Riverine nitrogen footprint of agriculture in the Mississippi-Atchafalaya River Basin: Do we trade water quality for crop production?

Chaoqun Lu[1*], Jien Zhang[1], Bo Yi[1], Ignacio Calderon[1], Hongli Feng[2], Ruiqing Miao[3], David Hennessy[2], Shufen Pan[4,5], Hanqin Tian[6]

[1] Department of Ecology, Evolution, and Organismal Biology, Iowa State University, Ames, Iowa, USA

[2] Department of Economics, Iowa State University, Ames, Iowa, USA

[3] Department of Agricultural Economics & Rural Sociology, Auburn University, Auburn, Alabama, USA

[4]Department of Engineering and Environmental Studies Program, Boston College, Chestnut Hill, MA 02467, USA

[5]College of Forestry, Wildlife and Environment, Auburn University, Auburn, AL 36830, USA

[6]Schiller Institute for Integrated Science and Society, Department of Earth and Environmental Sciences, Boston College, Chestnut Hill, MA 02467, USA

* Corresponding author: clu@iastate.edu

Abstract Increasing food and biofuel demands have led to the cascading effects from cropland expansions, raised fertilizer use, to increased riverine nitrogen (N) loads. However, little is known about the current trade-off between riverine N pollution and crop production due to the lack of predictive understanding of ecological processes across the land-aquatic continuum. Here we propose a riverine N footprint (RNF) concept to quantify how N loads change along with per unit crop production gain. Using data synthesis and a well-calibrated hydro-ecological model, we find that the RNF within the Mississippi-Atchafalaya River Basin peaked at 1.95 g N kg^{-1} grain during the 1990s, and then shifted from an increasing to a decreasing trend, reaching 0.65 g N kg^{-1} grain in the 2010s. This implies decoupled responses of crop production and N loads to key agricultural activities approximately after 2000, but this pattern varies a lot among sub-basins. Our study highlights the importance of developing a food-energy-water nexus indicator to examine the region-specific trade-offs between crop production and land-to-aquatic N loads for achieving nutrient mitigation goals while sustaining economic gains.

Keywords Riverine Nitrogen footprint, agricultural management, crop production, nitrogen loading, land use land cover change, crop rotation, Mississippi Atchafalaya River Basin

Synopsis We use an innovative concept, the riverine N footprint (RNF) concept, and a data-model framework that couples land-aquatic processes to investigate the relationship between water quality and crop production.

Introduction

The hypoxic zone in the northern Gulf of Mexico is greatly influenced by the high nutrient (e.g., nitrogen and phosphorus) loads from the Mississippi Atchafalaya River Basin (MARB) (Boesch *et al* 2009, Lohrenz *et al* 2008, Turner and Rabalais 2003), which degrades biodiversity, impairs ecosystem function and threatens coastal economies (Altieri *et al* 2017). A wide variety of studies have examined the natural and anthropogenic drivers of streamflow and nutrient loading variations across the MARB. Climate variations, precipitation in particular, has been identified as an important contributor to the year-to-year variations in N loads through altering water discharge and biogeochemical processes (Battaglin et al., 2010, Smits et al., 2019, Lu et al., 2020). Meanwhile, intensive agricultural production serves as the principal catalyst for the growing nutrient loads in riverine environments, primarily attributed to the expansion of agricultural lands (Lambin and Meyfroidt 2011) and widespread increases in nitrogen (N) fertilizer uses (Stets *et al* 2020, Van Meter *et al* 2018, Oelsner and Stets 2019). With cropland encompassing roughly 58% of the basin's land area, the MARB drains about 41% of the contiguous United States and yields an economic value exceeding $100 billion, stemming from the farming and fishing sectors (Goolsby et al., 1999). Collectively, these attributes make the MARB an ideal testbed to understand the interplay of the Food-Energy-Clean Water nexus, and to quantitatively assess the relationship between water quality management and the attainment of sustainable food and biofuel production under climate changes (Gordon *et al* 2010).

Over the past several decades, cropland expansion required to meet the world's rising food and biofuel demand has increased the total N export (Seitzinger *et al* 2010). The increased cropland area is closely linked to the cumulative N input within agricultural systems (Lu et al., 2019). It also determines the accumulation of available soil legacy N that is readily mobilized when crop production decreases (Lee *et al* 2016). In the MARB, both USDA census data and process-based ecosystem modeling (Dynamic Land Ecosystem Model-DLEM in this case) show grain crop production in the 2010s increased by 75% compared to the 1970s' average, with increasing rates of 4.3–5 million metric tons (MMT) yr^{-1} (range of model estimate to survey average, $p < 0.01$, Figure 1a). To meet the increases in crop production in the Basin, total N fertilizer input increased by almost 43% from 1970 to 2017, rising from 4.4 Tg yr^{-1} in the 1970s to 7.7 Tg yr^{-1} in the 2010s (Figure 2). However, dissolved inorganic N (DIN) exports from the MARB to the Gulf, estimated by both water quality monitoring data (i.e., the USGS LOADEST, WRTDS) and the process-based modeling, demonstrate large inter-annual variations with a slightly decreasing trend during 1980-2017, except for a few peak years (e.g., 2008 and 2017 Figure 1b). We hypothesized that the decoupling between increasing agricultural N input and the "near-flat" trend in riverine N load from the MARB would be caused by a looser connection between crop production and water quality. To test this hypothesis, in this study, we used a process-based modeling approach and historical time-series datasets to quantify how key agricultural activities have regulated inorganic nitrogen loads while changing crop production (in the concept of riverine N footprint, i.e., the ratio of ΔN loads to Δcrop production) and how their impacts varied over space and time across the MARB.

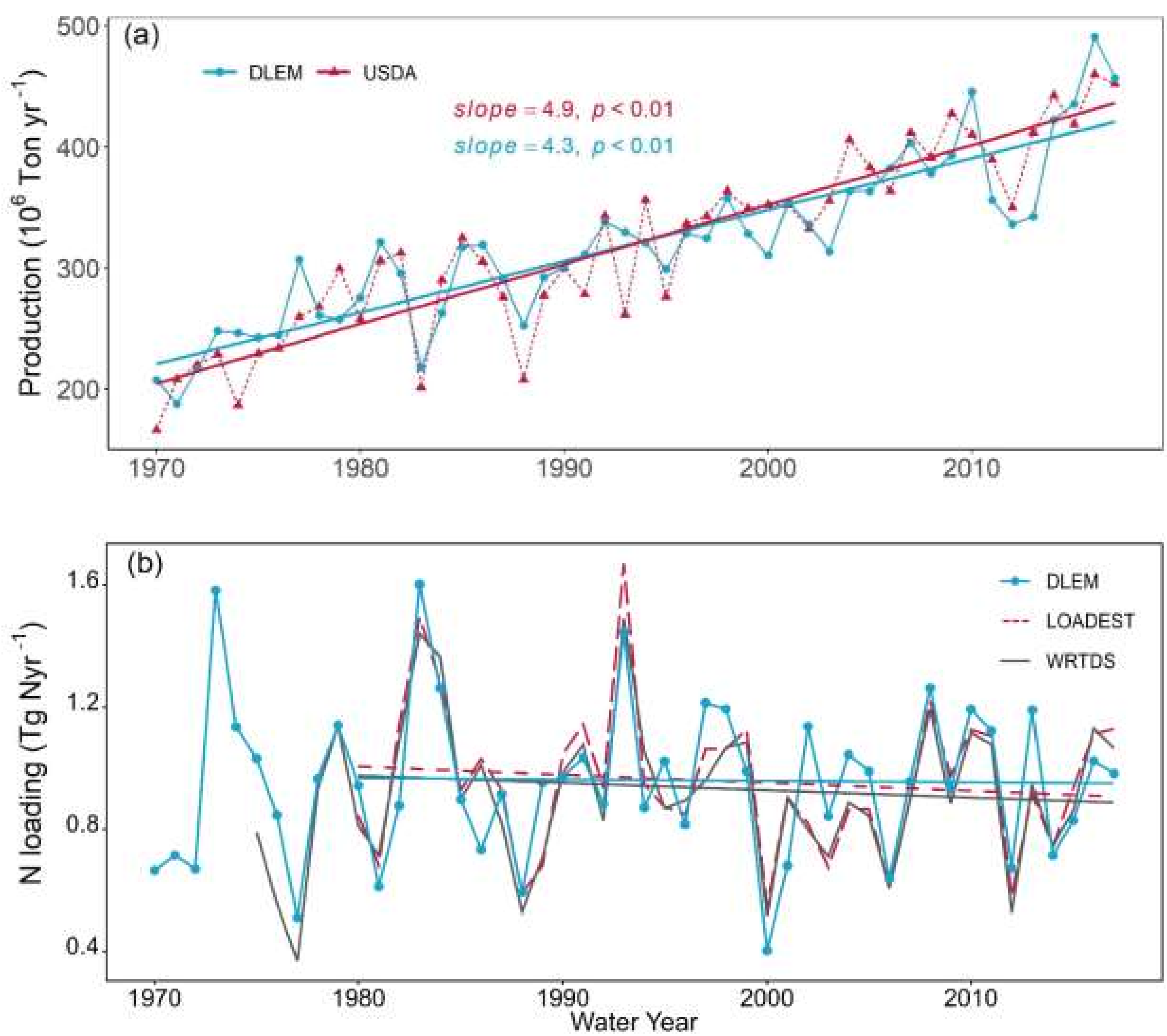

Figure 1. Annual total grain crop production and its trend as estimated by the Dynamic Land Ecosystem Model (DLEM) and USDA survey (a), and annual DIN load estimated by DLEM, the USGS LOADEST, and Weighted Regression on Time Season and Discharge (WRTDS) (b) in the MARB during 1970–2017. Note that the LOADEST N load data starts in 1980 and WRTDS starts in 1976 due to the limited data availability. The post-1980 estimates of DIN loads show a near-flat long-term trend with large inter-annual variations (slope of DLEM estimates = -0.0005, p=0.90; slope of LOADEST = -0.0026, p=0.46; slope of WRTDS = -0.0024, p=0.48).

To maintain the quality of the agroecosystems on which human society relies, modern agricultural activities seek the most sustainable synergy between increasing crop production and reducing N loads to the environment. Among these activities, some are primarily used to increase crop production and crop N uptake. For example, genetic improvement of crop varieties is widely adopted to boost crop yield and maximize economic profits (Evenson and Gollin 2003a, 2003b). Other activities, such as organic fertilizer application and crop rotation systems are more focused on reducing fertilizer input into the fields as well as lowering N loss to the environment (Wei *et al* 2020, Ma *et al* 2007). Despite previous studies that have examined how basin-wide agricultural management affects N export to the northern Gulf (National Research Council 2009, Marshall *et al* 2018, Robertson and Saad 2021, 2013), a comprehensive analysis quantifying the trade-offs between N pollution and crop production, driven by key agricultural activities, has not emerged. This is partially caused by limited modeling capability in tracking the carbon and nitrogen cycling in the plant-soil-water-river continuum, such as simulating crop production and riverine N loads at the same time. Additionally, it remains unclear how the agricultural activities driving the rise of crop production have contributed to riverine N loads among the sub-basins of the MARB. Insufficient understanding and quantification of food-energy-water conflicts hinders our ability to identify the alternative practices and assess the related costs to arrive at these alternatives (Tian *et al* 2018).

The DLEM model we used in this study is capable of simultaneously simulating plant physiology, biogeochemical and hydrological cycles, and vegetation dynamics at a daily time step. The DLEM can estimate the fluxes and pool sizes of carbon, nitrogen, and water in terrestrial ecosystems. At the same time, DLEM can simulate the transfer and delivery of carbon and nitrogen from terrestrial ecosystems to aquatic systems (e.g., streams, rivers, and lakes). In this version of DLEM, we emphasize crop-specific management practices. Agricultural management practices considered in this study include nitrogen fertilizer use, manure nitrogen application, irrigation, tillage, tile drainage, crop rotation, and technology improvement on crop yield at various time steps (Cao *et al* 2018, Yu and Lu 2018, Yu *et al* 2018). Along with the management practice data, we use the prescribed input data, such as data on daily climate (average, minimum, and maximum temperature, precipitation, and shortwave radiation), annual land use patterns, monthly concentration of atmospheric CO_2, and annual nitrogen deposition, to characterize the key environmental changes and to drive the model simulations. We also parameterize the model to capture the magnitude and historical trajectories of yield for multiple major crops, including corn, soybean, wheat, rice, cotton, barley, sorghum, and so on. The DLEM is shown to capably capture the long-term trend and inter-annual variations in crop production and N loading in the MARB during 1970-2017 (Figure 1, Figures S1-S7) (Lu *et al* 2018, 2020, 2021, Yu *et al* 2019, Tian et al., 2020, Zhang et al., 2022).

By using a series of simulation experiments, we attributed the relationship between riverine N loading and crop production to four key agricultural activities from 1970 to 2019. The impacts of other drivers and point-source contributions have been excluded in this study as we keep them constant across the simulations and they cancel out in calculating the impacts of drivers of interest. The agricultural activities examined in this study include synthetic N fertilizer application (referring to N fertilizer), manure N application, land use/land cover change (LUCC, including cropland expansion, abandonment, and inter-annual crop rotation), and crop technology improvement (referring to the crop genetic improvement in enhancing crop community photosynthesis rates and N uptake capability. More details can be found in Lu *et al* (2018). Crop technology improvement was incorporated into the DLEM through two mechanisms: (1) an increase in the harvested amount of a crop based on a crop-specific time-series harvest index, and (2) improved productivity achieved by enhanced crops nitrogen uptake capacity. The key parameter values in regulating the model-estimated long-term trends of crop yield under changing climate and management practices were calibrated against the national crop yield records for each crop type from the USDA National Agricultural Statistics Service (NASS) crop databases (http://www.nass.usda.gov/index.asp). It is noteworthy that the impact of N fertilizer use examined in this study specifically refer to how fertilizer use rate change for each crop type has affected crop production and N loading across the MARB. The paired model simulations (comparing with and without fertilizer use change) maintain consistency in cropland area dynamics, with differences solely reflecting the influence of historical fertilizer use rate changes (Table S1 in Supplementary Material). In our simulation design, we disable management practice alterations in the paired experiments involving scenarios with and without LUCC. This allows us to isolate the effect of LUCC and exclude the influence of management practices on the newly cultivated or abandoned croplands. Moreover, as we incorporate annual crop type maps into our LUCC input, the estimation of LUCC impacts on crop production and N load encompasses alterations in N fixation caused by area change of legume crops. In determining the N fixation of crops such as soybean, alfalfa, and other nitrogen-fixing varieties, we simulate their annual dynamic productivity across various climate, soil, and management conditions and utilize crop-specific N-fixing parameters.

We depicted the trade-off using a concept of riverine nitrogen footprint (RNF) of grain crop production (i.e., the ratio between the change of N load and the change of total grain crop production associated with the aforementioned agricultural activities, where the unit is g N kg^{-1} grain). Using the RNF as an N footprint indicator, we aim to investigate how agricultural activities in the MARB have contributed to altering the water quality and grain crop production since 1970. It is noteworthy that these agricultural activities also play an important role in affecting the production of “other non-grain crops,” which are excluded from the RNF estimation in this study due to their relatively small share of cropland area and anthropogenic N input. Positive RNF values mean that crop production and riverine N load are changing “concurrently”, while negative values indicate “counter-current” changes between crop production and riverine N load. Under the circumstance of concurrent changes, water quality deterioration (i.e., positive N load change) occurs while crop production reaches a net gain (i.e., positive production change), and vice versa (i.e., negative changes in both riverine N load and crop production). We also quantify the economic revenues from major crop types driven by these activities, linking monetary gains with environmental costs. The results reflected by our study contribute to quantitative insights into food-biofuel-clean water conflicts in other agriculture-dominated river basins across the globe.

Methods

Model simulations

We used the DLEM model to quantify N leaching to local waters at each simulation grid as N yield (in a unit of g N m^{-2} day^{-1}) and accumulated N delivered to rivers and coastal areas at the river outlet grids as N load (in a unit of Tg N day^{-1}). Daily estimates were aggregated to monthly and annual total for analysis and comparison purposes. The N considered in DLEM includes dissolved organic N (DON), dissolved inorganic N (DIN: NO_3^-N and NH_4^+N), and particulate organic N (PON). We only quantified DIN loading in this study, which accounts for about 70% of the total N load to the Gulf of Mexico (Dominguez-Faus *et al* 2009). Annual DIN load at the river outlet to the Gulf was also validated by comparing DLEM estimates with the USGS LOADEST (i.e., the software is made available by the USGS for estimating constituent loads in streams and rivers based on a regression model given a time-series of streamflow, additional data variables, and constituent concentration, https://water.usgs.gov/software/loadest/) and Weighted Regression on Time Season and Discharge (WRTDS) simulation results from Stackpoole et al. (2021). For both LOADEST and WRDTS, we combined NO_2-NO_3 and NH_3 from the datasets to obtain annual DIN loading. N loading data from WRTDS and LOADEST are strongly correlated at an annual scale ($R^2 = 0.94$). The Nash–Sutcliffe model efficiency coefficient (NSE) and Percent bias (PBIAS) between DLEM estimates of annual DIN loading and the WRTDS data are 0.61 and 4.5%, respectively (Figure 1b). The calculations were performed using the "hydroGOF" R package (Zambrano-Bigiarini, 2017). We set up a series of counterfactual model simulation experiments by diminishing the change of driving factors one at a time, and compare them with the "best-estimate run" in which all the driving forces vary over time (Table S1). Their differences are used to quantify the changes in crop production and riverine DIN loading in response to changes in anthropogenic agricultural drivers in the MARB. Details of the DLEM model, model drivers, model calibration and validation, and simulation design can be found in Supplementary Material.

Examining the relationship between N balance and N loads

Based upon the existing data, we quantified annual anthropogenic N inputs, outputs, and N balance across the MARB during 1970-2017, and investigated the relationship between N balance and flow-normalized N loads during the same period. Anthropogenic N inputs considered in this study include atmospheric N deposition, synthetic N fertilizer use, manure N application, and N fixation by legume crop cultivation. The total N fixation amount is estimated based on the DLEM-simulated yield of legume crops (e.g., soybean, alfalfa, etc.) that has been validated against the USDA NASS database. The remaining N input data are derived from the time-series gridded database we developed in previous work for characterizing how human activities have altered N cycling and forcing the ecosystem modeling (e.g., Cao et al., 2018; Bian et al., 2021). N outputs in this study include crop-harvested N and gaseous N emissions. The annual amount of harvested N is obtained from the NuGIS (Fixen et al., 2018), which comprises 21 crops (i.e., alfalfa, apples, barley, dry beans, canola, corn for grain, corn for silage, cotton, other hay, oranges, peanuts, potatoes, rice, sorghum, soybeans, sugar beets, sugarcane, sunflower, sweet corn, tobacco, and wheat) during the period 1987-2017. We calculated the quantity of harvested N from 9 major crops grown in the MARB since 1970 using the methodologies detailed in Zhang et al., (2021), relying on county-level crop-specific yield and harvested acreage data as reported by NASS. Subsequently, we utilize the ratio of the harvested N by the 9 major crops to that of the 21 major crops reported by the NuGIS that is available during 1987-2017 to estimate the harvested N from the 21 major crops for the period preceding 1987. Nitrogen emissions in this study are determined by three pathways, namely direct N_2O emissions (1% of total anthropogenic N inputs), N_2 emissions resulting from denitrification (1.7 times the direct N_2O emission, Sabo et al., 2019), and indirect N_2O emissions due to N leaching (1.1% of the total regional N leaching estimated by DLEM in this study, Hergoualc'h et al., 2019). All the N input and output variables are processed within the MARB region. The flow-normalized TN (total N) loads in the MARB are obtained from the WRDTS model (Stackpoole et al. 2021) to reflect the anthropogenic trend in riverine N loads with random streamflow changes removed.

Definition of Partial Riverine Nitrogen Footprint of Grain Crop Production

Based on the above experiments, we quantified how agricultural activities (including land use changes and extensive management practices) have simultaneously altered riverine N loading and production of eight major grain crops in the MARB since 1970. We examined the relative contributions of key agricultural activities to changes in N loading and grain crop production across the Basin. Due to the lack of detailed crop type, distribution, agronomic and management information, the production of non-grain and other crops were excluded in this analysis, although their production contributed to N yield and loading to the Gulf as well. Therefore, the RNF could partially reflect the water quality cost of per unit grain production gain under historical agricultural resource (e.g., land, nutrient) allocation since 1970. We also summarized the sub-basin riverine N footprint on a decadal scale for comparison purposes. To identify the N reduction potential of each agricultural activity, we defined the partial riverine N footprint as:

$$N_{footprint,i} = \Delta N_{loading,i} / \sum_i \Delta Production_i \quad (1)$$

where $N_{footprint,i}$ is the riverine N footprint driven by land use or management practice i, $\Delta N_{loading,i}$ is N loading change driven by each activity, and $\sum_i \Delta Production_i$ is the net change of crop production driven by all four activities considered in this study. The concurrent changes in riverine N footprint indicate that either water quality is sacrificed while pursuing a crop production gain or water quality is improved at the cost of losing crop production. Under the circumstance of counter-current changes, however, water quality improvement (i.e., negative N load change) can be accompanied by crop production gain (i.e., positive crop production change), such as the impacts of crop technology improvement. The other possible scenario under this circumstance is that water quality deterioration (i.e., positive N load change) is accompanied by crop production loss (i.e., negative crop production change), which likely occurs under extreme climate conditions. In our analyses, a net crop production gain is seen in most cases. However, changes in crop production during the 2000s in the Lower Mississippi sub-basin (LM) is close to zero, which results in an extraordinarily large value of N footprint. Therefore, to avoid providing a misleading interpretation of riverine N footprint, we excluded the calculations for the LM in the 2000s. Furthermore, our analysis did not consider the drainage area changes over the past decades in the sub-basin within the MARB.

Estimation of crop-specific revenue changes

We calculated the revenue change of the eight major grain crops within the MARB, driven by the four agricultural activities. First, we obtained the state-level crop-specific price data from the USDA NASS survey database. To make sure the price in one year is comparable to the price in another year, we deflated the price data to remove the inflation factor using the Gross Domestic Product Implicit Price Deflator (https://fred.stlouisfed.org/series/GDPDEF) with 2010 treated as the base year. We then multiplied the model-estimated annual crop production change, which was driven by the four agricultural activities in the DLEM model, by the deflated price data for each crop in each state. The product was treated as the annual crop-specific revenue change given the assumption that all the crop productions were traded in the market. Then, we summarized the sub-basin total revenue change on a decadal scale for comparison purposes.

Results

Historical patterns of agricultural activities in the MARB

In the MARB, the total cropland area averaged 90.3 ± 3.7 Mha (mean ± SD, SD is quantified to reflect year-to-year variations) with little inter-annual variations and a relatively stable pattern (trend of 0.99 Mha decade^{-1}) during 1970-2021. Among them, the eight major grain crops accounted for 73% ± 5% of the total cropland areas in the region, exhibiting a significant increasing trend of 3.6 Mha decade^{-1} (p<0.05) over the past five decades. In addition, the total synthetic N fertilizer input averaged 6.2 ± 1.2 Tg N yr^{-1}, a N export from the MARB mong which 81% ± 5% were applied for the production of these eight major crops. The amount of synthetic N fertilizer received by all the crops and these eight major crops demonstrates a similar increasing trend (0.73 vs 0.61 Tg N decade^{-1}, p<0.05). The total area of the eight major crops increased by 16 Mha (27% of the 1970s average) over the past five decades. Five out of the eight major crop types presented an increase in their planting areas (Figure 2a), with corn and soybean each increasing by ~10 Mha, followed by spring wheat (0.58 Mha), rice (0.35 Mha), and durum wheat (0.16 Mha). In contrast, the planting areas of sorghum and barley decreased by 3 Mha and 1.2 Mha, respectively, followed by winter wheat (–0.5 Mha). Overall, the land and nutrient resources invested to these major crop types grew substantially during the past five decades although the total cropland area in this region remained stable.

We found that among the six sub-basins, the expanded croplands are primarily found in the Missouri (MO, 8.5 Mha) and Upper-mid Mississippi (UM, 5 Mha) river basins. Specifically, the corn area in the MO and the UM increased by 6.2 Mha and 3.8 Mha, respectively (Figure 2a). N fertilizer input increased by 1.0 Tg and 0.7 Tg in the MO and the UM, respectively, which was mainly driven by the corn area expansion (Figure 2b). Soybean area increased by 3.7 Mha and 2 Mha in these two sub-basins, respectively, which is about half of the corn acreage increases. In the Ohio river basin (OH), the total agricultural land area increased by 2 Mha, among which soybean area increased by 1.6 Mha (80% of net cropland area change) and corn area only increased by ~0.7

Mha (35% of net change). The increasing soybean used in rotation with corn only led to an increase of 0.2 Tg N fertilizer use, which potentially reduced the N loss per unit crop production gain in this sub-basin. The areas of the major crop types in the other sub-basins remained relatively stable over the past five decades (Figure 2a).

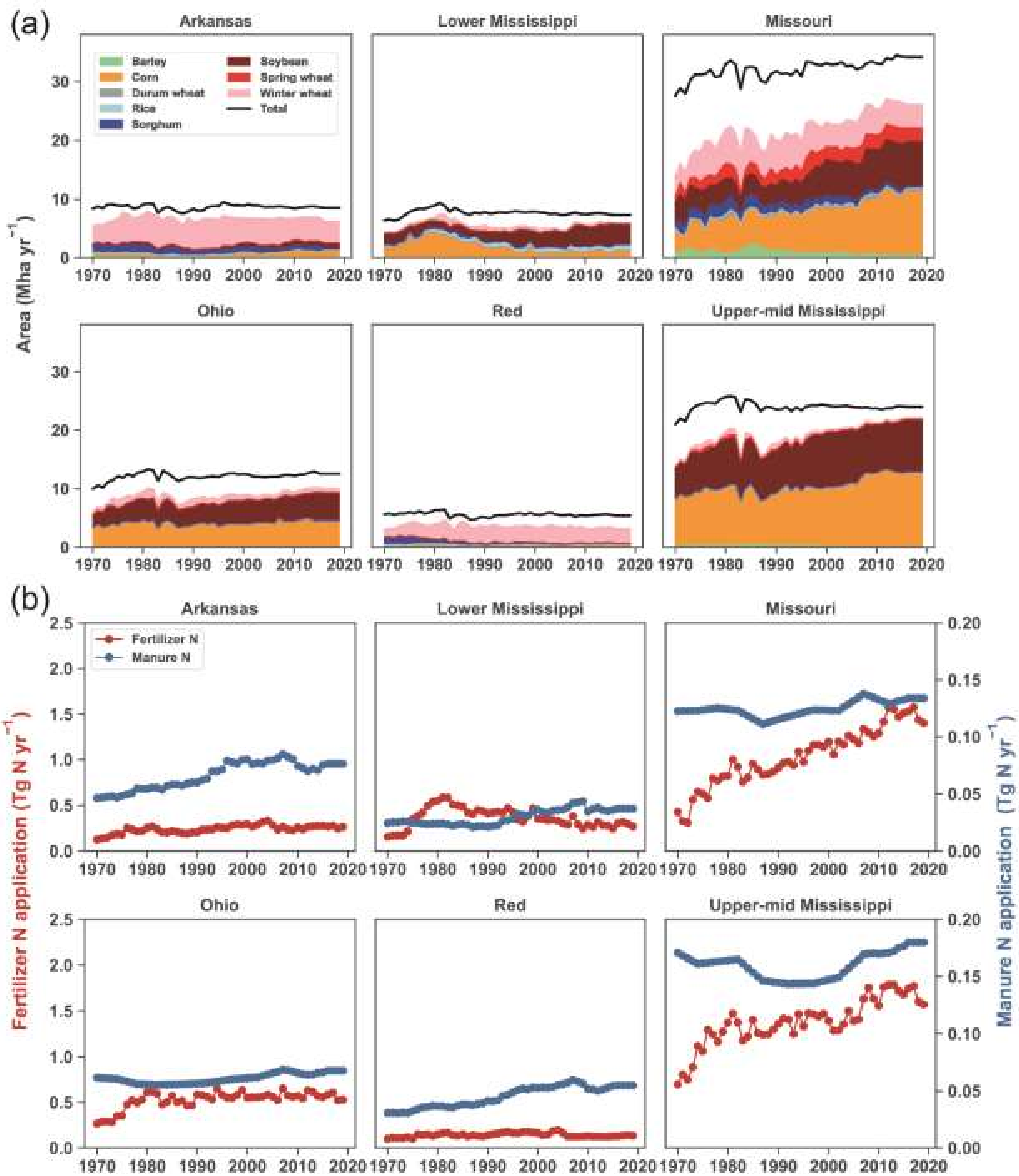

Figure 2. Annual areas of eight major grain crops and total cropland (a), and total synthetic N fertilizer and manure N application (b) from 1970 to 2017 in the sub-basins of the MARB.

Anthropogenic N balance on land and riverine N load

To further investigate how terrestrial anthropogenic N balance affects N loads from the MARB, we compiled the time-series data of N inputs, outputs, N balance from model input database and survey-based crop N budget data (Fixen et al., 2018, Zhang et al., 2021), as well as the flow-normalized N load derived from Stackpoole et al (2021). Our data analysis (Figure 3a) indicates that the anthropogenic N inputs received by the MARB land surface, including N fixation by legume crop cultivation, atmospheric N deposition, synthetic N fertilizer use, and manure N application in croplands, have increased from 9.5 Tg N yr^{-1} (1 Tg = 10^{12} g) in the 1970s to 14.4 Tg N yr^{-1} in the 2010s, with an increasing trend of 0.12 Tg N yr^{-2} (R^2 = 0.89). On the other hand, the total N outputs, including crop harvested N and N emissions, have increased from 8.2 Tg N yr^{-1} to 13. 1 Tg N yr^{-1} during the same period, exhibiting a similar increasing trend but larger inter-annual variations (0.13 Tg N yr^{-2}, R^2 = 0.79). Despite large year-by-year fluctuations in the annual N balance, the smoothed N balance shows a clear trend with an initial increase since 1970, followed by a continued decline after the mid-1980s. We find this trend corresponds well with the flow-normalized TN load (Figure 3b), an indicator that removes the random effects of streamflow change, and therefore, could better represent the anthropogenic trend of the land N pollution to water bodies. However, there is also evidence of decoupling between them: the terrestrial N balance reaches its peak a few years later than the flow-normalized TN load (1988 vs 1983); furthermore, while the N balance continues to decline thereafter, the flow-normalized TN load remains stable and has even increased slightly in the recent decade. This implies that the rise in land N balance consistently contributed to deteriorating water pollution in the Gulf prior to the mid-1980s, and the subsequent decrease in N balance correlates with the enhancement of water quality between the mid-1980s to mid-1990s. Nevertheless, beyond that time frame, the decline in N balance, driven by augmented crop uptake, has had a diminished impact on further improving water quality. This finding is consistent with the relationship between N balance and flow-normalized TN loads reported by Stackpoole et al (2021).

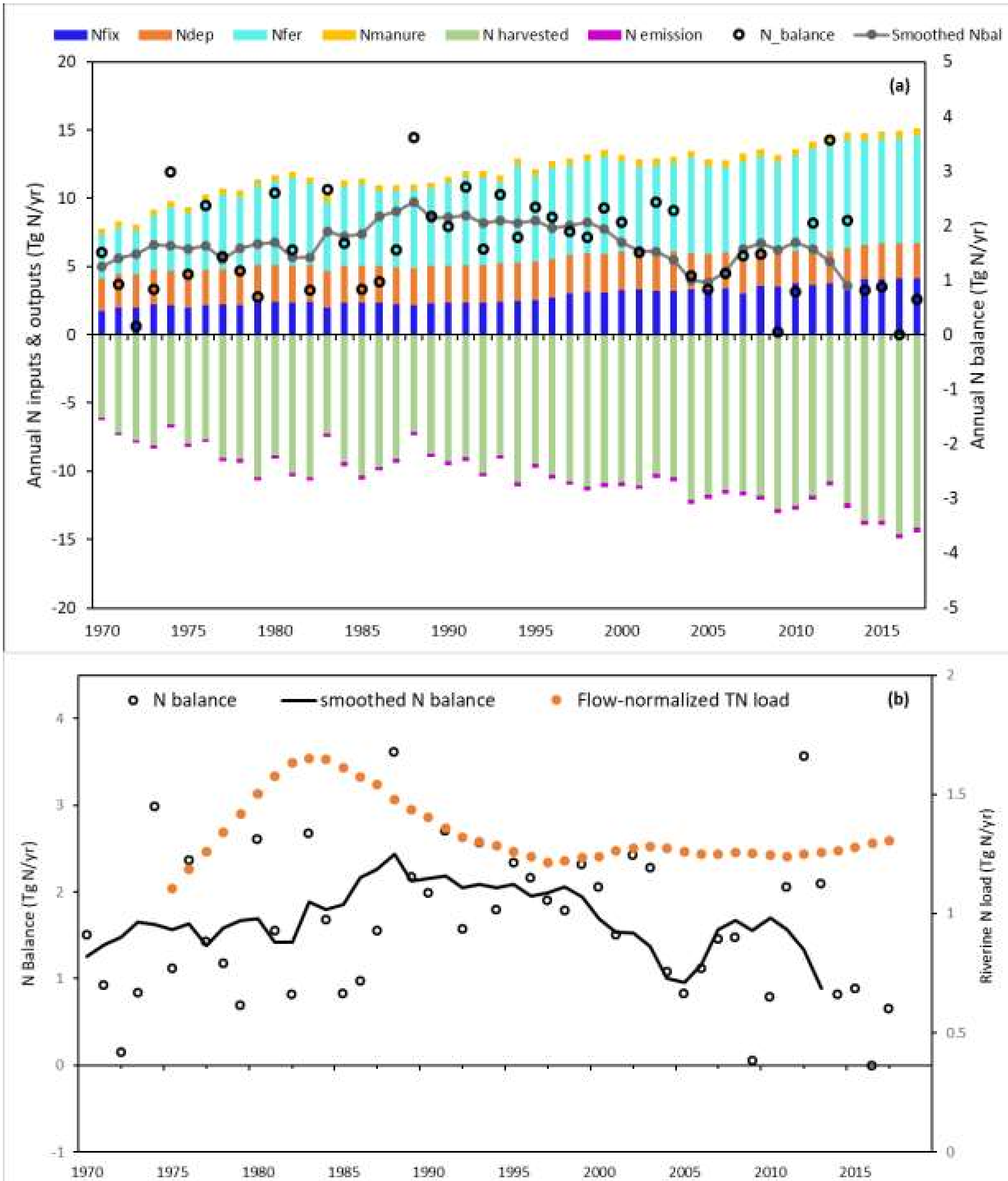

Figure 3 Annual time-series N inputs, outputs, and N balance (a) and comparison between N balance and flow-normalized N load (b)in the MARB during 1970-2017. Nfix, Ndep, and Nfer in Fig (a) stand for N fixation, N deposition, and synthetic N fertilizer use, respectively. N manure

refers to manure N application in the croplands. Smoothed Nbal refers to smoothed N balance derived from 6-year moving average.

Impacts of agricultural activities on N loading, grain crop production, and RNF

Our model attribution indicates that the major agricultural activities considered in this study together led to a net crop production increase, ranging from 43 MMT yr^{-1} in the 1970s to 107 MMT yr^{-1} in the 2010s across the Basin (Figure 4a). The impacts of LUCC on crop production changes varied a lot during the study period, with its contribution ranging from 12% in the 1990s to 86% in the 1970s. However, apart from 1970s, crop technology improvement was identified as a primary contributor, leading to 29% to 56% of the net change in crop production. Meanwhile, the growing usage of N fertilizer contributed to 18% to 36% of the increase in this region but manure N application played a trivial role.

Similar to the trend in flow-normalized N load (eliminating random streamflow effects, Figure 3b), the changes in model-estimated riverine N load due to these agricultural activities (human impacts) also follow a pattern of initial increase followed by leveling off, spanning the last fifty years. (Figure 4b). We find that agricultural activities together drove a net increase of 0.07 Tg N yr^{-1} in N load in the 2010s. Among these activities, N fertilizer use plays a dominant role in leading to N load increase ranging from 0.02 Tg N yr^{-1} in the 1970s to 0.16 Tg N yr^{-1} in the 2010s, while LUCC grows to be an important contributor next to fertilizer input since the 1980s, with its contribution growing by over threefold from 0.03 Tg N yr^{-1} in the 1980s to 0.1 Tg N yr^{-1} in the 2010s. Crop technology improvement, however, decreased N load, and its "mitigation" effects on N load increased by over tenfold from −0.018 Tg N yr^{-1} in the 1970s to −0.19 Tg N yr^{-1} in the 2010s. Our simulation indicates that manure N application in the croplands had a negligible impact on N loading changes in the Basin, compared to other driving factors. This is possibly caused by the fact that we only consider the part of manure N applied in the croplands, rather than the total manure N produced by the livestock sector. Globally, in 2017, the estimated ratio of manure N application to production stands at 0.18 (Bian et al., 2021).

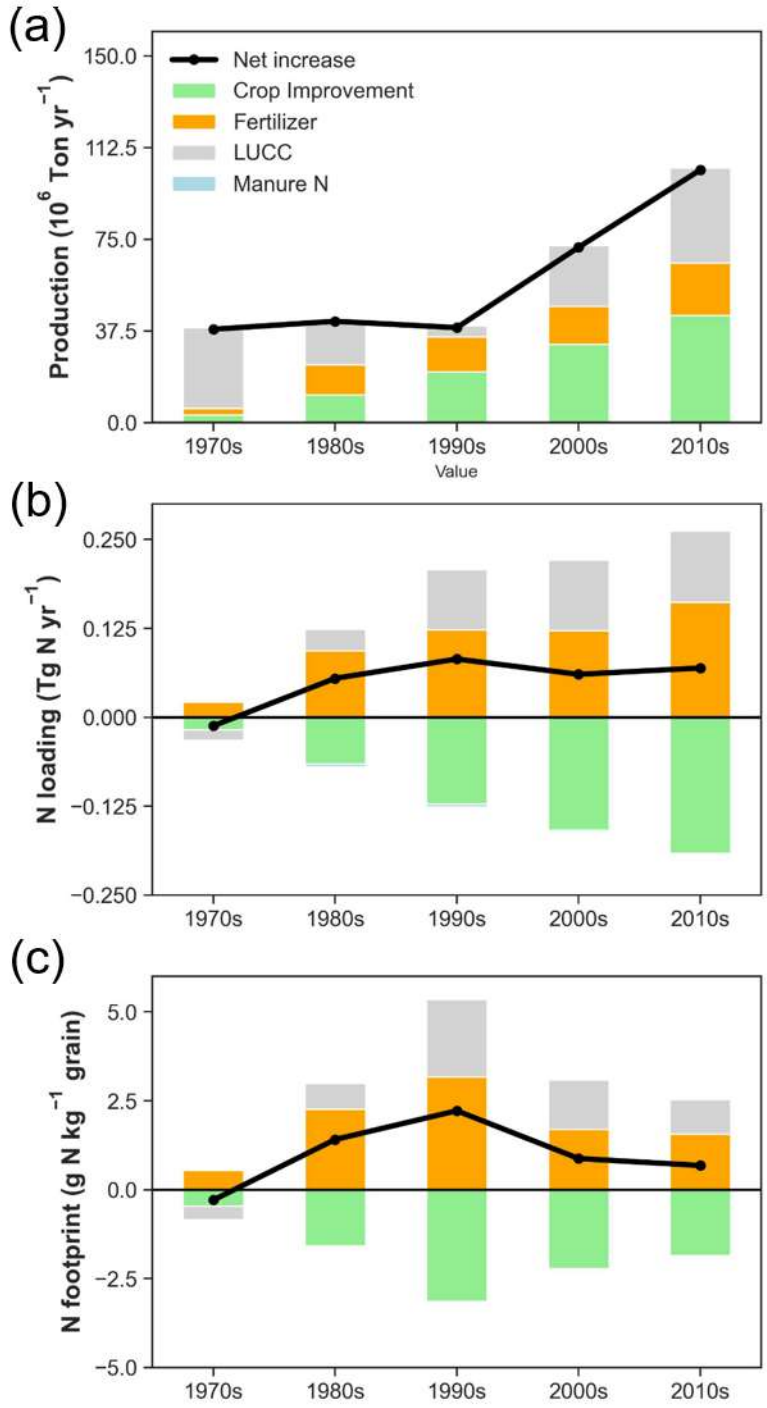

Figure 4. Model-estimated factorial contributions to changes in crop production (a), N load (b), and riverine N footprint (c) from the 1970s to the 2010s. Major drivers considered here include crop technology improvement (Crop Improvement), synthetic N fertilizer use (Fertilizer), land use

and land cover change (LUCC), and manure N application (Manure N). The black dot curve is the net change driven by all these four agricultural activities.

The model estimation shows that the RNF (the ratio of $\Delta N_{loading}/\Delta$Production; see Methods for details) of grain crop production in the MARB has increased from −0.28 g N kg^{-1} grain in the 1970s to its peak, 1.95 g N kg^{-1} grain, in the 1990s, followed by a two-decade decline to 0.65 g N kg^{-1} grain in the 2010s (Figure 4c). This implies that the increases in Basin-wide crop production were not necessarily associated with a proportionate degradation in water quality during recent two decades. On the other hand, the four agricultural activities show varied impacts on the RNF. The N fertilizer-induced RNF has increased six-fold, from 0.49 g N kg^{-1} grain in the 1970s to 2.94 g N kg^{-1} grain in the 1990s (Figure 4a). It has declined and then remained stable at approximately 1.5 g N kg^{-1} grain in the recent two decades. This could be linked to the observation that the rate of increase in synthetic N fertilizer use for each crop has decelerated in the last twenty years (Cao et al., 2018), and the rise in N loading due to fertilizers is less than the overall increase in crop production since the 1990s (Figure 4). It is noteworthy that the LUCC impacts on the RNF has reversed from -0.33 g N kg^{-1} grain in the 1970s to 0.67 g N kg^{-1} grain in the 1980s. Likewise, the LUCC-induced RNF also peaked in the 1990s (2.01 g N kg^{-1} grain), and then declined to 0.93 g N kg^{-1} grain in the 2010s. The impacts of crop technology improvement on reducing RNF demonstrates a similar "V-shape" trend with its peak in the 1990s (-2.89 g N kg^{-1} grain), followed by a decline to -1.78 g N kg^{-1} grain in the 2010s. This indicates its efficiency on improving water quality diminished during the most recent decades. Together, these shifts imply a disconnection between the rising rates of crop production and N loading caused by the four agricultural activities since the 1990s.

Sub-basin-wide patterns of agricultural activities' impact on RNF

We find that the RNF in the Upper-mid Mississippi (UM) river basin has increased from a negative value (−1.3 g N kg^{-1} grain, represented by the black point in Figure 5) in the 1970s to 2.6 g N kg^{-1} grain in the 1990s, and then decreased to 0.54-0.66 g N kg^{-1} grain in the past two decades (Figure 5). In this sub-basin, N fertilizer input has consistently increased the N footprint, with the largest contribution found in the 1980s (5.2 g N kg^{-1} grain), followed by a substantial decline thereafter. Crop technology improvement has constantly reduced the N footprint, with its peak impact in the 1980s. In contrast, LUCC impacts on the RNF shifted from a negative value in the 1970s to positive values thereafter, implying a complicated land use changes at the sub-basin scale over time. However, starting from the 1990s, LUCC and N fertilizer inputs have played a comparable role in augmenting the RNF, and their impacts have decreased in tandem. Crop production increase in the UM river basin accounts for approximately one-third to half of the MARB total in the past half century (Figure S8). The decreasing riverine N footprint in this region since the 1990s indicates that enhancements in crop production have resulted in a diminishing impact on water pollution.

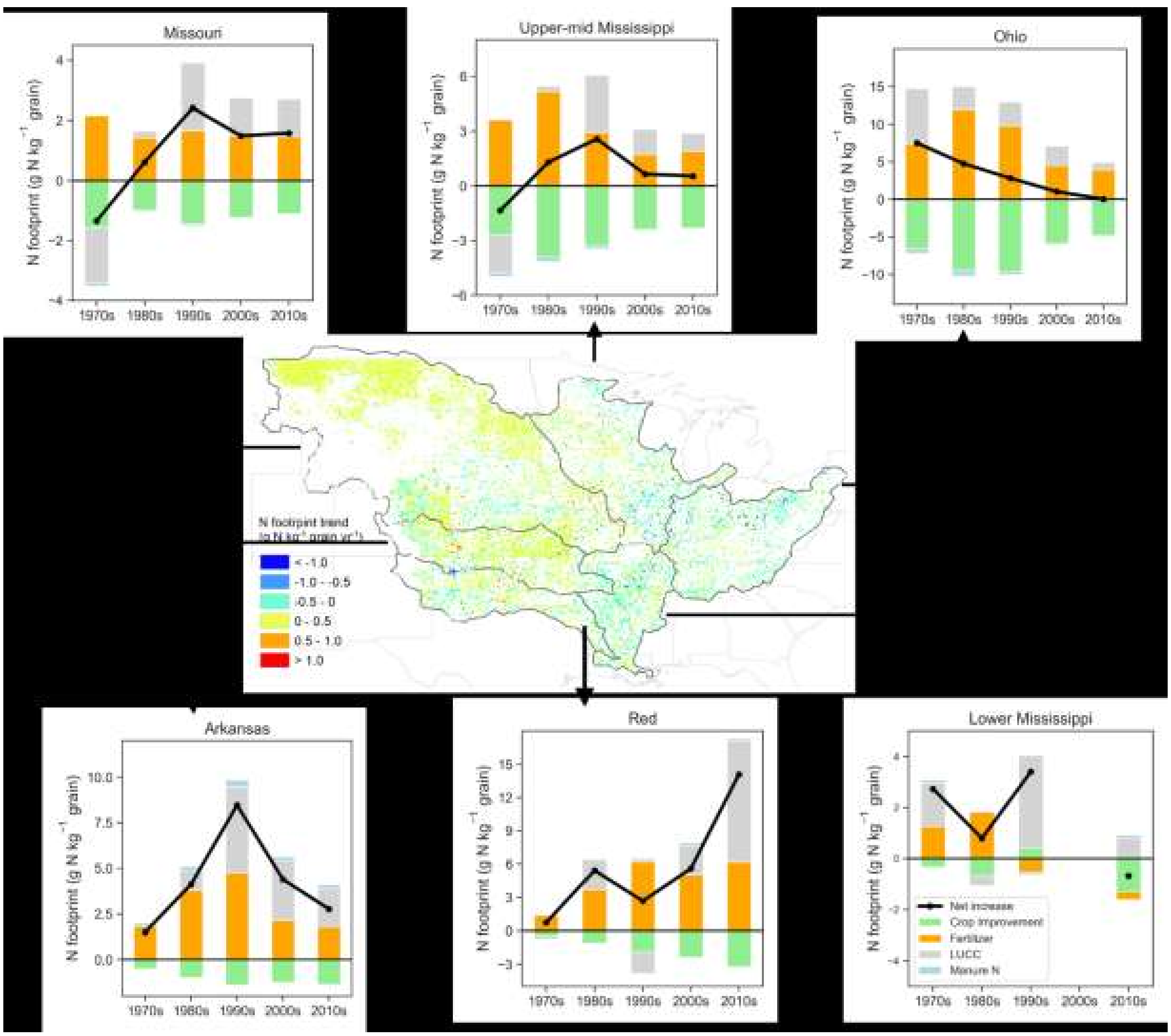

Figure 5. Modeled factorial contributions to changes in riverine N footprint of crop production in the six sub-basins from the 1970s to the 2010s. The centered map indicates the significant trends of N footprint, calculated as the ratio of the change in N yield (i.e., N leaching determined by both surface and sub-surface runoff) to the change of crop yield, across the MARB. (Note: due to the

close-to-zero values in crop production changes in the Lower Mississippi river basin (LM) in the 2000s 0.9 MMT yr^{-1}), the N footprint would be extraordinarily large, which is less informative in comparison with the values in other sub-basins. Therefore, the N footprint in these two decades in the LM was not plotted in the figure. We also ignored the N footprint in the grids where the absolute value of crop yield change is less than 0.01 kg m^{-2}.)

The Missouri river basin (MO) demonstrated a similar pattern of RNF as in the UM, increasing from a negative RNF (−1.3 g N kg^{-1} grain) in the 1970s to its peak (2.4 g N kg^{-1} grain) in the 1990s, followed with a decline to 1.5-1.6 g N kg^{-1} grain in the last two decades. Similar to the UM, in the MO river basin since the 1990s, LUCC and N fertilizer inputs have played a comparable role in enhancing the N footprint. Nevertheless, the reduction in N footprint due to crop technology improvement has remained relatively consistent over time in this region. Crop production increase in the MO accounts for 23% to 47% of total rise in the MARB during the study period (Figure S8). The declining N footprint, in this context, indicates a loose connection between grain production gain and water pollution. The RNF in the Arkansas (AR) river basin has demonstrated a similar inverted V-shaped pattern, increasing first and then declining. However, in this region, the RNF dynamics has been predominantly affected by LUCC and fertilizer inputs, while the effects of crop technology improvements have been much smaller. Despite the incomplete trajectory of RNF estimation in the Lower Mississippi River basin, we are still able to identify a decreasing trend in RNF following the initial rise. The similar RNF patterns in these sub-basins suggest that although boosting crop production, the expansion of cropland and increased N fertilizer use have escalated the riverine N footprint over the first three decades. However, this trend has waned over the recent two decades, partially due to the fact that the response of N load to these agricultural activities has become less rapid than the response of crop production.

An encouraging pattern is found in the Ohio river basin (OH, Figure 5). Although the RNF started at a high level (i.e., on average 7.5 g N kg^{-1} grain in the 1970s), it has decreased to a near-zero value (0.04 g N kg^{-1} grain) in the 2010s. This notable decrease was primarily propelled by the reduced pollution sources arising from both LUCC and N fertilizer. Collectively, the impacts of LUCC and N fertilizer on RNF declined from approximately 15 g N kg^{-1} grain in the 1970s to around 5 g N kg^{-1} grain in the 2010s. We find that LUCC and N fertilizer have increased N load by only 0.09 Tg N yr^{-1} and 0.06 Tg N yr^{-1} over decades, while they have raised crop production by 11.2 MMT yr^{-1} and 8.0 MMT yr^{-1}, respectively (Figure S8). Meanwhile, crop technology improvements have exerted significant impacts on reducing the RNF, ranging from -4.8 g N kg^{-1} grain in the 2010s to -9.6 g N kg^{-1} grain in the 1990s. Even though the rise in crop production within the Ohio river basin comprises only 10% to 17% of the overall increase in the MARB, the steady reduction in its RNF suggests a promising path toward sustainable agriculture and a diminishing environmental footprint.

Different from other sub-basins, the Red river basin demonstrates a growing RNF, with its peak of 14.1 g N kg^{-1} grain in the 2010s. Even though the Red River basin contributes a smaller proportion to the overall increase in crop production (<5% of the MARB total in the recent three decades), the increasing RNF serves as a warning that even modest crop production gains might come at the expense of deteriorating water quality in certain regions. Although the changing trend in the RNF varied substantially across the MARB over the 48 years, it aligns closely with the decadal patterns (centered map in Figure 5).

Discussions

Interpretation of modeling limitations and uncertainties

In this study, the model-estimated concurrent or counter-current change in crop production and N loads influenced by agricultural activities and the consequent RNF is different from the latent process that was investigated and reported to be important in reducing TN load since the mid-1980s (Stackpoole et al., 2021). Their study argues that increased N retention in watershed, as well as slowed increase rate of N balance, have reduced TN loads in recent decades. This is consistent with what we find through crop-specific N balance data analysis and counterfactual modeling assessment (e.g., the declining N balance since 1988 in Figure 3, and decreasing riverine N footprint since the 1990s in Figure 4). From the perspective of the crop N budget that was entirely estimated by the USDA census data, our earlier research (Zhang et al., 2021) also revealed that the crop N surplus at the national level (referring to the total N input not utilized by field crops) exhibited a relatively consistent pattern from the 1980s to the 2000s within US cropping systems. Subsequently, a reduction occurred during the 2010s, bringing the surplus to a level akin to that of the early 1970s. Despite concentrating solely on crop nitrogen budgets, Zhang et al. (2021) mirrors a comparable trend to our study, underscoring the stabilization or reduction in N pollution to the environment within the principal cropping systems of the US over the recent decades.

Mounting evidence has indicated that effective land management and sustainable agricultural practices in upstream watersheds could significantly improve downstream water quality (Vitousek et al 1997, Blann et al 2009, Yang et al 2016). Upstream conservation programs in the MARB have targeted reductions in N moving to surface waters through adopting best management practices (e.g., cover crops and side-dressing N application) and increasing crop nitrogen use efficiency (NUE). But our study aims to investigate how crop production and N loads simultaneously respond to a given agricultural activity change, in which the impacts of best management practices (BMPs) such as prairie strips, planting of cover crops, are not examined. For example, we include historical tillage records and tile drainage as model inputs to drive the DLEM for historical simulations (details can be found in the appendix and Lu et al., 2022). However, the impacts of these practices are not teased out and quantified in this study. In addition, the model performance in estimating water discharge and DIN loads vary among sub-basins (Figure S6-S7). This variability can be attributed in part to the following factors: (1) Limited availability of input data: The accuracy of our model relies on the quality of input data used for driving it, such as crop area and distribution maps, N fertilizer utilization, and manure N application maps. However, these data sources are constrained by the extent of state-level crop-specific surveys. Consequently, there might be discrepancies between the data and the actual land and nutrient management practices within the boundaries of individual sub-basins. (2) Representation of flood control and dam operations: The Dynamic Land Ecosystem Model (DLEM) employed in our study does not adequately capture changes in flood control and dam operations, potentially resulting in an overestimation of N loading in sub-basins characterized by dense reservoir networks during flooding years (Lu et al., 2020). It's important to highlight that our analysis concentrates exclusively on anthropogenic influences. Notably, we have excluded the effects of climate on interannual fluctuations in N loads by computing the disparity between two simulation experiments.

Research has demonstrated that climate variability and extreme events exert a substantial influence on historical riverine N loading originating from the MARB. (Battaglin et al., 2010, Smits et al., 2019, Lu et al., 2020). Future climate change and more frequent extreme climatic events are projected to increase N export from the basin, even with hypothetically unchanged N fertilizer inputs and cropland expansion in the scenario testing (Zhang et al 2022). While this study does not explicitly quantify the effects of historical climate variations and changes in atmospheric CO_2 concentrations, their interplay with agricultural activities has been considered. These factors influence model-based estimations of crop growth and nitrogen loading by modulating the availability of resources such as radiation, light, water, and CO_2, as well as influencing biogeochemical processes. We acknowledge that models used for estimation and attribution have various sensitivities to environmental changes such as rising temperature, increasing atmospheric CO_2 concentration, N fertilization, irrigation. Modeling approach has limitations in accurately quantifying the impacts of individual factors due to limited knowledge on the complex process, interactions, and confounding factors (Ruehr et al., 2023). In this case, the DLEM estimates of crop production and N loads have been calibrated and validated against long-term monitoring, measurement, and survey data across the MARB and the country (Fig 1, Fig S1-S7). The model-estimated responses to LUCC, agricultural N inputs, and crop technology improvement have been cross-validated with other studies in our previous work (Yu et al., 2019; Lu et al., 2018, 2020, 2021) and averaged at the decadal level in this study to reduce the influence of inter-annual variations over a half-century study period. To constrain modeling results, in terms of factorial contribution assessment, more future studies are needed to provide long-term measurement data under paired experiments (e.g., control and treatment) across various climate and soil conditions to inform and evaluate modeling work.

Implications for future nutrient management across the MARB and beyond

Agricultural practices associated with corn cultivation are the major cause of N load in the U.S. agricultural systems (Broussard and Turner 2009). As a result, the expansion of corn would significantly increase the total N load in the river systems (Ni *et al* 2021). At recommended N rates, corn-soybean rotation systems would lead to 14 to 36% lower NO_3–N concentrations than in continuous corn systems (Helmers *et al* 2012). However, soybean cultivation may lead to a higher phosphorus (P) yield than corn cultivation (Ni *et al* 2021). Therefore, future research efforts are needed to evaluate the trade-offs between N loads and P loads in rotation systems.

The results of this study provide evidence that LUCC, crop choices, and follow-up nutrient management play a key role in determining if a region improves water quality while maintaining or increasing crop production. In the MO and the UM, although cropland expansions, combined with the widespread use of fertilizer, increased crop production, they came at the cost of water quality degradation (Ramankutty *et al* 2018). Nevertheless, in the OH, increased crop production was accompanied by a reducing riverine N load during the study period (Figure S8). These divergences were due to grain crop production increases caused by different crops and different drivers among sub-basins. For example, corn production change in the MO, rising from 0.1 MMT yr^{-1} in the 1970s to 27 MMT yr^{-1} in the 2010s, accounted for 80% of the total crop production increase over the study period (Figure 6). Corn production increase in the UM went up to 25 MMT yr^{-1} in the 2010s, accounting for 83% of the total change. However, soybean showed secondary increases over decades in these two sub-basins, with a net increase of 11 MMT yr^{-1} (28%) and 8 MMT yr^{-1} (23%) in the MO and the UM in the 2010s, respectively. These results indicated that corn production consistently dominated and had a large share of the grain crop production increase in the Midwest regions, which were associated with water quality deterioration in the U.S. Midwestern basins in the first three decades. However, primarily driven by crop technology improvement, the rise in crop production during the recent two decades was not accompanied by a proportionate N loading increase in these two basins (Figure S8).

The changing trends we find in other sub-basins indicate that a reduction in RNF is achievable through properly managing agricultural lands where high N fertilizer-consuming crops (e.g., corn) used to dominate the crop shares. For instance, grain-crop production increase in the OH only rose from 3 MMT yr^{-1} in the 1970s to 18 MMT yr^{-1} in the 2010s (Figure 5). Among crops, while corn's contribution to the overall production change increased from 54% in the 1970s to 60% in the 2010s, the proportion of soybean also rose significantly from 29% to 43%, with negligible or negative contribution from the remaining crops. The significant rise in soybean production share also elucidates the consistent trend of synthetic fertilizer inputs in the OH since the 1980s, despite the ongoing increase in total crop production (Figure 2, Figure S8). Based on the NASS database, our previous study found that the total harvested area of corn in Ohio, Indiana, Tennessee, and West Virginia, which consist of the majority of the Ohio river basin, increased by only 1% in the 2010s, compared with the 1970s (Lu *et al* 2019). Such changes in crop choice and N input align well with the detected RNF decrease in the 2000s and the 2010s in this sub-basin (Figure 5). Furthermore, we noticed that the cropping system in the LM experienced an alteration from corn-dominated to soybean- and rice-dominated over decades (Figure 6). These changes in crop production indicate that, although corn-planting-induced N loss was likely replaced by rice-planting due to their high N demands, the elevated share of soybean production still reduced N input and led to a “near-neutral” N loading change (Figure S8).

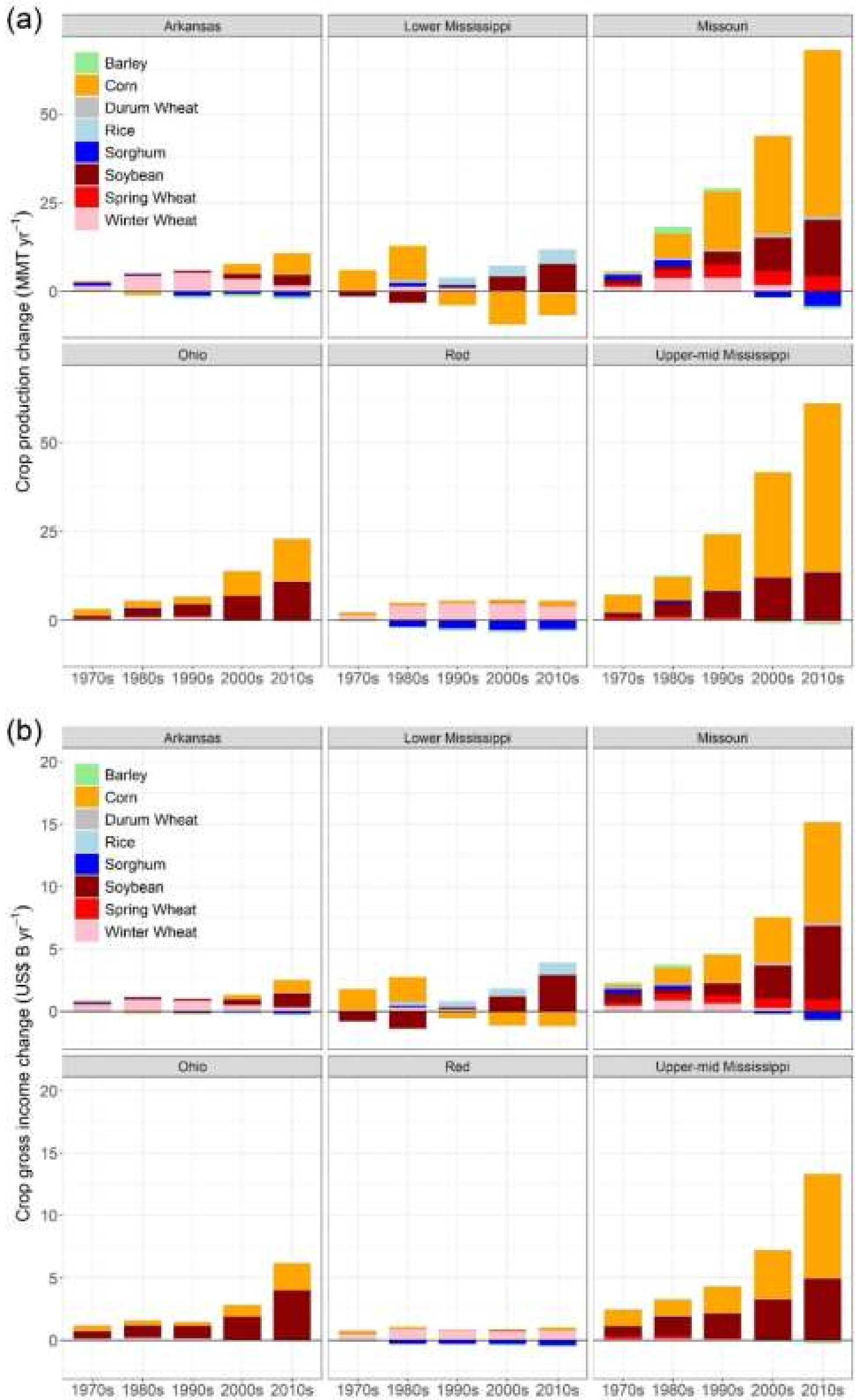

Figure 6. Decadal average production change (a) and revenue change (b) of major crops resulting from the four agricultural activities in the sub-basins of the MARB.

Economically, corn contributed 53% (e.g., US$ 5.4 Billion (B) yr^{-1}) and 64% (US$ 6 B yr^{-1}) of the total revenue changes of the eight major grain crops in the MO and the UM in the 2010s, respectively (Figure 6b), which are lower than its shares of grain production. In contrast, soybean-induced revenue change was about 44% (US$ 4.4 B yr^{-1}) and 38% (US$ 3.5 B yr^{-1}) in the MO in the UM, respectively, exceeding its share in grain production in the same decade. In the OH, soybean-planting induced revenue change in the 2010s (i.e., US$3.1 B yr^{-1}, 63%) has already exceeded the contributions of corn (US$ 2.0 B yr^{-1}, 39%), suggesting that soybean has emerged as the primary crop for increasing economic income. Soybean's price is higher than corn, although its yield per unit area is lower. Also, soybean would require less cost (e.g., the need for a small amount of fertilizer). Currently, boosted by biofuel prices, the share of continuous corn growing is increasing over time in the U.S. Midwest (Wang and Ortiz-Bobea 2019). Therefore, soybean has the potential to serve as a key cash crop in the future to maintain the trade-off between water pollution and farmers' economic profits. Beyond managing crop choices, we may target adopting other restoration practices, such as restoring less productive agricultural lands to natural lands (Wu *et al* 2013, Cheng *et al* 2020), using slow-releasing fertilizers in fields, replacing monoculture with polyculture (Crews *et al* 2018), and postponing fertilizer input to meet plant nutrient demands (Lu *et al* 2020).

Outlook

Our results indicate that the temporal changes in crop production and N loads in response to the major agricultural activities became decoupled after 2000. This transition led to a reversal of the trend in RNF, shifting from an increasing to a decreasing pattern (Figure 4c). The findings suggest that cropland expansion, increasing share of corn and soybean, and elevated anthropogenic N input before 2000 have consistently sacrificed water quality for higher crop production to meet food and energy demand, which is supported by earlier work (Foley *et al* 2005, Daniel *et al* 2010, Secchi *et al* 2011, Lark *et al* 2022). However, after 2000, crop technology improvement is more effective in reducing N load as well as increasing crop production than LUCC and N fertilizer input, which on average led to a reducing RNF over the past two decades (Figure 4c). Our result implies that N loading has changed at a slower pace under the agricultural resource use and management efforts in pursuing the same unit grain production in the recent two decades, which is an encouraging signal for water quality improvement.

Local stakeholders should raise awareness of environmental degradation while managing agricultural land to achieve long-term environmental sustainability (Handmaker *et al* 2021). Farmers' choices over what to plant will be driven by markets and policies. However, influenced by the frequent extreme weather conditions in the largest crop-producing countries (e.g., Brazil: the 1st soybean-producing and 3rd largest corn-producing country), the market effects of trade wars, the lingering supply issues induced by coronavirus pandemic, and food crisis due to the Russia-Ukraine (the 6th largest corn-producing and the 8th largest soybean producing country) conflict, crop prices will likely become more unpredictable in the following years. Therefore, it is difficult to predict how the choices of crops will change regionally and globally. These uncertainties underscore the significance of comprehending the intricate trade-offs between food and biofuel production and environmental quality, given agricultural management and economic event-induced market effects. Our conclusions drawn here have important ramifications regarding land and nutrient resource management for achieving long-standing policy goals for agricultural and environmental sustainability.

Author Contributions

CL conceived the idea; JZ conducted model simulations; CL, JZ, BY and IC analyzed the model outputs; CL and JZ wrote the original draft; all authors contributed to interpreting the results, reviewing and editing the manuscript.

Data Availability Statement

The data that supports this study, including anthropogenic nitrogen balance and model-estimated riverine nitrogen footprint in the Mississippi-Atchafalaya River Basin during 1970-2019, can be found in https://doi.org/10.6073/pasta/62f779fab3be6e90f008d163455f8559

Acknowledgments

This work is supported by the Iowa Nutrient Research Center, the Postdoc seed grant award of Iowa State University, NSF grant (1903722), and NSF CAREER (1945036). We extend our gratitude to Dr. Robert Sabo at the United States Environmental Protection Agency and two anonymous reviewers for their constructive comments, which significantly improve the quality of this manuscript.

Conflict of interest

The authors declare no competing interests.

References

Altieri A H, Harrison S B, Seemann J, Collin R, Diaz R J and Knowlton N 2017 Tropical dead zones and mass mortalities on coral reefs *Proceedings of the National Academy of Sciences* **114** 3660–5

Blann K L, Anderson J L, Sands G R and Vondracek B 2009 Effects of agricultural drainage on aquatic ecosystems: a review *Critical reviews in environmental science and technology* **39** 909–1001

Boesch D F, Boynton W R, Crowder L B, Diaz R J, Howarth R W, Mee L D, Nixon S W, Rabalais N N, Rosenberg R and Sanders J G 2009 Nutrient enrichment drives Gulf of Mexico hypoxia *Eos, Transactions American Geophysical Union* **90** 117–8

Broussard W and Turner R E 2009 A century of changing land-use and water-quality relationships in the continental US *Frontiers in Ecology and the Environment* **7** 302–7

Cao P, Lu C and Yu Z 2018 Historical Nitrogen Fertilizer Use in Agricultural Ecosystems of The Contiguous United States during 1850–2015: Application Rate, Timing, and Fertilizer Types *Earth System Science Data* **10** 969–984

Chen G, Tian H, Liu M, Ren W, Zhang C and Pan S 2006 Climate impacts on China's terrestrial carbon cycle: an assessment with the dynamic land ecosystem model *Environmental modeling and simulation* 56–70

Cheng F Y, Van Meter K J, Byrnes D K and Basu N B 2020 Maximizing US nitrate removal through wetland protection and restoration *Nature* **588** 625–30

Crews T E, Carton W and Olsson L 2018 Is the future of agriculture perennial? Imperatives and opportunities to reinvent agriculture by shifting from annual monocultures to perennial polycultures *Global Sustainability* **1**

Daniel G, He L, Jensen K L and English B C 2010 Expanded ethanol production: Implications for agriculture, water demand, and water quality *Biomass and bioenergy* **34** 1586–96

Dentener F J 2006 Global Maps of Atmospheric Nitrogen Deposition, 1860, 1993, and 2050 *Data set. Available on-line (http://daac. ornl. gov/) from Oak Ridge National Laboratory Distributed Active Archive Center, Oak Ridge, TN, USA*

Dominguez-Faus R, Powers S E, Burken J G and Alvarez P J 2009 The water footprint of biofuels: a drink or drive issue?

Evenson R E and Gollin D 2003a Assessing the impact of the Green Revolution, 1960 to 2000 *Science* **300** 758–62

Evenson R E and Gollin D (eds) 2003b *Crop variety improvement and its effect on productivity the impact of international agricultural research,* Cabi Publishing

Fixen P E, Williams R, and Rund Q B 2012 *NUGIS: A nutrient use geographic information system for the US*. Brookings, SD. Retrieved from: International Plant Nutrition Institute. http://nugis.ipni.net/Publication/

Foley J A, DeFries R, Asner G P, Barford C, Bonan G, Carpenter S R, Chapin F S, Coe M T, Daily G C and Gibbs H K 2005 Global consequences of land use *science* **309** 570–4

Gordon L J, Finlayson C M and Falkenmark M 2010 Managing water in agriculture for food production and other ecosystem services *Agricultural Water Management* **97** 512–9

Handmaker O, Keeler B L and Milz D 2021 What type of value information is most valuable to stakeholders? Multi-sector perspectives on the utility and relevance of water valuation information *Environmental Science & Policy* **115** 47–60

Helmers M J, Zhou X, Baker J L, Melvin S W and Lemke D W 2012 Nitrogen loss on tile-drained Mollisols as affected by nitrogen application rate under continuous corn and corn-soybean rotation systems *Canadian Journal of Soil Science* **92** 493–9

Hergoualc'h K, Akiyama H, Bernoux M, Chirinda N, del Prado A, Kasimir Å, MacDonald J D, Ogle S M, Regina K, & van der Weerden T J 2019 N_2O emissions from managed soils, and CO2emissions from lime and urea application. In E. Calvo Buendia, K. Tanabe, A. Kranjc, J. Baasansuren, M. Fukuda, S. Ngarize, A. Osako, Y. Pyrozhenko, P. Shermanau, & S. Federici (Eds.), 2019 Refinement to the 2006 IPCC guidelines for national greenhouse gas invento-ries, volume 4: Agriculture, forestry and other land use switzerland. Intergovernmental Panel on Climate Change (IPCC).

Lambin E F and Meyfroidt P 2011 Global land use change, economic globalization, and the looming land scarcity *Proceedings of the National Academy of Sciences* **108** 3465–72

Lark T J, Hendricks N P, Smith A, Pates N, Spawn-Lee S A, Bougie M, Booth E G, Kucharik C J and Gibbs H K 2022 Environmental outcomes of the US Renewable Fuel Standard *Proceedings of the National Academy of Sciences* **119** e2101084119

Lee M, Shevliakova E, Malyshev S, Milly P C D and Jaffé P R 2016 Climate variability and extremes, interacting with nitrogen storage, amplify eutrophication risk *Geophysical Research Letters*

Licht M and Archontoulis S V 2017 Influence of Drought on Corn and Soybean *Integrated Crop Management News, Iowa State University Extension and Outreach* Online: https://crops.extension.iastate.edu/cropnews/2017/07/influence-drought-corn-and-soybean

Liu M, Tian H, Chen G, Ren W, Zhang C and Liu J 2008 Effects of Land-Use and Land-Cover Change on Evapotranspiration and Water Yield in China During 1900-2000 1 *Journal of the American Water Resources Association* **44** 1193–207

Liu M, Tian H, Yang Q, Yang J, Song X, Lohrenz S E and Cai W J 2013 Long-term trends in evapotranspiration and runoff over the drainage basins of the Gulf of Mexico during 1901-2008 *Water Resources Research* **49** 1988–2012

Lohrenz S E, Redalje D G, Cai W-J, Acker J and Dagg M 2008 A retrospective analysis of nutrients and phytoplankton productivity in the Mississippi River plume *Continental Shelf Research* **28** 1466–75

Lu C, Yu Z, Hennessy D A, Feng H, Tian H and Hui D 2022 Emerging weed resistance increases tillage intensity and greenhouse gas emissions in the US corn–soybean cropping system *Nature Food* **3** 266–74

Lu C, Yu Z, Tian H, Hennessy D A, Feng H, Al-Kaisi M, Zhou Y, Sauer T and Arritt R 2018 Increasing carbon footprint of grain crop production in the US Western Corn Belt *Environmental Research Letters* **13** 124007

Lu C, Yu Z, Zhang J, Cao P, Tian H and Nevison C 2021 Century-long trends and drivers of soil N2O emissions across the contiguous United States *Global Change Biology* **28** 2505–24

Lu C, Zhang J, Cao P and Hatfield J 2019 Are We Getting Better in Using Nitrogen?: Variations in Nitrogen Use Efficiency of Two Cereal Crops Across the United States *Earth's Future* **7** 939–52

Lu C, Zhang J, Tian H, Crumpton W G, Helmers M J, Cai W-J, Hopkinson C S and Lohrenz S E 2020 Increased extreme precipitation challenges nitrogen load management to the Gulf of Mexico *Communications Earth & Environment* **1** 21 Online: https://doi.org/10.1038/s43247-020-00020-7

Ma L, Malone R W, Heilman P, Jaynes D B, Ahuja L R, Saseendran S A, Kanwar R S and Ascough Ii J C 2007 RZWQM simulated effects of crop rotation, tillage, and controlled drainage on crop yield and nitrate-N loss in drain flow *Geoderma* **140** 260–71

Malone R W, Kersebaum K C, Kaspar T C, Ma L, Jaynes D B and Gillette K 2017 Winter rye as a cover crop reduces nitrate loss to subsurface drainage as simulated by HERMES *Agricultural water management* **184** 156–69

Mananze S, Pôças I and Cunha M 2019 Agricultural drought monitoring based on soil moisture derived from the optical trapezoid model in Mozambique *Journal of Applied Remote Sensing* **13** 24519

Marshall E, Aillery M, Ribaudo M, Key N, Sneeringer S, Hansen L, Malcolm S and Riddle A 2018 *Reducing nutrient losses from cropland in the Mississippi/Atchafalaya River Basin: Cost efficiency and regional distribution*

Mesinger F, DiMego G, Kalnay E, Mitchell K, Shafran P C, Ebisuzaki W, Jović D, Woollen J, Rogers E and Berbery E H 2006 North American regional reanalysis *Bulletin of the American Meteorological Society* **87** 343–60

Van Meter K J, Van Cappellen P and Basu N B 2018 Legacy nitrogen may prevent achievement of water quality goals in the Gulf of Mexico *Science*

Mitchell T D and Jones P D 2005 An improved method of constructing a database of monthly climate observations and associated high-resolution grids *International Journal of Climatology* **25** 693–712

National Research Council 2009 *Nutrient control actions for improving water quality in the Mississippi River Basin and Northern Gulf of Mexico* (National Academies Press)

Ni X, Parajuli P B, Ouyang Y, Dash P and Siegert C 2021 Assessing land use change impact on stream discharge and stream water quality in an agricultural watershed *Catena* **198** 105055

Oelsner G P and Stets E G 2019 Recent trends in nutrient and sediment loading to coastal areas of the conterminous US: Insights and global context *Science of the Total Environment* **654** 1225–40

Ramankutty N, Mehrabi Z, Waha K, Jarvis L, Kremen C, Herrero M and Rieseberg L H 2018 Trends in global agricultural land use: implications for environmental health and food security *Annual review of plant biology* **69** 789–815

Ruehr S, Keenan TF, Williams C. et al. 2023 Evidence and attribution of the enhanced land carbon sink. *Nat Rev Earth Environ* 4, 518–534. https://doi.org/10.1038/s43017-023-00456-3

Robertson D M and Saad D A 2021 Nitrogen and phosphorus sources and delivery from the Mississippi/Atchafalaya River basin: An update using 2012 SPARROW models *JAWRA Journal of the American Water Resources Association* **57** 406–29

Robertson D M and Saad D A 2013 SPARROW models used to understand nutrient sources in the Mississippi/Atchafalaya River Basin *Journal of Environmental Quality* **42** 1422–40

Sabo R D, Clark C M, Bash J, Sobota D, Cooter E, Dobrowolski J P, et al. 2019 Decadal shift in nitrogen inputs and fluxes across the contiguous United States: 2002–2012. *Journal of Geophysical Research: Biogeosciences*, 124, 3104–3124. https://doi.org/10.1029/2019jg005110

Secchi S, Gassman P W, Jha M, Kurkalova L and Kling C L 2011 Potential water quality changes due to corn expansion in the Upper Mississippi River Basin *Ecological Applications* **21** 1068–84

Seitzinger S P, Mayorga E, Bouwman A F, Kroeze C, Beusen A H W, Billen G, Van Drecht G, Dumont E, Fekete B M and Garnier J 2010 Global river nutrient export: A scenario analysis of past and future trends *Global biogeochemical cycles* **24**

Stackpoole S, Sabo R, Falcone J, and Sprague L 2021. Long-term Mississippi River trends expose shifts in the river load response to watershed nutrient balances between 1975 and 2017. *Water Resources Research,* 57, e2021WR030318. https://doi.org/10.1029/2021WR030318

Stets E G, Sprague L A, Oelsner G P, Johnson H M, Murphy J C, Ryberg K, Vecchia A V, Zuellig R E, Falcone J A and Riskin M L 2020 Landscape drivers of dynamic change in water quality of US rivers *Environmental Science & Technology* **54** 4336–43

Tian H, Lu C, Pan S, Yang J, Miao R, Ren W, Yu Q, Fu B, Jin F-F and Lu Y 2018 Optimizing resource use efficiencies in the food–energy–water nexus for sustainable agriculture: From conceptual model to decision support system *Current Opinion in Environmental Sustainability* **33** 104–13

Tian H, Xu R, Pan S, Yao Y, Bian Z, Cai WJ, Hopkinson CS, Justic D, Lohrenz S, Lu C and Ren W 2020 Long-term trajectory of nitrogen loading and delivery from Mississippi River Basin to the Gulf of Mexico. *Global Biogeochemical Cycles*, **34(5)**, p.e2019GB006475.

Tian H, Xu X, Liu M, Ren W, Zhang C, Chen G and Lu C 2010 Spatial and temporal patterns of CH4 and N2O fluxes in terrestrial ecosystems of North America during 1979–2008: application of a global biogeochemistry model *Biogeosciences* **7** 2673–94

Turner R E and Rabalais N N 2003 Linking landscape and water quality in the Mississippi River

Basin for 200 years *Bioscience* **53** 563–72

U.S. Environmental Protection Agency 2001 *Action plan for reducing, mitigating, and controlling hypoxia in the Northern Gulf of Mexico* (Washington, DC: Office of Wetlands, Oceans, and Watersheds, US Environmental Protection Agency)

Vitousek P M, Aber J D, Howarth R W, Likens G E, Matson P A, Schindler D W, Schlesinger W H and Tilman D G 1997 Human alteration of the global nitrogen cycle: sources and consequences *Ecological Applications* **7** 737–50

Wang H and Ortiz-Bobea A 2019 Market-driven corn monocropping in the US Midwest *Agricultural and Resource Economics Review* **48** 274–96

Wei Y, Liu S, Huntzinger D N, Michalak A M, Viovy N, Post W M, Schwalm C R, Schaefer K, Jacobson A R, Lu C, Tian H, Ricciuto D M, Cook R B, Mao J and Shi X 2014 The north american carbon program multi-scale synthesis and terrestrial model intercomparison project - Part 2: Environmental driver data *Geoscientific Model Development* **7** 2875–93

Wei Z, Ying H, Guo X, Zhuang M, Cui Z and Zhang F 2020 Substitution of mineral fertilizer with organic fertilizer in maize systems: A meta-analysis of reduced nitrogen and carbon emissions *Agronomy* **10** 1149

Wu Y, Liu S, Sohl T L and Young C J 2013 Projecting the land cover change and its environmental impacts in the Cedar River Basin in the Midwestern United States *Environmental Research Letters* **8** 24025

Yang H, Wang G, Wang L and Zheng B 2016 Impact of land use changes on water quality in headwaters of the Three Gorges Reservoir *Environmental Science and Pollution Research* **23** 11448–60

Yu Z and Lu C 2018 Historical cropland expansion and abandonment in the continental US during 1850 to 2016 *Global Ecology and Biogeography* **27** 322–33

Yu Z, Lu C, Cao P and Tian H 2018 Long-term terrestrial carbon dynamics in the Midwestern United States during 1850-2015: Roles of land use and cover change and agricultural management *Global change biology* **24** 2673–90

Yu Z, Lu C, Tian H and Canadell J 2019 Largely underestimated carbon emission from land use and cover change in the conterminous U.S. *Global Change Biology* **25** 3741–52 Online: doi:10.1111/gcb.14768.

Zambrano-Bigiarini Mauricio. 2017 Package 'hydroGOF': Goodness-of-fit functions for comparison of simulated and observed hydrological time series. R package version 0.4-0. URL https://github.com/hzambran/hydroGOF. DOI:10.5281/zenodo.839854.

Zhang J, Cao P and Lu C 2021 Half-century history of crop nitrogen budget in the conterminous United States: Variations over time, space and crop types *Global Biogeochemical Cycles* e2020GB006876

Zhang J, Lu C, Crumpton W, Jones C, Tian H, Villarini G, Schiling K and Green D 2022 Heavy Precipitation Impacts on Nitrogen Loading to the Gulf of Mexico in the 21st Century: Model projections under future climate scenarios *Earth's Future* e2021EF002141