition inputs for each crop each year. However, before 2008 (the earliest year when CDL-derived crop type maps are available for the entire nation), the crop-specific N deposition rates we extracted in this study were likely biased by limited knowledge of crop type and distribution patterns within a county. Finally, the crop N recovery coefficient and the coefficient determining the soil residual N from soybean N fixation were assumed to not vary with time and space because there are no systematic methods that account for these variations. Future research may provide more insights and efforts in gap-filling, reducing uncertainties in data and methods, and assess the roles of these limitations in characterizing spatiotemporal patterns of crop N budget in the U.S.

5 Conclusion

This study investigated the N budget of eight major crops in the U.S., including barley, corn, cotton, durum wheat, spring wheat, winter wheat, sorghum, and rice. This study differed from previous analyses by using county-scale long-term data from 1970 to 2019 for the eight major crop types and comparing the responses of crop recovered N, NUE, and N surplus to N input over space and time. Our results showed that national crop NUE estimated from the eight major crops slowly increased from 0.5 kg N kg^{-1} N in the 1970s to 0.65 kg N kg^{-1} N in the 2010s, with an average of 0.59 kg N kg^{-1} N. The N surplus for these eight major crops on average remained at around 41% (27% to 55%) of the total N input during the study period, which is consistent with other estimates (Houlton et al., 2013; Lassaletta et al., 2014). The corn N surplus in the 2010s decreased to a level close to the 1970s (Figure 5b and S15b), which drove the large decline of the national total N surplus (Figure 6). This was likely caused by policy-driven and state-oriented efforts on reducing agricultural N loss. Spatial patterns in NUE and N surplus over the past half-century suggest some regions had historically improved crop N utilization but were still facing a high risk of losing N when N input increases. Increases in N surplus in some regions, including portions of the Great Plains (e.g., corn N surplus) and Rice-Belt (e.g., rice N surplus) over the most recent decades, may be a cause of concern. Furthermore, N input has been a major factor in driving yield increase. Yet, for some crops such as wheat and barley, the crop yield has likely plateaued despite increasing N inputs. With more extreme weather events and changing climate, future crop NUE is less certain, and some regions like the Midwest and the Great Plains appear to be at higher risk than other regions for future N-related pollution if crop yield does not increase or extra N is applied. Extensive farm-level data collections across diverse conditions of climate, soil, and management regimes are expected to reveal the dominant factors controlling NUE variations in different crop types across the nation.

Data availability

The annual crop-specific NUE and N surplus data can be found via https://doi.org/10.6084/m9.figshare.13030436 (Zhang et al., 2021).

Competing interests

The authors declare no competing interest.

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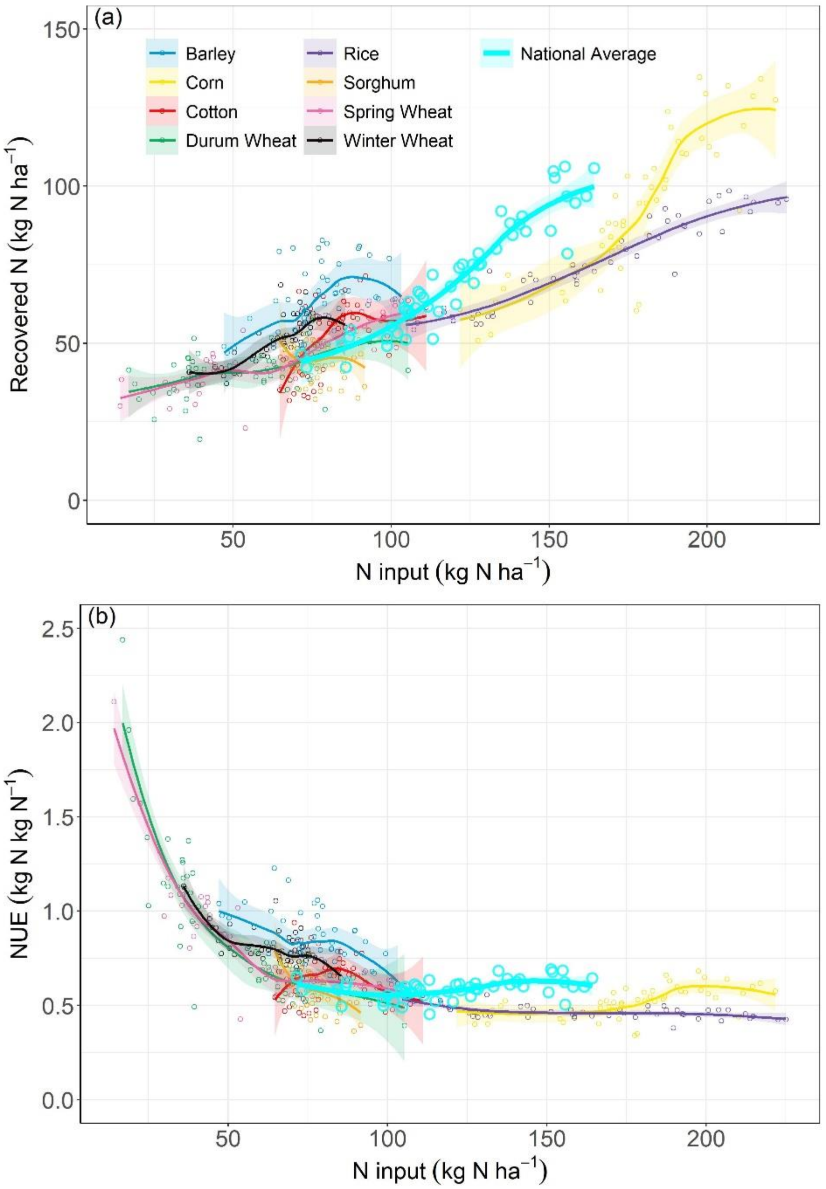

Figure 1. The relationship between crop yield and total nitrogen (N) input rate (a) and the relationship between crop nitrogen use efficiency (NUE) and total N input rate (b) of eight major crops from 1970 to 2019 in the U.S. The solid curves are the “loess” fitted correlations for each crop type over the analyzed years (1970-2019). The shaded areas around the fitted curves indicate the 95% confidence interval. The national average yield and N input data from 1970 to 2019 are aggregated from the eight crops based on the planting area. The national crop NUE

from 1970 to 2019 is calculated as the ratio of total crop harvested N to the total N input of the eight crops. The national-level data are shown by the cyan dots and cyan solid line for each pair of the variables at a national level.

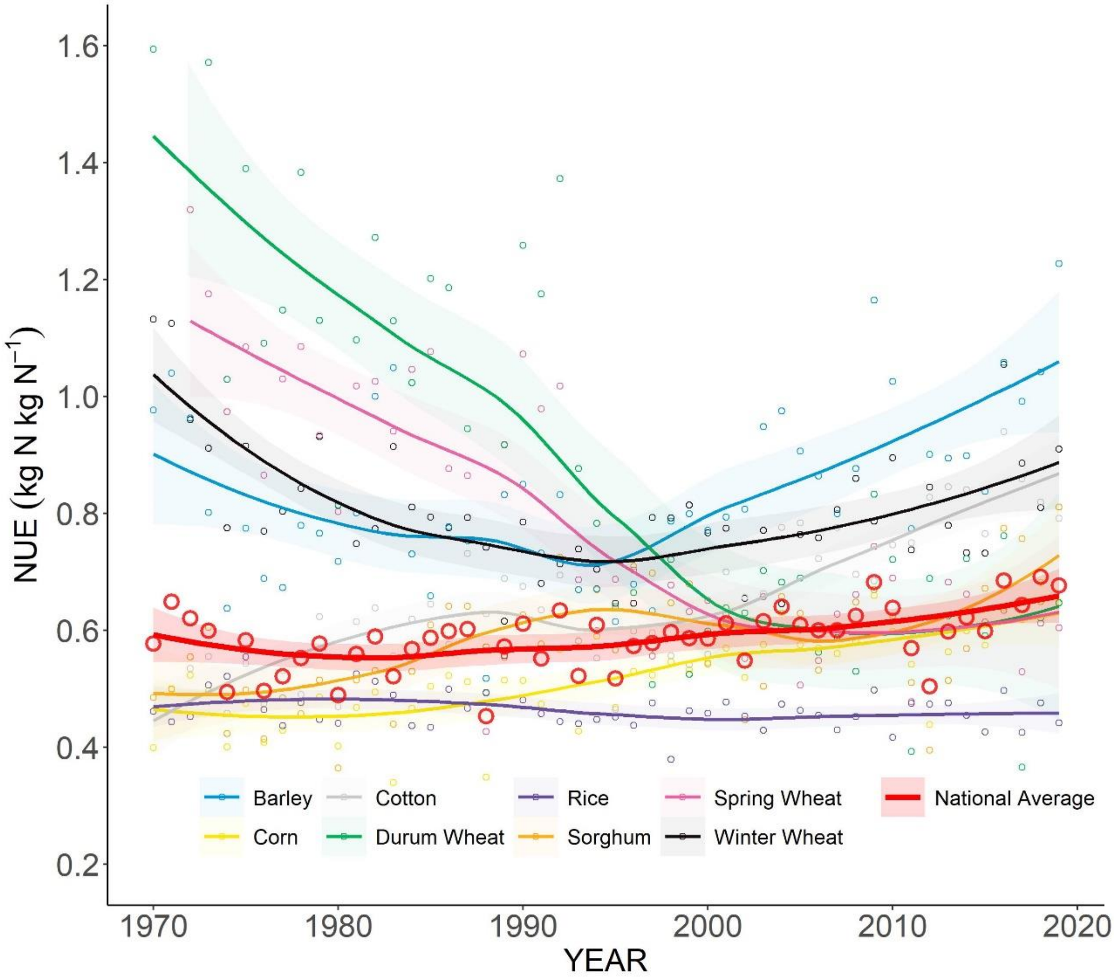

Figure 2. National-scale nitrogen use efficiency (NUE) time series of eight studied crops and the national average crop NUE from 1970 to 2019 in the United States. The solid curves are the “loess” fitted curves for each crop over the analyzed years (1970-2019). The shaded areas around the fitted curves indicate the 95% confidence interval. The red solid curve indicates the non-linear NUE trend at a national level. The crop NUE, including the crop-specific and national

average, is calculated as the ratio of total crop harvested N to the total N input for each crop type or the eight crops counted together.

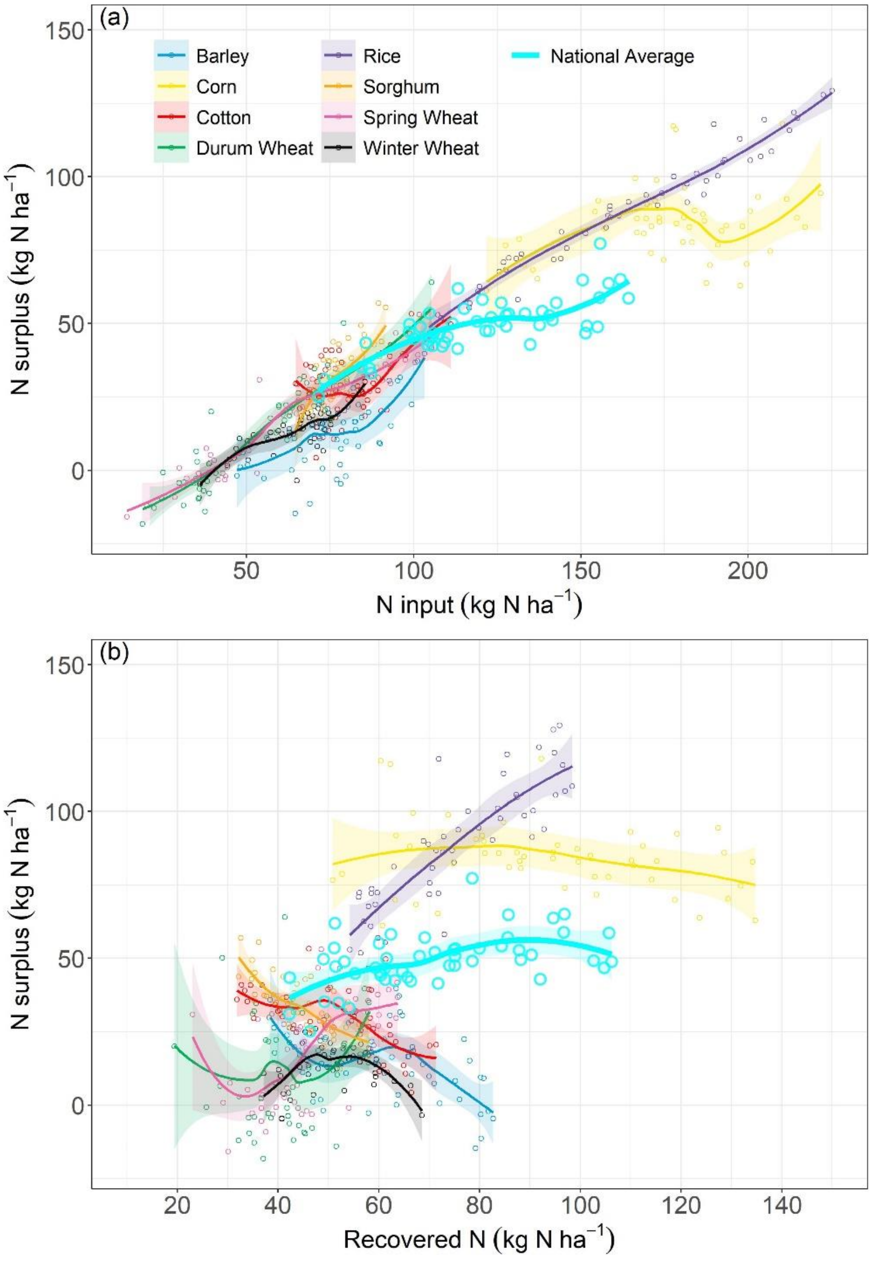

Figure 3. Dynamics of N surplus over the gradient of total N input rate (a), and crop yield (b) for eight major crops in the U.S. from 1970 to 2019. The solid curves are the “loess” fitted curves for each crop type over the analyzed years (1970-2019). The shaded areas around the fitted

curves indicate the 95% confidence interval. The cyan solid line indicates the non-linear trend for each pair of variables at the national level. The national average N surplus, N input, and crop yield are aggregated from the eight crops based on the planting area, shown by the cyan dot.

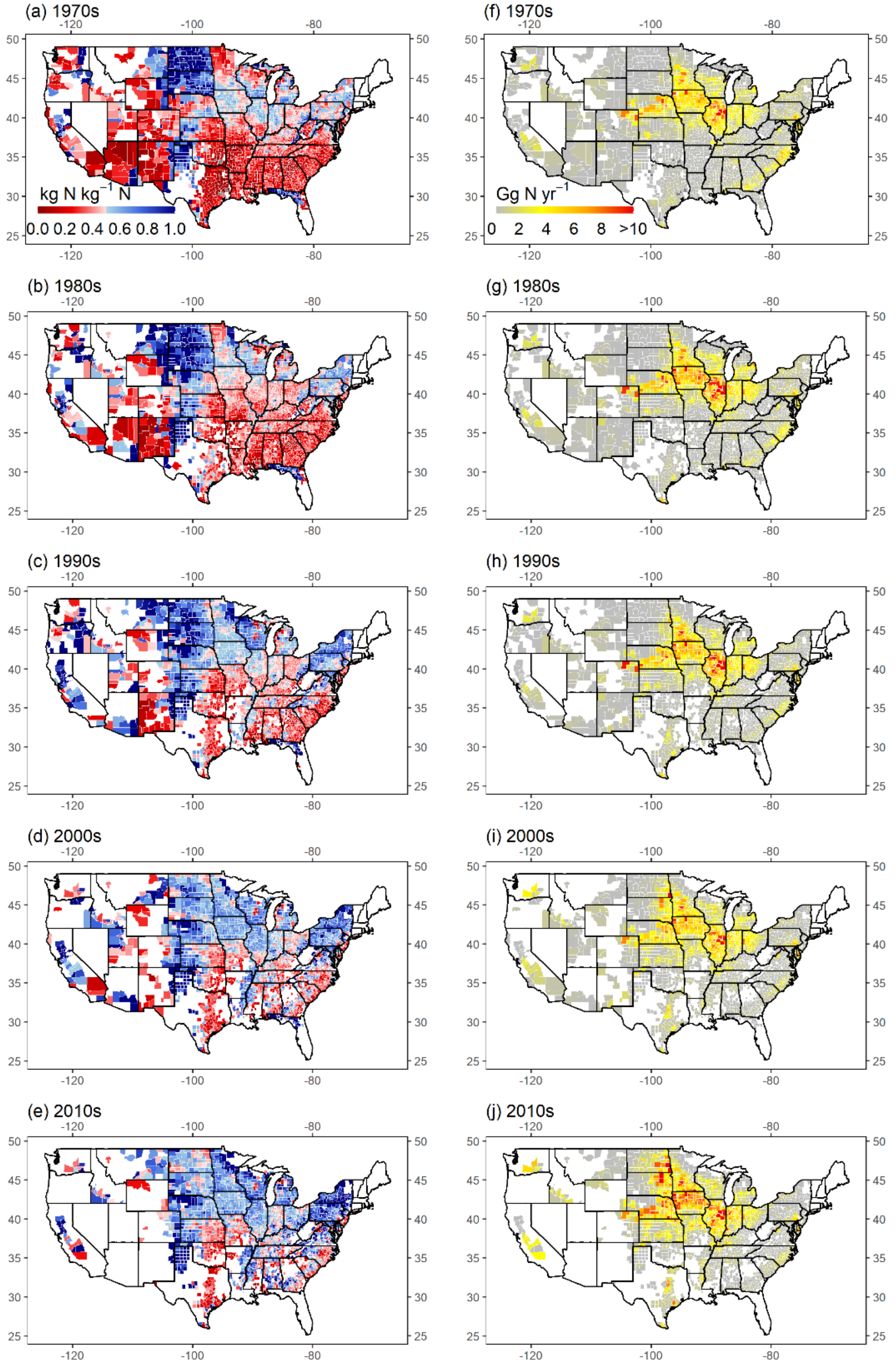

Figure 4. Decadal average NUE (a-e, unit in kg N kg^{-1} N yr^{-1}) and N surplus (f-j, unit in Gg N yr^{-1}) of corn in each county of the contiguous U.S. Annual planting area in each county has been used to calculate the county-level N surplus.

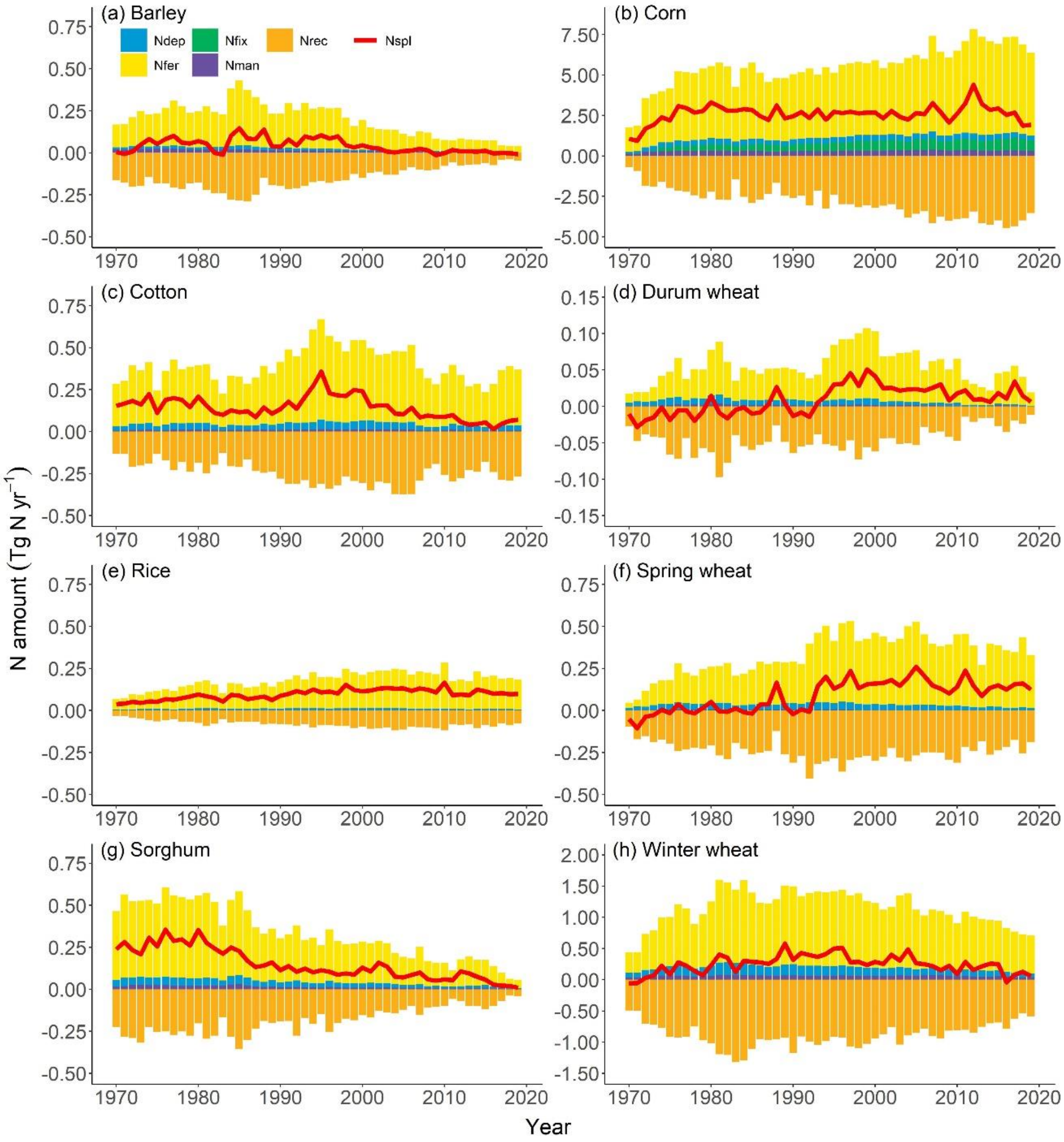

Figure 5. Crop-specific nitrogen budget in the U.S. from 1970 to 2019. N inputs include manure (Nman), atmospheric deposition (Ndep), fertilizer (Nfer), and biological N fixation (Nfix), and N output includes crop recovered N (Nrec). The time series of nitrogen surplus (Nspl) is shown by

the red curve. The data are aggregated from county-level crop-specific N budget. The y axis scales in corn, durum, and winter wheat differ from other crops.

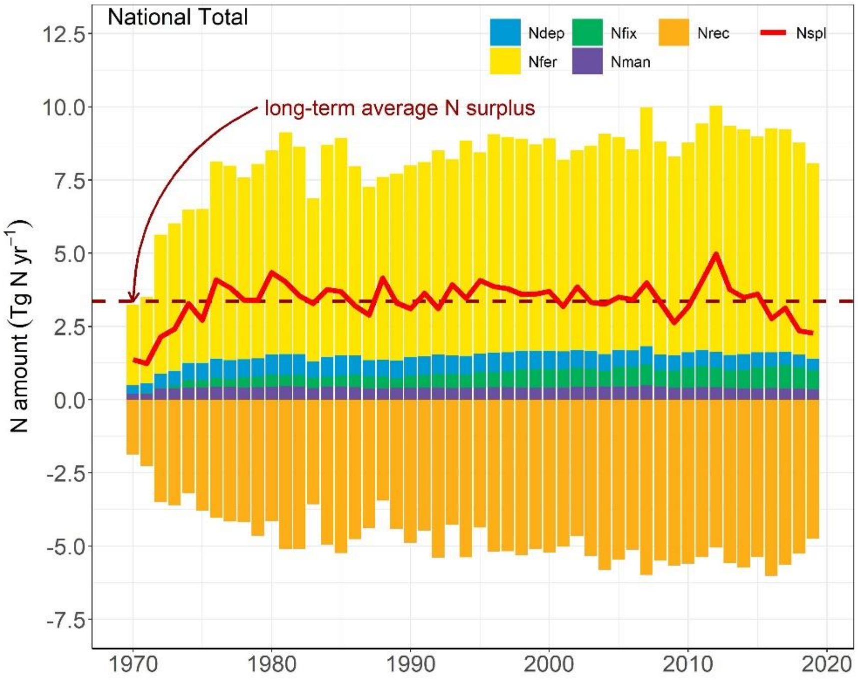

Figure 6. Nitrogen budget aggregated from the eight crops in the U.S. from 1970 to 2019. N inputs include manure (Nman), atmospheric deposition (Ndep), fertilizer (Nfer), and biological N fixation (Nfix), and N output includes crop recovered N (Nrec). The time series of nitrogen surplus (Nspl) is shown by the red curve. The dashed line is the long-term average (1970-2019) N surplus.

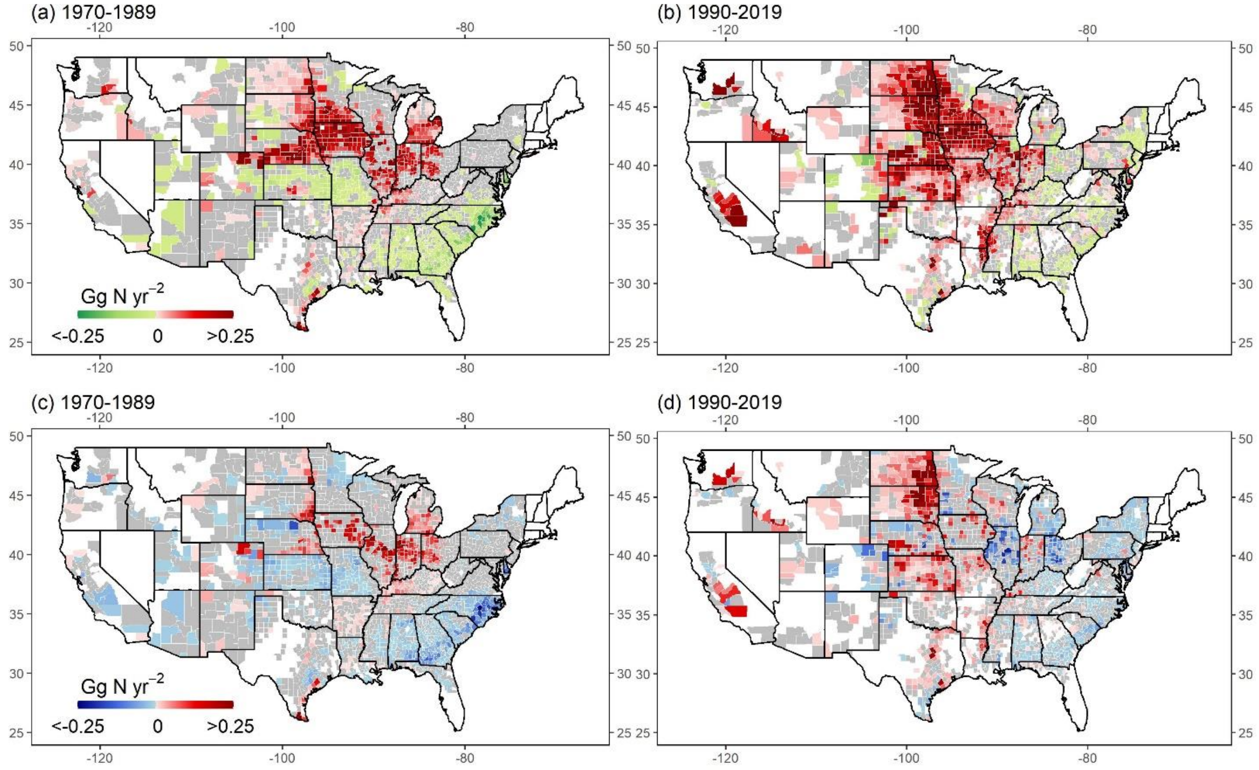

Figure 7. The total nitrogen (N) input trend (a and b, unit in Gg N yr^{-2}) and N surplus trend (c and d, unit in Gg N yr^{-2}) of corn in each county of the contiguous U.S. during the periods 1970-1989 and 1990-2019. The green colors and the red colors in (a) and (b) represent the significantly decreasing and increasing trends ($p<0.05$) in total N input, respectively. The blue colors and the red colors in (c) and (d) represent the significantly decreasing and increasing trends ($p<0.05$) in N surplus, respectively. Grey area indicates no significant trend at 0.05 level according to the Mann-Kendall test.

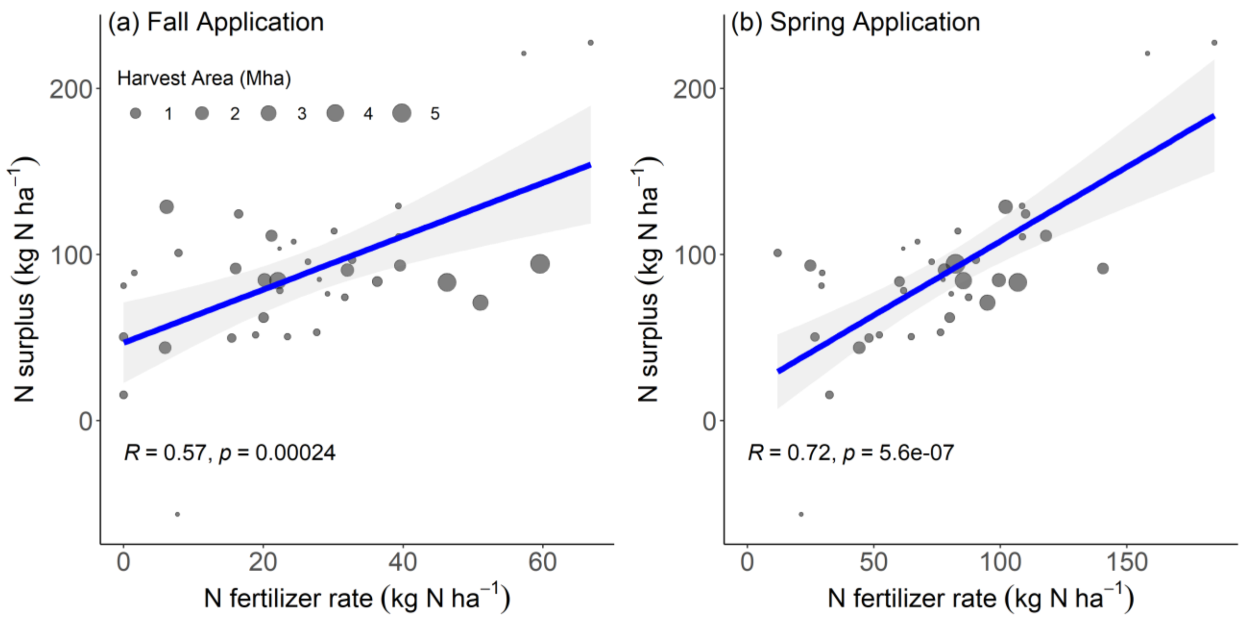

Figure 8. Relationship between state-level corn annual total nitrogen surplus and N fertilizer rates at four application timings from 2010 to 2019. Each dot represents an individual state. The letter "*R*" is the Pearson correlation coefficient. Based on the state-level corn-specific N fertilizer application timing survey published by USDA-ERS, we extract the N fertilizer use during fall (previous year) and spring (before planting). Due to limited data, we adopt the timing information from the latest survey for each state. More details can be found in Cao et al. (2018).