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RESEARCH ARTICLE

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Special Section:

Quantifying Nutrient Budgets for sustainable nutrient management

Key Points:

- Responses of yield and N use efficiency to N fertilizer input exhibit different trajectories between corn and winter wheat and vary across the states
- State-level NUE of corn and winter wheat has improved in recent decades, but peak NUE occurs at a medium level of N fertilizer input
- Interannual variations in N surplus at a national scale are closely tied with grain yields in corn and with N fertilizer use rate in winter wheat

Supporting Information:

Supporting Information S1

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Are We Getting Better in Using Nitrogen?: Variations in Nitrogen Use Efficiency of Two Cereal Crops Across the United States

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Abstract Spatial variation and temporal trajectory of crop nitrogen use efficiency (NUE) have important implications for nitrogen management and environmental conservation. Previous studies have examined cross-nation divergences in crop NUE but often overlooked its spatial heterogeneity and cross-crop differences at subnational scales. We examined the relationship between state-level NUE and nitrogen fertilizer use for two major fertilizer-consuming crops, corn and winter wheat, which account for over half of national N fertilizer use in the United States. Since 1970, as N fertilizer use rates have changed, the responses of crop yield and NUE have exhibited large temporal and spatial variations. It is evident that NUE of corn begins to decline when N fertilizer application rate exceeds ~150 kg N ha⁻¹ yr⁻¹, and that yield response of winter wheat slows down with annual N fertilizer input above \sim 50 kg N ha⁻¹ yr⁻¹. State-level NUE in both crops has risen in recent decades, which could potentially reduce N loss from agricultural production. Across the United States, some major corn-producing states demonstrate a shift from an increasing trend of NUE during the period 1970 to 1999 to a decreasing trend after 2000, whereas winter wheat-producing states present an opposite pattern. Furthermore, this study indicates that annual dynamics of N surplus in corn is closely tied with grain yields, while that in winter wheat significantly correlates with N fertilizer input. A larger proportion of N loss would be anticipated if no further increase in corn yield was obtained or fertilizer use kept rising in winter wheat.

Plain Language Summary There have been growing concerns about a mismatch between agricultural nitrogen supply and crop nitrogen demand. To reduce nitrogen pollution from cultivation, it is essential to know how nitrogen use efficiency (NUE) of crops has changed over space and time, and how far we are from their potentials. Here we examined the NUE patterns in two major fertilizer-consuming crops across the United States since 1970. Despite large spatial variations, we found that NUE in corn and winter wheat has improved in recent decades, potentially reducing nitrogen loss. We also identified a turning point of nitrogen fertilizer use rate for each crop, above which NUE begins to decline or yield response slows down. In addition, our study shows that nitrogen demand and supply play different roles between crops in regulating nitrogen surplus.

1. Introduction

Global use of synthetic nitrogen (N) fertilizer has increased ninefold since the 1960s (Lu & Tian, 2017) and is closely associated with increased agricultural food production worldwide (Cassman, 1999; Hirel et al., 2007). An increasing amount of N fertilizer that is not utilized by crops is lost to the environment via nitrification, denitrification, volatilization, and leaching (Cassman et al., 2002; Tilman et al., 2002). It is reported that 76% of the total anthropogenic N applied to the global land surface returned to the atmosphere, transported to the ocean, or percolated to the surface and subsurface water bodies (Schlesinger, 2009), and that this part of N accounts for nearly 40% of global total N inputs with biological N fixation accounted (Liu et al., 2010). Tremendous N losses cause detrimental impacts on the diversity and functioning of recipient ecosystems (Barak et al., 1997; Bouwman et al., 2002; Kumazawa, 2002; van Meter et al., 2017). Given the increasing N loss, improving crop nitrogen use efficiency (NUE)—the crop production harvested per unit N input—is one of the most effective means for increasing crop productivity and, consequently, alleviating environmental degradation (Davidson & Kanter, 2014; Giller et al., 2004; Howarth et al., 2002).

Understanding crop NUE is critical to address the challenges of food security, environmental degradation, and climate change, from the perspective of both technological innovation and socioeconomics (Zhang et al., 2015). It was concluded that a 20% improvement in "full-chain" NUE by 2020 would lead to an annual saving of nearly 20 million tons (Mt) of global N fertilizer (Sutton et al., 2013). Recently, NUE has been widely proposed as an indicator for assessing progress in agricultural sustainability by many countries (Norton et al., 2015). Increases in NUE have been much larger in North America and Europe than in other regions around the globe (Lassaletta et al., 2016). As of 2007, when we count the part of intentional N inputs entering food, livestock feed, biofuel, and industrial products, the United States national NUE was close to 55% (Houlton et al., 2013). In contrast, flat or even negative NUE responses to crop yield increases were observed in some developing countries, implying that the increases in crop yield in these regions are at the expense of introducing greater N surplus to environments (Ciampitti & Vyn, 2014; Lassaletta et al., 2014; Zhang et al., 2015). For example, in China, grain yield improved by only 28% at the expense of a 55% increase in N fertilizer consumption from 37 Mt in 1990 to 57 Mt in 2010 (Meng et al., 2016).

Although national records of crop yield and N input have facilitated spatial comparisons and syntheses of crop NUE, examining previous trends and responses of crop NUE at a subnational level can help us understand when and where crops have reached the peak of utilizing N, identify the remaining potentials of crops in using N, and determine the critical thresholds of N addition that may deteriorate environmental pollutions. According to a few existing studies, N fertilizer application rate varied among subregions within a single nation, which is likely the results of differences in the physical environment and the state-oriented policy (Bierman et al., 2012; Cao et al., 2018). For instance in the United States, while annual total inorganic N fertilizer use for corn in 2015 varied from 67 kg N/ha in New York to 186 kg N/ha in Missouri (Cao et al., 2018), corn yields were very similar in these two states (9,009 kg grain/ha in New York and 8,946 kg grain/ha in Missouri, based on the statewide average yields summarized by the USDA National Agricultural Statistics Service). Differences in N fertilizer application amount, timing and types, and additional N sources (e.g., manure application, N fixation) may lead to a large spatial heterogeneity of crop NUE and N losses, creating unprecedented challenges for N management practices in agriculture. However, few studies have examined long-term crop NUE trends and their spatial heterogeneity at a subnational scale.

Based on long-term data sets in the United States, our goal is to conduct a spatiotemporal analysis that combines subnational information on crop yields, N fertilizer use rates, and other N input sources for corn (*Zea mays* L.) and winter wheat (*T. aestivum* L.) to better understand how yield response and NUE have changed with fertilizer use rates and to identify critical thresholds of fertilizer uses. The following questions guided this study: (i) how has NUE of corn and winter wheat in the United States changed spatially and temporally? (ii) has NUE of these two crops reached its peak, and when? and (iii) what is the dominant driver responsible for the dynamics of N surplus in these two crops?

2. Methods and Data

We obtained historical state-level crop yield data, from the USDA National Agricultural Statistics Service (NASS; http://www.nass.usda.gov/index.asp) crop databases, and state-level annual N-fertilizer use data developed by a recent study (Cao et al., 2018). Other anthropogenic N sources considered in this study include manure N application and atmospheric N deposition for the two crops, and residual N from previous-year soybean fixation for corn only. We analyzed the responses of yield and NUE to N fertilizer application from 1970 to 2015. To examine the changing trend of state-level NUE for the periods 1970–1999 and 2000–2015, we used autoregressive moving average time series models that account for autocorrelation among residuals. We also analyzed the responses of N surplus to N fertilizer input rates and crop productivity at a national scale.

2.1. Crop Yield and Anthropogenic N Input Data Sources 2.1.1. Crop Yield

The annual yield for each crop in each state was calculated as the sum of total crop production divided by its total harvested area from 1970 to 2015. We used 1970 as the starting point to restrict the analyses to a period when private sectors in the United States have heavily invested in modern hybrids, advanced management practices, and scientific technologies to improve crop production (Evenson & Gollin, 2003; Tilman, 1998). Due to the small planting areas, the state of Nevada was excluded for the analyses of corn, and Maine, Vermont, New Hampshire, Massachusetts, Connecticut, and Rhode Island were excluded for the analyses

of both corn and winter wheat. The summary of the studied states is listed in Table S1 in the supporting information.

2.1.2. N Fertilizer Use

N fertilizer consumption of these two crops accounts for more than 50% of national total N fertilizer use in the United States (USDA-ERS, 2013). Based on the N fertilizer survey conducted by USDA-NASS, we generated state-level crop-specific annual average N fertilizer use rate by multiplying the state-level crop-specific N fertilizer use rate with the percentage of the fertilized area ranging from 1970 to 2015 (Figure S1). The survey records cover approximately 90% of national corn and winter wheat planting area (Table S1). We gap-filled the missing data in the N fertilizer use rates by cubic spline approach when the missing years are no more than three years and by distance-weighted imputation approach for gaps more than three years. For the states without any surveyed data of N fertilizer use, the amount of N fertilizer use rates were adopted in the states where no survey data were reported during the period 1970–2010 and 2014–2015. For winter wheat, national average N fertilizer use rates were only used in the states without survey data from 1970 to 1999. These states only account for ~10% of national planting areas for both crops. Details of the development of N fertilizer use rates and gap-filling are described in Cao et al. (2018).

2.1.3. Atmospheric N Deposition

We obtained 4,134-m resolution N deposition data from 2000 to 2015 from the National Atmospheric Deposition Program (http://nadp.slh.wisc.edu/MDN/annualmdnmaps.aspx), which were then resampled to 5-arc-minute maps in ArcMap 10.4 (ESRI, 2018). We reconstructed the N deposition for the period 1970–1999 by taking the National Atmospheric Deposition Program map of N deposition in 2000 as the endpoint and the trend of N deposition developed by interpolating three-year N deposition data with N emission patterns from EDGAR (Dentener, 2006; Wei et al., 2014). The change rate of EDGAR-derived N deposition between two consecutive years for each pixel was first calculated, which was then used to extrapolate the National Atmospheric Deposition Program-derived N deposition data to the period before 2000.

2.1.4. Manure N Application

The county-level plant recoverable manure N data, which represent the amount of nutrients from excreted manure that would be available to apply to the land as fertilizer, were obtained from the Nutrient Use Geographic Information System (http://nugis.ipni.net/). While the N deposition data cover the entire studied period, the manure N data are only available for the years 1987, 1992, 1997, 2002, 2007, and 2010-2012. We used linear interpolation provided by the stats() package in RStudio 1.1.447 (R core team, 2018) to derive the county-level manure N for the gap years. For instance, manure N application from 1988 to 1991 was linearly interpolated at four equally spaced points spanning the interval between 1987 and 1992. In addition, we assumed the pre-1987 (1970-1986) and post-2012 (2013-2015) manure N levels remained the same as the level of 1987 and 2012, respectively. We then resampled the annual manure N to 5-arc-minute resolution maps in ArcMap 10.4 (ESRI, 2018). To get the crop-specific N addition rate, we extracted the manure N and N deposition data specifically for corn and winter wheat using 5-arc-minute time series crop distribution maps. These maps are aggregated from 1-km maps developed by a recent study (Yu & Lu, 2018), in which the county-level harvested area of each crop type in each year was kept consistent with the county-level survey records provided by NASS, USDA (http://www.nass.usda.gov/index.asp). Then the annual crop-specific manure N application and N deposition rates at a 5-arc-minute resolution were aggregated to the state-level through an area-weighted approach.

2.1.5. N Residues From Previous-Year Soybean Fixation

We also considered the residual benefit of N fixation from previous year soybean cultivation when calculating corn NUE. Using the 1-km crop distribution maps resampled from 30-m resolution cropland data layer data from 2008 to 2017 (https://www.nass.usda.gov/Research_and_Science/Cropland/SARS1a.php), we first identified those pixels in which soybean rotates with corn in three consecutive years with rotation sequences such as soybean-corn-soybean, soybean-corn-corn, or soybean-soybean-corn. We calculated the percentage of soybean that rotated with corn in a three-year moving window to the total soybean acreage in each state each year. Following the same definition, national acreage of soybean that rotated with corn in three consecutive years was reported to account for 93% of total soybean acreage in 1993 (https://www.ers.usda.gov/web-docs/publications/41882/30078_arei4-2.pdf?v=0) and 91% in 1997 (Padgitt et al., 2000). Following the state-level soybean rotation pattern in 2008, we interpolated the aforementioned national rate in 1993 and 1997 to each state and gap-filled the missing years by assuming a linear changing trend during the periods of 1993–

1997 and 1997–2008. Since soybean farming in the United States started in ~1940 (https://ncsoy.org/mediaresources/history-of-soybeans/), we assumed that soybean was 100% used to rotate with corn by then. Linear interpolation was used to fill the soybean rotation percentage in gap years between 1940 and 1992 for each state. The residue of soybean-fixed N that is left for corn grown in year *t*, $N_{fix,t}$, is expressed as

$$N_{fix,t} = \left(a \times Y_{s,t-1} \times H_{s,t-1} \times P \times R\right) / H_{c,t}$$

$$\tag{1}$$

where *a* is the N recovery capacity by per unit soybean yield (0.054 kg N/kg grain) based on the Nutrient Use Geographic Information System-recommended value (http://nugis.ipni.net/), $Y_{s,t-1}$ is the state-level soybean yield in year t - 1, $H_{s,t-1}$ is the state-level harvesting area of soybean in the same year, $H_{c,t}$ is the state-level harvesting area of corn in year t, P is the area percentage of soybean that rotated with corn to the total soybean area in year t - 1, and R is the below-ground residue of fixed N (assumed to be 20% of soybean N recovery according to Nyatsanga and Pierre (1973)). Then we assumed that the residue of N fixation is evenly distributed to corn acreage in the next year. If the rotated soybean acreage in year t - 1, $P \times H_{s,t-1}$, is larger than the harvesting area of corn in year $t(H_{c,t})$, instead of using $P \times H_{s,t-1}$, we used $H_{c,t}$ to calculate the residual N to avoid overestimating N input for corn in the following year. The total amount of residual N from soybean fixation can be found in Figure S2 in the supporting information. According to the cropland data layer, the percentage of winter wheat-soybean rotation was only 1.2% of the total winter wheat planting area in 2008. Therefore, the residual benefit of N fixation by soybean was ignored in our estimates of total N input to winter wheat.

The aforementioned anthropogenic N input sources were summed to get the total N input for assessing annual corn and winter wheat NUE for the period from 1970 to 2015. The percentage and total amount of anthropogenic N input for corn and winter wheat in each state are shown in Figure S3 in the supporting information, and we summarized them to the national level in Figure S2 in the supporting information.

2.2. Calculation of State-Level Crop-Specific NUE and N Surplus

The NUE is calculated as a ratio of crop-recovered N to crop-specific N input in units of kg N/kg N (referring to a percentage (%) of total N input), which is converted from the unit of kg grain per kg N input. The crop-recovered N is calculated as crop yield multiplied by a crop N recovery coefficient, indicating how much N is retained in per unit crop yield. We adopted the N recovery coefficients from the Nutrient Use Geographic Information System (http://nugis.ipni.net/), which is 0.012 kg N/kg grain for corn and 0.0194 kg N/kg grain for winter wheat. We also obtained an annual national yield survey for corn and winter wheat from USDA NASS to calculated national-level NUE.

To represent the potential N losses to the air and water systems, we defined the portion of N that is not recovered in crops as N surplus. This indicator has been extensively used to assess the excessive N input beyond crop N demands, and the potentials of agricultural N loss (Wachendorf et al., 2006; P. Xu et al., 2006). N surplus (in units of kg N ha⁻¹ yr⁻¹) is estimated as total N input received by each crop minus the crop N recovery rate.

2.3. Trend Detection in Crop-Specific NUE Time Series

To examine the temporal trend of NUE time series data, we used models with autoregressive moving average residuals (Box et al., 2015) to account for temporal autocorrelation. Temporal autocorrelation occurs when unexplained variation at one time step has lingering effects over additional time steps (Ives et al., 2010; Michener, 1997); thus, if events that affect NUE in one year have lasting effects in subsequent years, they may generate autocorrelation. In our time series models, "unexplained variation" refers to any variation not captured in the trends of NUE, which are explicitly included in the model. Even though they are "unexplained" in the model, they are usually driven by environmental changes such as weather conditions, market forces that change management practices, and deployment of new technologies. Specifically, Autoregressive Integrated Moving Average ((p,d,q)) model is used to fit the NUE data, where *p* is the order of the autoregressive parameters. The time series are treated as an Autoregressive Integrated Moving Average (1,0,0) process with an autocorrelation process. The model we used to fit the time series of NUE is a linear equation:



$$NUE_t = a + bt + z_t$$

$$z_t = \beta z_{t-1} + t$$
(2)

in which NUE_t represents NUE (kg N/kg N) and z_t the residuals in year *t*, *a* and *b* are the regression parameters, and ε_t is the white noise. The parameter β represents the autocorrelation in the residual (AR-process with lag 1) explaining the unexplained variation. We divided the data into two periods, 1970–1999 and 2000–2015, and used the linear regression coefficient to analyze the trend of each time series. To give a measure of statistical support for the linear trends, we compared the likelihoods between models with and without the linear trends. Specifically, we computed the log-likelihood ratio (LLR) between the model produced by equation (2) including the linear term ($b \neq 0$) and the model without the linear term (b = 0). For a single time series, the LLR between nested models (if the parameters in Model A are a subset of the parameters in Model B, Model A is nested in Model B) approximately follows a chi-square distribution. In such a case, the LLR gives a significance test for the parameters that differ between models. For models that differ in a single parameter, if the values of double the LLR are greater than 3.84 (i.e., LLR values are greater than 1.92), the alternative model is significant at the 0.05 level. In our case, time series NUE values spanning from 1970 to 2015 were obtained in 41 and 42 states for corn and winter wheat, respectively.

3. Results

3.1. Yield Response in Corn and Winter Wheat to N Fertilizer Across the United States

Cross-state yield responses in corn demonstrate a three-stage trend as the N fertilizer application rates increase (Figure 1a). With annual N fertilizer input increasing from 25 to 100 kg N/ha, corn yield increased from 3 to 6 t grain/ha, most of which occurred in the 1970s and the 1980s. When the N fertilizer use level moved up to the range between 100 and 150 kg N ha⁻¹ yr⁻¹, corn yield showed a quick jump from 6 to 8 t grain/ha. This jump is consistent with the national yield responses to N fertilizer use, which is shown by the red circles and the dashed linear regression line in Figure 1a. It is noteworthy that the largest yield response was found at N fertilizer use rate around 150 kg N ha⁻¹ yr⁻¹, above which state-level corn yield leveled off, despite large cross-state variations (indicated by the width of the shaded area around the fitted trend curve). Corn yield responses to N fertilizer input after 2000 (shown by dark blue dots and the cyan trend curve in Figure 1a) were generally higher than that with all years considered (shown by the blue trend curve and the grey shaded area). Our analysis also demonstrates increases in the harvested area of corn and intensive corn farming in major states (larger size dots in recent years).

Cross-state yield responses in winter wheat demonstrate a two-stage trend, with a cutoff value of the N fertilizer application rates at ~50 kg N ha⁻¹ yr⁻¹ (Figure 1b). The yield of winter wheat steeply increased as N fertilizer application rate increased from nearly 0 to 50 kg N ha⁻¹ yr⁻¹, which occurred in the 1970s and 1980s, and then became less steep when N fertilizer rate exceeded this rate, which occurred mostly during the periods after the 1990s. Likewise, national-level crop yield responses to N fertilizer input demonstrate a medium increasing trend with a narrower range of fertilizer use rate. Compared to the trend with all years included, the post-2000 yield response of winter wheat showed a similar trend but with a slightly higher magnitude. In addition, the peak of winter wheat yield response was evident with N fertilizer input around 50 kg N ha⁻¹ yr⁻¹.

3.2. Nitrogen Use Efficiency Responses to N Fertilizer Use Across the United States

Corn NUE first declined with low levels of N fertilizer use rates (25 to 110 kg N ha⁻¹ yr⁻¹), and then slightly increased with medium levels of N fertilizer use rates (110 to 150 kg N ha⁻¹ yr⁻¹). Interestingly, corn NUE decreased again with a further increase of N fertilizer, with a peak of ~0.5 kg N/kg N at the N fertilizer use rate of around 150 kg N ha⁻¹ yr⁻¹ (Figure 1c). However, the national corn NUE only demonstrates a linearly increasing trend (shown by the red dots and the red linear trend line with a shaded area) with a narrower N fertilizer input range and misses this nonlinear NUE pattern observed at the state level. Corn NUE with a larger harvested area after 2000, which is shown by the large size of dark blue dots, was generally higher than those before 2000. In the states with 3 Mha or more corn harvested area, corn NUE presents a similar nonlinear trend (shown by the green trend line and the shaded area), while the magnitude is higher than that with all harvested areas included. The corn NUE of these regions reached



Figure 1. The relationship between state-level yield and N fertilizer input rate for (a) corn and (b) winter wheat from 1970 to 2015, and the relationship between state-level NUE and N fertilizer input rate for (c) corn and (d) winter wheat from 1970 to 2015. The blue solid curve is the "loess" fitted correlation for each pair of the variables for all the analyzed years (1970–2015), and the cyan dashed line is the loess fitted correlation between yield and N fertilizer rate after 2000. The green dashed lines are the loess fitted correlation between NUE and N fertilizer rate in those states where planting areas of corn or winter wheat are more than 3 and 1 million hectares (Mha), respectively. The shaded areas around the fitted curves indicate the 95% confidence interval. The national annual data from 1970 to 2015, which are from the NASS data set, are shown by the red dot, and the red dashed line indicates the linear trend for each pair of the variables at a national level. The size of the data dots indicates the harvested area in million hectares, and the color gradient stands for the year ranging from 1970 to 2015.

the peak with N fertilizer use rate around 140 kg N ha^{-1} yr⁻¹, which is about 10 kg N ha^{-1} yr⁻¹ less than that when all the states are considered.

In contrast to corn, the state-level NUE of winter wheat showed a sharp decline with N fertilizer input rate ranging from 0 to 30 kg N ha^{-1} yr⁻¹ and then leveled off with a further increase of N fertilizer input rate (Figure 1d). The >1.0 kg N/kg N NUE of winter wheat was likely due to the small amount of N fertilizer input, which indicates that other sources of N might contribute to the N recovery of winter wheat, that is, soil mining (Huggins & Pan, 1993), which were not considered in this study. There are large variations in winter wheat NUE across the entire spectrum of N fertilizer input rates, especially when N fertilizer rates ranged from 30 to 60 kg N ha^{-1} yr⁻¹. If we excluded those states with low fertilizer input (i.e., 0 to 30 kg N ha^{-1} yr⁻¹), our analysis indicates that winter wheat NUE generally reached the peak with N fertilizer rate at ~50 kg N ha^{-1} yr⁻¹ in the 2000s and the 2010s. Compared to corn, the NUE of winter wheat was less varied over time, although N fertilizer use rates in the 2000s and the 2010s were slightly higher than those in previous decades. The national-level NUE trend in winter wheat not only showed a slightly decreasing trend with N fertilizer input rates from 3 to 80 kg N ha^{-1} yr⁻¹ but also, surprisingly, presented a small year-to-year variation. Contrary to corn, harvested areas of winter wheat have been declining since the 1990s (see Figure S4 in the supporting information). The states with larger winter wheat harvested area (\geq 1.0 Mha) on average exhibited a lower NUE (shown by the green trend curve) than that with all states accounted.



Figure 2. NUE trend of (a and b) corn and (c and d) winter wheat in each state of the continental United States during the periods 1970–1999 and 2000–2015. The red and blue colors indicate increasing and decreasing trends in NUE, respectively. Grey color indicates a flat trend. The unit of NUE trend is in kg N kg⁻¹ N yr⁻¹. The yellow asterisk indicates that the trend is significant by the log-likelihood ratio test. National average crop-specific N fertilizer use rates are adopted for those states masked by oblique lines because no survey data were reported in these states during the study periods. An exception is the period of 2011–2013, in which all-state survey data are available for corn (national average rate is partially used in oblique-line masked states in Figure 2b).

3.3. Spatial Pattern of NUE Changing Trend Across the United States

The pre- and post-2000 cross-state NUE trends showed a distinct spatial pattern between corn and winter wheat (Figures 2 and S5). During the period from 1970 to 1999, only 3 out of 41 states showed a decreasing or a flat trend of corn NUE, while the remainder of the states experienced increasing NUE (Figure 2a). Since 2000, the increasing trend of NUE in most corn-producing states has switched to a marginally decreasing trend or remained flat (Figure 2b). During the same period, corn NUE increase rate peaked at 0.02 kg N kg $^{-1}$ N yr⁻¹ in some southeastern and northeastern states such as New York and Mississippi.

The NUE of winter wheat showed a decreasing trend in most states from 1970 to 1999; however, a number of states have been characterized by increasing NUE since 2000 (Figures 2c and 2d). The largest NUE increase rate (above 0.1 kg N kg⁻¹ N yr⁻¹) was found in Arizona. In contrast, the remaining states, including Oklahoma, Idaho, Montana, South Dakota, Iowa, Georgia, and West Virginia, kept a decreasing trend in winter wheat NUE over the entire study period, although this decreasing trend became smaller or insignificant after 2000.

3.4. Relationships Between Crop Yield, N Surplus, and N Fertilizer Use Rate Across the United States

Using the area-weighted statistics, our analyses demonstrate that national annual average yield and N fertilizer input have increased since 1970 in both corn and winter wheat (Figures 3a and 3b). Specifically, inorganic N fertilizer applied to corn on average plateaued around 140 kg N/ha after 2000, while N fertilizer applied to winter wheat leveled off around 60 kg N/ha since 2007. The yield and N fertilizer rate at the national scale show a positive correlation for both corn ($R^2 = 0.45$) and winter wheat ($R^2 = 0.53$), indicating that N fertilizer input played an important role in enhancing the yield of these two crops (Figures 3c and 3d). The national-level N surplus in corn is negatively correlated to crop yield ($R^2 = 0.44$), while N surplus in winter wheat is positively associated with the N fertilizer input rates ($R^2 = 0.72$; Figures 3e and 3h). This implies that the dynamics of N surplus was mainly demand-driven in corn and supply-driven in winter wheat.





Figure 3. (a and b) Time series of national yield (t ha⁻¹ yr⁻¹) and N fertilizer use rate (kg N ha⁻¹ yr⁻¹) and the relationship between (c and d) yield and N fertilizer use rate, (e and f) N surplus (kg N ha⁻¹ yr⁻¹) and yield, and (g and h) N surplus and N fertilizer use rate in the United States during 1970–2015. The left column is for corn, and the right column for winter wheat.

4. Discussion

This study provided a comprehensive analysis of NUE and yield responses to N fertilizer input of two major crops in the United States using the long-term data. We found that a few major corn- and winter wheat-producing states have passed their NUE peaks and are likely to lose more N if anthropogenic N input kept rising (Figure S5). The NUE of corn was enhanced in most states after 2000, and it was shown to decline when N fertilizer application rate exceeded ~150 kg N/ha. We also found a slight decline in the NUE of winter wheat as N fertilizer use rate exceeded 50 kg N/ha, with its yield response slowing down with N additions. However, large variations in corn and winter wheat NUE still existed among states, indicating that the specific environmental conditions and differences among crop varieties should be considered while N management recommendations are provided at local scales. Across the United States, our analysis in the major corn-

producing states (harvesting area \geq 3.0 Mha) demonstrates an earlier turning point of NUE when N fertilizer rate passes 140 kg N/ha, whereas major winter wheat-producing states (harvesting area \geq 1.0 Mha) present a leveled-off pattern similar to the national average. This study provided insights for understanding and investigating the long-term responses of crop yield and NUE to N fertilizer input and identifies the potential for N fertilizer management from state to national scales.

4.1. Comparison of NUE in Corn and Winter Wheat With Previous Studies

Previous studies have shown a rapid yield response to medium-level N fertilizer use rates ($125-175 \text{ kg N ha}^{-1} \text{ yr}^{-1}$) in corn production of the United States by using national data of yield and N fertilizer consumption (Lassaletta et al., 2014; Zhang et al., 2015). In this study, we identified NUE declines in corn in response to a high level of N input rates, particularly in the intensive corn producing states after 2000 (e.g., Corn Belt states), which have been missed in national-scale analyses. On the other hand, although the NUE trend in winter wheat kept relatively stable with N addition, state-level data revealed an NUE peak at N fertilizer level of 50 kg N ha⁻¹ yr⁻¹. These findings indicate that the spatiotemporal variabilities in crop yield, NUE, and N fertilizer use rate are likely to be averaged when within-nation details are aggregated. Therefore, it is critical to develop subnational analyses for understanding the role of N addition in promoting crop production and future N loss potentials, especially in areas with a large amount of N fertilizer input.

Crop NUE has been used as a performance indicator for agricultural production and a policy tool for regulating N fertilizer use (Erisman et al., 2018; Powell et al., 2010). For example, beginning in the late 1980s and through the early 2000s, increases in NUE in several European Union countries coincided with changes in the European Union Common Agricultural Policy, which reduced crop subsidies, and adoption of the European Union Nitrates Directive, which limited manure application rates on cropland (van Grinsven et al., 2012, 2015). Within the United States, increases in NUE since the 1990s were largely caused by increasing crop yields, resulting from improved crop varieties, increased irrigation, and other technological improvements (van Grinsven et al., 2015; Xu et al., 2013), and steady input of N fertilizer, resulting from regulatory programs of nutrient management (Ferguson, 2015). However, limited by the unavailability of a longterm crop-specific database, comparing crop NUE is therefore challenging, and heterogeneity among countries should be interpreted cautiously till the methods and data are proven comparable (Erisman et al., 2018).

The timing of N fertilizer application driven by policy and market also affects NUE. In the U.S. Midwest, for instance, fall N application, which is proven to have negative impacts on ecosystem services, is still popular largely due to the lower fertilizer prices (Dinnes et al., 2002). The ratio of fertilizer to crop prices, together with yield responses to fertilizer input, have been widely used to advise farmers on fertilizer application rates to maximize economic returns (Robertson & Vitousek, 2009; Setiyono et al., 2011). Our state-level results confirmed that finer-scale analysis (e.g., county-level crop-specific NUE) is likely to do a better job for disentangling the relationship among crop NUE, yield responses, and the consequent economic returns. More importantly, our study reconfirmed that future studies on crop NUE should cautiously interpret the national estimates, especially while advising policy.

4.2. Reasons for the Variations of NUE in Corn and Winter Wheat

Although cereal yields and fertilizer N consumption have increased in a highly correlated near-linear fashion during the past decades, large differences existed in the historical trends of N fertilizer usage and NUE among world regions and crops (Dobermann & Cassman, 2005). Improved NUE in the United States has been achieved by increased stress tolerance of modern hybrids, improved management of crop production such as conservation tillage, higher plant densities, and improved N-fertilizer management (Cassman, 2017; Cassman et al., 2002; Duvick & Cassman, 1999; Ferguson, 2015). In this study, however, we still found that state-level data in corn and winter wheat both feature a wide spread of crop yield under the same N fertilizer application level within the United States. Likewise in Europe, large variations in NUE of foodproducing systems were recently reported for the period of 1980–2011 (Erisman et al., 2018). The N uptake capacities and their controlling factors are different between corn and winter wheat (Delogu et al., 1998; Scharf et al., 2002; Vanotti & Bundy, 1994). Their NUE variations were likely derived from a broad spectrum of soil fertility, local climate conditions, other management practices, and different capability of N uptake and yield among crop varieties (Kang, 1997; Peterson et al., 1992). For instance, historically within the United States, a combination of freezing and warming impacts were found to be responsible for wheat yield variations in Kansas (Tack et al., 2015), and large variations in corn yield were found closely related to climatic variation in the Midwest (Hatfield et al., 2018). Other than climate impacts, crop genotypes have a genetic variability in removing nitrogen (Fageria & Baligar, 2005), which can largely result in variations in NUE (Duvick, 2005).

The N-management improvements become increasingly popular, including shifting fall application to spring, using multiple small N-fertilizer applications to replace a single large N application (Zhang et al., 2012), promoting organic agriculture (Scialabba & Müller-Lindenlauf, 2010), implementing rotations (Dias et al., 2015), and using residual soil nitrate as N sources (Bakhsh et al., 2000). Despite these advantageous factors, differences in the scale of farming operations still likely result in variations in NUE (Cassman et al., 2002). Sustaining NUE of various crop hybrids should consider improving not only crop genotypes but also N management and the dynamics of environmental conditions such as solar radiation, temperature, and moisture regimes.

4.3. Crop Yield and NUE Response to Further N Fertilizer Input

Our results demonstrated that NUE declined and yield plateaued in corn at high levels of N fertilizer addition, implying that an increase in corn yield may not be achieved by current hybrids or by traditional N fertilizer types. For example, in two major corn-producing states, Indiana and Illinois, we found that NUE plateaued and decreased during 2000–2015, although fertilizer input kept increasing in these two states (see Figure S5 in the supporting information). A recent study in China also found that N uptake first decreased and then plateaued with grain yield improvement (Meng et al., 2016). Through a three-year field experiment, however, another study evaluated the performance of enhanced efficiency fertilizers on corn grain yield in Iowa, USA, and found a consistent higher yield with these types of fertilizer application, compared to the traditional N fertilizers (Hatfield & Parkin, 2014), indicating that fertilizer improvement likely stimulates further yield increases for corn. However, it remains untested whether these enhanced efficiency fertilizers can sustain a long-term corn yield increase. Currently, most of the corn-growing areas still rely on traditional N fertilizers. During the period of 1970 to 2015, ammonium-N is the dominant fertilizer type across the United States (Cao et al., 2018), which has a high potential for volatilization loss and leaching loss after being nitrified. Therefore, N loss can be expected if future N fertilizer management relies on traditional fertilizer types. Studies focusing on the consistency of new fertilizer types in boosting long-term yield are needed.

A decreasing trend of NUE occurred in winter wheat before 1990. While the N fertilizer use rate increased from a low level since the 1970s, the winter wheat yield remained stable in most states from 1990 to 1999, which results in a decreasing NUE trend across the nation (see Figure S5 in the supporting information). From 2000 to 2015, although the national average N fertilizer use rate in winter wheat has decreased by ~ 20 kg N⁻¹ ha⁻¹ yr⁻¹ (Cao et al., 2018), its yield and NUE in most states substantially increased in the same period (Figures 2d and 3b). This recent jump in winter wheat yield was likely due to the U.S. Conservation Reserve Program, which converts highly erodible cropland to environmentally beneficial uses (Farm Service Agency, USDA; https://www.fsa.usda.gov/). On average, 20% of winter wheat planting area was categorized as highly erodible cropland and was converted to reserve land by the Conservation Reserve Program, leaving productive lands to support a higher NUE (Vocke & Ali, 2013). A recent study evaluating a panel of 407 winter wheat cultivars for six characteristics of spike and kernel development suggested that some of the key traits were the basis of grain yield gains over the past decades (Würschum et al., 2018). This experiment points out that further exploitations of the available trait variations, integrated with genomic approaches, may assist wheat breeding in continuing to increase yield levels globally. Generally, due to its C3 plant constraints in utilizing water and nutrients, wheat has a smaller NUE than C4 crops such as corn (Cassman et al., 2002). The N loss from wheat fields would be triggered more easily than C4 crops, especially when application timing is out of step with its growth phases (Meng et al., 2016).

4.4. N Loss Risk at the State and National Level and Recommendations for Future N Management

The ultimate goal of agriculture is to achieve food security, sustainability, and ecosystem services efficiently at regional and global scales (Foley et al., 2011). To meet global cereal demand, a 60% increase in global N use would be required if NUE cannot be increased substantially, which would eventually result in major environmental issues (Dobermann & Cassman, 2005; Ladha et al., 2016). Our analyses suggested that the N surplus is likely reduced by the increased yield in corn and the resultant increase in N uptake

(Figures 3g and S6a). Using 15-N isotopes, a study showed that a spring N application led to increases of 0.5 to 1.2 t/ha in yields in corn, resulting in ~9-kg N/ha reduction in N loss (Buzicky et al., 1983). However, corn yield would likely level off when N fertilizer exceeds its threshold level at approximately 150 kg N/ha, with large cross-state variations revealed from our analyses (Figure 1a). Hence, without a considerable crop variety improvement to produce higher yield, N fertilizer above such level will likely increase the risk for N loss in the United States. Interestingly, in the major corn-producing states, we found that the critical threshold of N input rate was 10 kg N/ha lower than that with all states considered. Compared to the remaining areas, consistently intensive N fertilizer use in these states may lead to an accumulation of residual soil N, which is likely to push the N application threshold lower than the national average. Therefore, for these states, efficient N management in corn fields is of particular importance for policy- and decision-making that aims at reducing N-related air and water pollution (Mitsch et al., 2001; Rabalais et al., 2002).

Interannual variation in N surplus from national corn cultivation is shown to be negatively correlated to the corn yield, implying that rising grain gain can potentially increase crop N demands and reduce N loss. However, winter wheat yield only explains ~8% of interannual variations in N surplus (Figure 3f). Although winter wheat yield was generally responsive to N addition at both state level and national level (Figures 1b and 3d), N surplus in winter wheat was predominantly controlled by N fertilizer input. We ascribed the different drivers of N surplus between corn and winter wheat to the following reasons. First, the majority of N fertilizer is applied to both cropping systems before planting in the United States, with March-April for corn and August to September for winter wheat (Cao et al., 2018). Conducting experiments in Oklahoma in 2006 and 2007, Girma et al. (2011) demonstrated that more than 45% of the total N accumulated in corn by growth stage V8 (the eighth leaf collar fully unfolded, temporally around late May) and more than 61% of the total N accumulated at later growth stages (F5, temporally around March) of winter wheat. Such finding suggests that the peak of N demand can be lagged behind the peak of N supply for five to six months for winter wheat but just one to two months for corn. Therefore, the amount of N fertilizer input, rather than yield or crop N demands, is a stronger driver for N surplus variations in winter wheat. Second, the grain N content of winter wheat is generally higher than that of corn, despite receiving a lower level of N fertilizer input. It implies a larger N recovery capacity of winter wheat, or in other words, a lower N demand to support the same amount of grain production compared to corn. Last but not least, other N sources, soil conditions, and management practices (e.g., irrigation) may substantially affect yield gain of winter wheat and eventually affect NUE and N surplus (Ladha et al., 2016). For example, N recovery rate could be up to 120% with conventional tillage for wheat in the Great Plains in the United States, indicating the occurrence of soil mining of N (Huggins & Pan, 1993). Higher grain yield and NUE of wheat can be achieved with low application rates of N fertilizer if the crop is irrigated with treated effluent containing nitrogen (Hussain et al., 1996). Overall, loose coupling between N fertilizer input and winter wheat yield gain (due to timing, crop N demand, and additional N sources) implies that interannual variation in winter wheat N surplus is more regulated by fertilizer input, rather than by grain yield. Extensive farm-level observations across diverse conditions of climate, soil, and management regimes are expected to reveal the dominant factors controlling NUE variations in winter wheat across a region.

5. Conclusion

This study comprehensively examined historical yield responses to N input for two major N fertilizerconsuming crops in the United States, corn and winter wheat, to study spatiotemporal changes in NUE and to detect critical thresholds in N fertilizer input rates. To estimate yield responses to changing N inputs, this study combined state-level crop-specific data of grain yield, N fertilizer use, manure application, and atmospheric N deposition from 1970 to 2015 for two crops, and residual N from previous-year soybean fixation for corn only. We also examined the relationship between annual N surplus and N demand (i.e., crop yield) and supply (i.e., N fertilizer input) for national corn and winter wheat planting during the past half century.

The state-level analysis indicated that corn yield plateaued and its NUE decreased at N fertilizer use rate above 150 kg N ha⁻¹yr⁻¹, albeit with large spatial variations. Although yield and NUE of winter wheat are less responsive than corn with increases in N input rate, we also found that NUE of winter wheat peaked at a fertilizer use rate of ~50 kg N ha⁻¹ yr⁻¹, and yield response slowed down as N input goes beyond that.

Our results clearly showed that NUE began to decline in some key corn-producing states, which suggests that N loss is likely aggravated if N fertilizer in these states continues increasing. Although NUE increase in winter wheat was found in many states after 2000, we are not optimistic to conclude a larger increase in the future. Our results indicated that the post-2000 NUE increase in winter wheat was likely caused by declined N fertilizer input, conversion of the unproductive field to Conservation Reserve Program, breed-ing technology improvement, and advanced management practices. Our national time series data revealed different roles of N demand and supply in affecting N surplus variations for corn and winter wheat. It implies that increasing yield is likely to reduce N surplus from corn planting while lowering N fertilizer supply is shown to contribute to reducing N surplus from winter wheat. More work is needed to identify ways that can maintain or improve crop NUE and reduce N loss across the United States.

Authors' Contributions

C.L. designed the research. J.Z. and P.C. compiled the nitrogen database. J.Z. compiled the crop yield database and carried out the data analysis. C.L. and J.Z. led the writing of the paper. All co-authors contributed to and reviewed the manuscript.

Data Availability

All data used in this study are publicly available. Crop yield data are from the USDA NASS survey at http:// www.nass.usda.gov/index.asp. The N deposition data can be accessed from the National Atmospheric Deposition Program (http://nadp.slh.wisc.edu/). The manure N data can be downloaded from the Nutrient Use Geographic Information System (http://nugis.ipni.net/). The N fertilizer use data set is publicly available via PANGAEA at https://doi.pangaea.de/10.1594/PANGAEA.883585. The cropland distribution map used to extract crop specific manure N and N deposition is available via https://doi.pangaea.de/ 10.1594/PANGAEA.881801. The corn and winter wheat maps used to process the residual N of corn-soybean fixation are available at the CDL database (https://www.nass.usda.gov/Research_and_Science/Cropland/ SARS1a.php).

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