

Regional Analysis of Nitrogen Flow within the Chesapeake Bay Watershed Food Production Chain Inclusive of Trade

Paniz Mohammadpour and Caitlin Grady*



Cite This: *Environ. Sci. Technol.* 2023, 57, 4619–4631



Read Online

ACCESS |

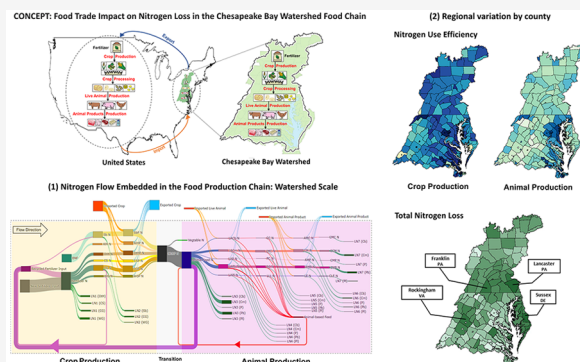
Metrics & More

Article Recommendations

Supporting Information

ABSTRACT: In the Chesapeake Bay Watershed, excess nitrogen has contributed to poor water quality, leading to nitrogen mitigation efforts to restore and protect the watershed. The food production system is a top contributor to this nitrogen pollution. While the food trade plays a vital role in distancing the environmental impacts of nitrogen use from the consumer, previous work on nitrogen pollution and management in the Bay is yet to carefully consider the effect of embedded nitrogen found in products (nitrogen mass within the product) imported and exported throughout the Bay. Our work advances understanding across this area by creating a mass flow model of nitrogen embedded in the food production chain throughout the Chesapeake Bay Watershed that separates phases of the production and consumption processes for crops, live animals, and animal products and considers commodity trade at each phase by combining aspects of both nitrogen footprint and nitrogen budget models. Also, by tracking nitrogen embedded in products imported and exported in these processes, we distinguished between direct nitrogen pollution and nitrogen pollution externalities (displaced N pollution from other regions) from outside of the Bay. We developed the model for the watershed and all its counties for major agricultural commodities and food products for 4 years 2002, 2007, 2012, and 2017 with a specific focus on 2012. Using the developed model, we determined the spatiotemporal drivers of nitrogen loss to the environment from the food chain within the watershed. Recent literature leveraging mass balance approaches has suggested that previous long-term declines in nitrogen surplus and improvements in nutrient use efficiency have stagnated or begun to reverse. Our results suggest that within the Chesapeake Bay, increased corn and wheat acreage and steadily increasing livestock/poultry production may have led to the stagnation in decreasing N loss trends from agricultural production observed over the past two decades. We also show that at the watershed scale, trade has reduced the food chain nitrogen loss by about 40 million metric tons. This model has the potential to quantify the effect of various decision scenarios, including trade, dietary choices, production patterns, and agricultural practices, on the food production chain nitrogen loss at multiple scales. In addition, the model's ability to distinguish between nitrogen loss from local and nonlocal (due to trade) sources makes it a potential tool to optimize regional domestic production and trade to meet local watershed's needs while minimizing the resulting nitrogen loss.

KEYWORDS: food production chain, food trade, nitrogen footprint, nitrogen budget, nitrogen loss, substance flow analysis



1. INTRODUCTION

While reactive nitrogen (Nr) is the primary nutrient source for plants, animals, and humans and plays a crucial role in food production efficiency, large amounts of Nr used in agricultural production are lost to the environment.^{1–5} The food production system is a top contributor to nitrogen pollution worldwide.^{6–8} In the Chesapeake Bay Watershed, agricultural runoff accounts for more than 40 percent of the Bay's nitrogen pollution, contributing substantially to the Bay's poor ecosystem health.⁹ To have a productive and environmentally protective agricultural system, it is necessary to balance the positive and negative impacts of Nr use in this system.

Nitrogen use efficiency (NUE) and Nr loss are two metrics that can quantify aspects of constructive and destructive effects of nitrogen (N) in a food production chain in an attempt to

understand nitrogen system balance. A food production chain incorporates commodity production, processing, distribution, and consumption processes.¹⁰ For a given commodity in the food chain, NUE is defined as a ratio of N available for the consumption of that commodity (output N) to the N input to produce it (input N), and Nr loss is the difference between input N and output N.¹⁰ To increase NUE and decrease Nr loss in the food chain, we first need to investigate how N inputs to the

Received: October 18, 2022

Revised: February 6, 2023

Accepted: February 7, 2023

Published: March 8, 2023



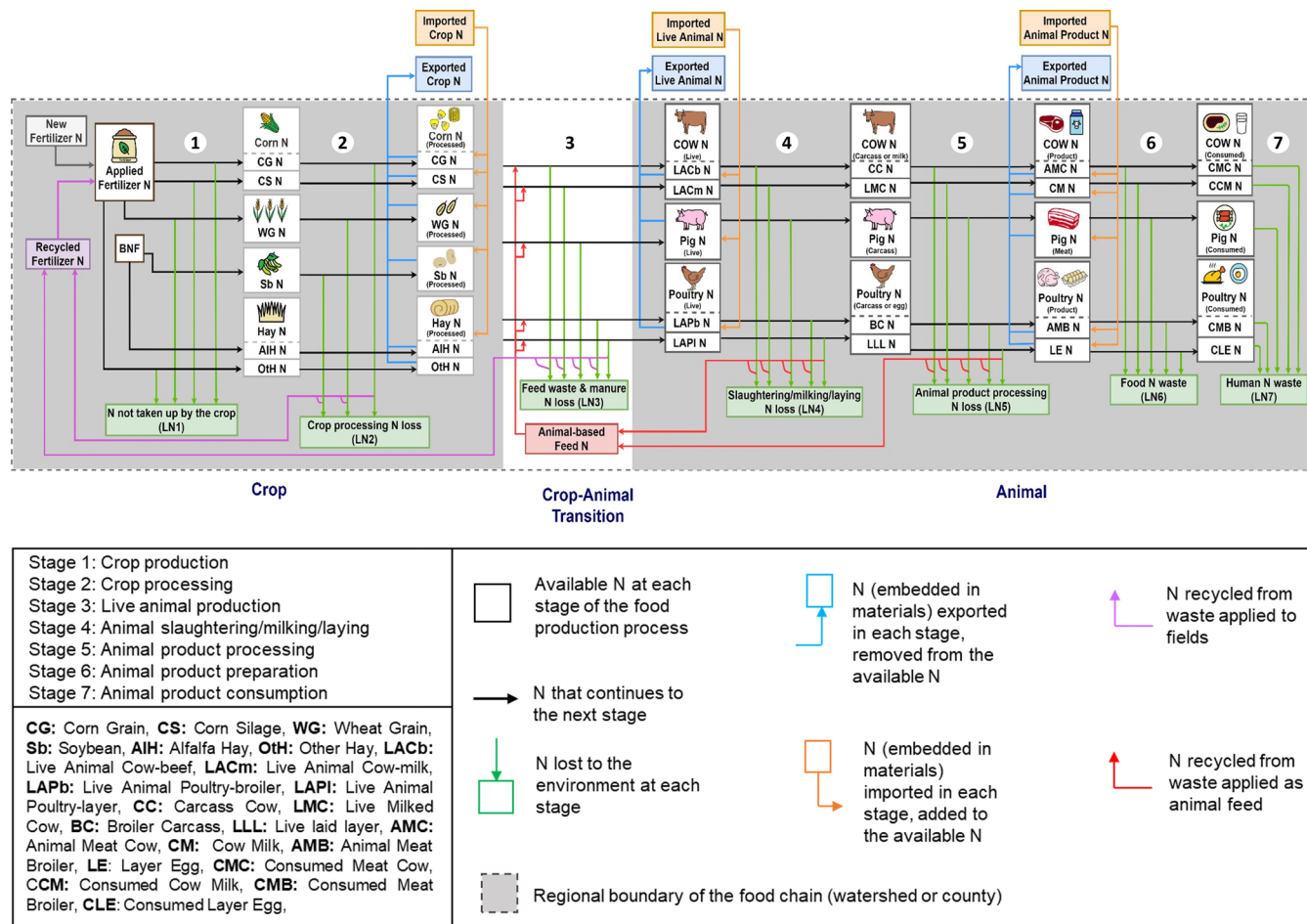


Figure 1. Nitrogen Flow model of the Chesapeake Bay watershed Food chain (NFCBF).

system flow through these processes.⁶ Nitrogen loss generated during these processes and NUE in the system are controlled by several factors, including landscape characteristics,^{11,12} production-based factors like land use,^{8,13,14} and agricultural practices,¹⁵ consumption-based factors like dietary choices,^{15,16} and food trade patterns.^{15–18} These factors also vary spatially and temporally.^{4,14,19–21} Combining these factors allows for the development of a comprehensive model that can quantify and track N flow and Nr loss in the food production chain in a region over time, in turn helping to manage Nr loss and NUE by determining the spatiotemporal drivers of Nr loss.

1.1. Embedded Nitrogen Accounting Tools. Nitrogen footprint (NF) and nitrogen budget (NB) methods are two common tools to track the mass flow of N embedded in commodities in a food production chain and quantify Nr loss and NUE. N-Calculator serves as an N footprint tool that estimates total nitrogen loss to the environment during the production and consumption of the food consumed by an individual.⁶ These models enable decision-makers and stakeholders to realize how consumers’ dietary choices relate to Nr loss to the environment. Despite this important use, these calculators do not determine where Nr loss occurs while this is crucial to find the Nr loss hotspots and prioritize regions for management. Also, they often do not provide detailed information on how commodity trade affects the Nr loss amount and spatial flow. Despite the further adjustments in NF models by incorporating food and feed trade¹⁵ and a per-area basis estimation of Nr loss intensity,^{1,22} they still estimate the

total Nr loss from an entity’s food consumption without determining where the loss is released.

Using the NB method, which models nitrogen inputs and outputs across a system’s boundary, we can quantify N flow and estimate NUE and Nr loss at each stage of the food chain for any region. Previous studies on food chain NB modeling have considered a wide range of inputs and outputs such as considering nitrogen fertilizer necessary for food production as system boundary imports and nitrogen loss from food product consumption as system boundary exports.^{10,14,23–27} Additionally, the food chain NB model has been used at multiple spatial scales, including farm level,^{27,28} regional, watershed,¹⁴ and country scale.²⁶ The N budget method has also been used to determine the final pathways of Nr loss^{4,27,30} and the spatial distribution of Nr loss and Nr pollution hotspots.^{14,24,25,31} In terms of agricultural efficiency, several studies have utilized an NB model to estimate NUE and determine its spatial variation.^{5,10,26,27,32–35} Hence, using the NB models alongside NF models advances it to include other aspects of a food production chain, like production and trade, rather than just consumption which enables the model to determine where the N loss occurs and estimate the displaced N loss due to trade.

1.2. Regional Dimension. Despite decades of nutrient management efforts to reduce nutrient loading in the Chesapeake Bay, nutrient pollution has remained a top challenge. Most research and practice in the Chesapeake Bay Watershed on reducing nitrogen pollution have focused on nitrogen leaching to the water bodies with less attention to total

Table 1. Developed Variables and Equations for the NFCBF Model

variable	Figure 1	description	equation	unit	eq
NI_{nk}	black arrow	N from stage $n - 1$ makes it to the stage n for commodity k	Table S1	ton N	
N_{nk-1}	black box	available N at stage $n - 1$ for consumption of commodity k	Table S1	ton N	
WN_{nk}	green arrow's start point	N waste produced at stage n for commodity k	$(N_{nk-1} - NI_{nk})$	ton N	1
RN_{nk}	purple and red arrows' start point	N recycled from N waste at stage n of commodity k	Tables S1 and S3	ton N	
Im_{nk}	NA	imported commodity k to the region at stage n	Table S6 and Section S1-2	ton N	
Ex_{nk}	NA	exported commodity k from the region at stage n	Table S6 and Section S1-2	ton N	
Nc_k	NA	N content of commodity k	Tables S2 and S4	$\frac{\text{ton N}}{\text{ton Commodity}}$	
ImN_{nk}	orange arrow	N embedded in imported commodity k to the region at stage n	$Im_{nk} \times Nc_k$	ton N	2
ExN_{nk}	blue arrow	N embedded in the exported commodity k from the region at stage n	$Ex_{nk} \times Nc_k$	ton N	3
RNI_{nk}	purple and red arrows' endpoint	N recycled input to stage n for commodity k	Tables S1 and S2	ton N	
N_{nk}	black box	available N of commodity k for consumption at stage n	$NI_{nk} + RNI_{nk} + ImN_{nk} - ExN_{nk}$	ton N	4
LN_{nk}	green arrow's endpoint	N loss at stage n for commodity k	$WN_{nk} - RN_{nk}$	ton N	5
NUE_{nk}	NA	nitrogen use efficiency at stage n for commodity k	$\frac{N_{nk}}{N_{nk-1} + ImN_{nk} - ExN_{nk} - RN_{nk}}$	ratio	6

nitrogen pollution considering the N_r loss to the atmosphere through volatilization and denitrification in addition to nitrogen leaching.^{36–38} Several previous studies targeting reducing N_r pollution have been at the field and farm scale using Best Management Practices (BMPs),^{38,39} neglecting the importance of multiple interactions at the landscape or regional scale.^{40–43}

Despite the important role food import and export play in distancing the environmental impacts of nitrogen use from where the commodity is produced to where it may be processed or consumed,^{15,17,18} current studies assessing nitrogen loss in the Chesapeake Bay have limited consideration for traded goods. In 2007, the Mid-Atlantic Water Program incorporated regional aspects in estimating total agricultural N_r loss in the Bay Watershed. They determined the regional variation of agricultural nitrogen balance in the Bay Watershed by developing nitrogen budgets of croplands in mid-Atlantic counties for 2007.⁴⁴ In addition, there are various recent studies on the Bay's nitrogen management, including Sabo et al.⁴⁵ and Ator et al.,³⁶ in which they investigated the spatiotemporal patterns and drivers of nitrogen loss sources in the watershed. Although these models successfully showcased agricultural N_r loss and NUE regional variation, the model's nitrogen inputs-outputs within the agricultural system focus on crop production and livestock waste. To further disentangle nitrogen loss, research on where within the food production chain N_r loss increases and NUE decreases could enhance knowledge of nitrogen loss and potential management solutions. Additionally, how N_r changes due to trade would further existing approaches.

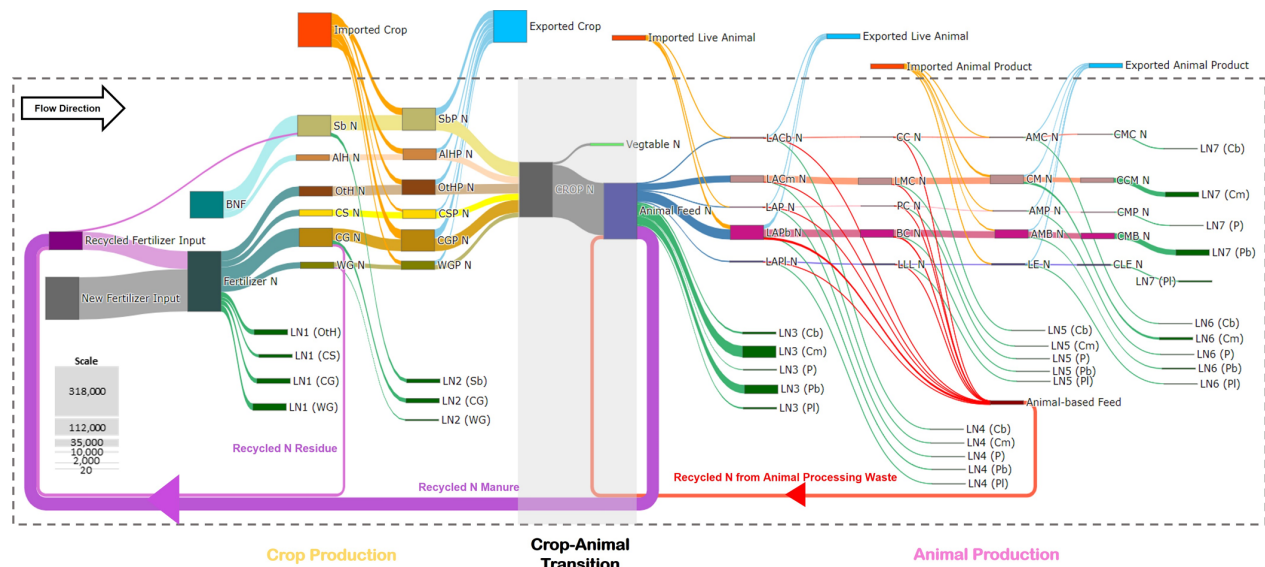
1.3. Research Goals. Leveraging nitrogen budget and nitrogen footprint models could enable tracking nitrogen flows embedded in food production, consumption, and trade in a given region. To improve understanding of spatiotemporal drivers of N_r loss in the Chesapeake Bay Watershed by highlighting the role of food trade on regional N_r loss and NUE, this research seeks to (1) develop detailed modeling of nitrogen flow of food production, consumption, and trade in the Chesapeake Bay Watershed at both watershed and regional scales; (2) determine NUE and N_r loss at each stage of the food production chain and show their regional variability in the watershed; (3) distinguish between local and nonlocal N_r loss due to commodity trade, and (4) analyze temporal trends of food production chain N_r loss to the environment across the watershed.

2. METHODOLOGY

The Chesapeake Bay Watershed spans 194 counties within the six states of Delaware (DE), Maryland (MD), New York (NY), Pennsylvania (PA), Virginia (VA), and West Virginia (WV), and the District of Columbia, collectively 195 regions (Figure S1). In line with our goals, we developed a Nitrogen Flow model of the Chesapeake Bay Watershed Food production chain (hereby referred to as NFCBF) for the watershed, and all its counties, by combining aspects of both nitrogen footprint and nitrogen budget models for 4 years 2002, 2007, 2012, and 2017 with a specific focus on 2012 (Figures 1 and S2). The model consists of a chain of commodity production, processing, consumption, import, and export within the boundaries of a region across seven stages (Figure 1). The model starts with fertilizer application to crops and Biological Nitrogen Fixation (BNF) of legumes which are considered as new nitrogen inputs to the model. As nitrogen flows throughout the chain through different stages, parts of the available nitrogen in commodities flow to the next stage, and the rest will be the nitrogen waste. Parts of the nitrogen waste at each stage may be recycled back to the system, serving as recycled nitrogen input, and the rest will be lost to the environment. By nitrogen loss, we are specifically estimating all N_r loss released to the environment, including water, air, and soil.

2.1. Construction of the NFCBF Model. The nitrogen mass-flow NFCBF model includes major agricultural commodities and food products and their production chains (Figure 1 and Section S1). This model separates phases of the production and consumption processes for crops, live animals, and animal products and considers regional import–export at each phase. For a given region in the watershed, food import–export consists of internal flows with the counties in the watershed as well as external flows with counties outside of the watershed. Nitrogen embedded in commodities imported to and exported from a region increases and decreases its available nitrogen, respectively. Model assumptions are listed in the supplemental document Section S2.

Primary input data to the NFCBF model are annual county-scale crop and animal production data from the US Department of Agriculture Census of Agriculture for 4 years 2002, 2007, 2012, and 2017,⁴⁶ food commodity trade data for 2012 from Freight Analysis Framework version 4 (FAF 4),⁴⁷ and Lin et al.,⁴⁸ crop and animal processing data, conversion factors, and



Legend	Crop Production																			
	CG	CS	WG	Sb	AIH	Oth	CGP	CSP	WGP	SbP	AIHP	OthP								
	Corn Grain	Corn Silage	Wheat Grain	Soybean	Wheat Hay	Other Hay	Corn Grain Processed	Corn Silage Processed	Wheat Grain Processed	Soybean Processed	Alfalfa Hay Processed	Other Hay Processed								
Animal Production	LACb		LACm		LAP		LAPb		LAPI		CC		LMC		PC		BC		LLL	
	Live Animal Cow-beef		Live Animal Cow-milk		Live Animal Pig		Live Animal Poultry-broiler		Live Animal Poultry-layer		Cow Carcass		Live Milked Cow		Pig Carcass		Broiler Carcass		Live Laid Layer	
	Animal Meat Cow		Cow Milk		Animal Meat Pig		Animal Meat Broilers		Layer Egg		Consumed Meat Cow		Consumed Cow Milk		Consumed Meat Pig		Consumed Meat Broilers		Consumed Layer Egg	
Nitrogen loss	LN: (j)																			
	Nitrogen Loss from commodity type j in stage i		1		2		3		4		5		6		7					
			N not taken up by crops		Crop processing N loss		Feed waste & manure N loss		Slaughtering/milking/laying N loss		Food Processing N loss		Food N waste		Human N waste					
		For crop commodities see crop production section (first row)																		
						Cb		Cm		P		Pb		PI						

Figure 2. Share of different components in the Nitrogen Flow of the Chesapeake Bay Watershed Food chain (year: 2012). The box size shows the greatness of available N (metric tons N) at a point (the larger the box, the more the amounts of available N at that point). The arrow's size shows the greatness of N flow between two boxes (the larger the arrow, the more the amounts of N flow between the boxes).

nitrogen content data (metric ton N/ton commodity) from agricultural agencies and the literature. It is worth mentioning that international trade flows are omitted in this study. We also conducted additional data processing for trade data to extract county-wise commodity flow data (Section S1-2). Detailed information on the data and data sources is presented in the supplementary document (Figure S2, Sections S1-1 and S1-2, and Tables S2 to S6).

To quantify different components of the model, we defined and used multiple variables and equations, a summary of which is discussed in Table 1, and the full information is provided in the supplementary document (Tables S1–S6). For any given region in the Chesapeake Bay Watershed, we developed nitrogen mass balance at each stage for every commodity (Table 1). Then, the NFCBF model components were developed and estimated (Table S1). The column titled “Figure 1” in Table 1 illustrates where major terms and equations fall in Figure 1. Throughout the manuscript we refer to ton to indicate metric ton (i.e., 1000 kg).

Finally, we calculated NUE at each stage for each commodity as a ratio of total N available for consumption (N embedded in the commodity in that stage) to total N inputs to produce that commodity minus the recycled N from waste in that stage (eq 6). Total N inputs at each stage include N available for consumption from the previous stage and net import N to the current stage. NUE varies between 0 and 1, indicating the least and the most efficient, respectively. We calculated NUE in our model following the approach presented by Erisman et al.¹⁰ and Guo et al.⁴

2.1.1. Uncertainty Analysis of Nitrogen Loss. We used several data sources, coefficients, and assumptions in our model subjected to uncertainties associated with many fields and calculation errors which raises the risk of uncertainty in the model outcomes. To advance knowledge around uncertainty for the NFCBF model, we assessed the effects of uncertainty in the models’ formulation, input data, and parameters on the Nr loss. We carried out uncertainty analysis through Monte Carlo simulation by assessing the range of variability within Nr loss due to the variability in the uncertain input data and parameters. To do so, we first determined the uncertain variables and their ranges of variability (Table S8). For any given uncertain variable, the range of variability has been determined according to the literature or the distribution that this variable follows. We then generated random numbers ($n = 5000$) using uniform or normal distribution for the uncertain variable (complete information is presented in the supplementary file in Section S4-2, Table S8). Finally, we obtained the variation of Nr loss at each stage and the total Nr loss by estimating them with random subsets of uncertain variables generated in the previous step.

2.2. Nitrogen Pollution Externalities in the Chesapeake Bay Watershed. Commodity import–export leads to spatial nitrogen flow due to the flow of nitrogen embedded in the commodities, displacing the Nr loss of food production–consumption and its environmental impacts.¹⁷ Analyzing the N flow due to commodity trade allows us to account for the environmental impacts of the commodity production within the exporter region while we transfer the impacts of commodity consumption Nr loss to the importer region (Figure S3). To

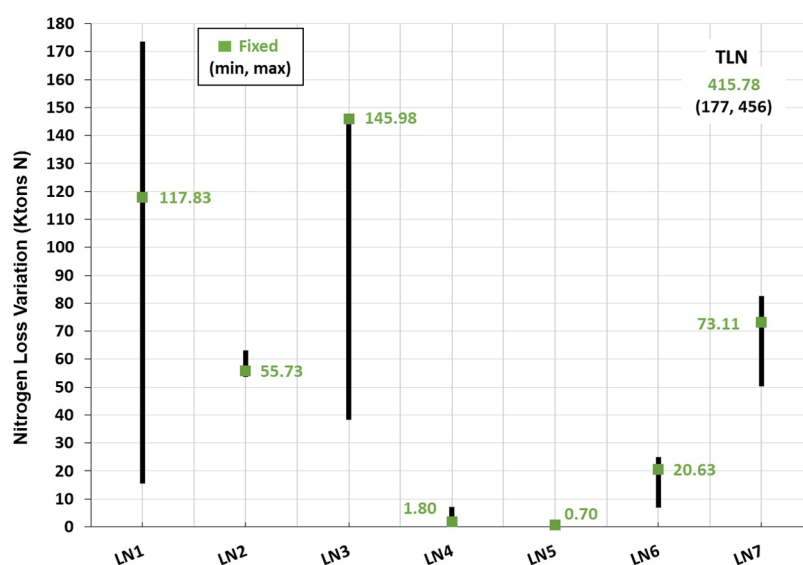


Figure 3. Fixed values of nitrogen loss at different stages (not considering uncertainty and estimated using constant variables for 2012, like Figure 2) compared to the extreme low and high thresholds and the range of indices variation (considering uncertainty in variables) utilizing Mont Carlo simulation (year: 2012) LN1: loss from crop production; LN2: loss from crop processing; LN3: loss from live animal production; LN4: loss from animal slaughtering/milking/laying; LN5: loss from animal product processing; LN6: loss from animal product preparation; LN7: loss from animal product consumption; TLN: total N Loss.

distinguish between the nitrogen pollution released to a region in the Chesapeake Bay Watershed resulting from food production and consumption in that region and outside of that region, we estimated the nitrogen pollution externalities of food production and consumption for a given region as eq 7:

$$\text{ExLN}_i = \sum_j \text{ExLN}_{ij} \quad (7)$$

where ExLN_i is the total nitrogen pollution externalities in region i (metric tons N), and ExLN_{ij} is nitrogen pollution externalities resulting from import and export of commodity j to and from region i (metric tons N). commodity j includes the commodities in the NFCBF model. We estimated ExLN_{ij} as eq 8:

$$\text{ExLN}_{ij} = \text{ExLNP}_{ij} + \text{ExLNC}_{ij} \quad (8)$$

where ExLNP_{ij} is production-based nitrogen pollution externality (metric tons N) and ExLNC_{ij} is consumption-based nitrogen pollution externality (metric tons N). We calculated ExLNP_{ij} and ExLNC_{ij} using eqs 9 and 10, respectively.

$$\text{ExLNP}_{ij} = \frac{\text{TLNP}_{ij} \times (\text{ExN}_{ij} - \text{ImN}_{ij})}{\text{DPN}_{ij}} \quad (9)$$

$$\text{ExLNC}_{ij} = \frac{\text{TLNC}_{ij} \times (\text{ImN}_{ij} - \text{ExN}_{ij})}{(\text{DPN}_{ij} + \text{ImN}_{ij} - \text{ExN}_{ij})} \quad (10)$$

where ImN_{ij} is nitrogen embedded in the imported commodity j to the region i (metric tons N), ExN_{ij} is nitrogen embedded in the exported commodity j from the region i (metric tons N), DPN_{ij} is nitrogen embedded in the domestic product of commodity j in the region i (metric tons N), TLNP_{ij} is the total Nr loss released to region i from the production of the domestic product of commodity j (metric tons N), and TLNC_{ij} is the total Nr loss released to region i from the consumption of available product of commodity j in region i (metric tons N) (more info in Section S2). For example, TLNP_{ij} for corn produced in region i is equal to

the sum of corn production (LN1) and corn processing (LN2) losses released to the region i , and TLNC_{ij} for live animal cow consumption in region i is the total of cow slaughtering (LN4) and cow meat processing (LN5) losses released to the region i .

Using the proposed approach (eqs 7–10), we determined nitrogen pollution externality of crops, live animals, and animal products for the Chesapeake Bay Watershed and each county in the watershed. For a county in the watershed, food import–export incorporates the flows between the county and other counties in the United States, including internal flows with other counties in the watershed as well as external flows with counties outside of the watershed. At the watershed scale, food import–export incorporates the flows between the watershed and all other counties in the United States outside of the watershed.

3. RESULTS

3.1. NFCBF Model Results. The NFCBF model results provide information on the share of N flows at each stage of the food chain, N recycled back to the system, the share of food trade in N flows, and the Nr loss at each stage for all commodities considered in the NFCBF model for the watershed (Figure 2, Table S7). Nitrogen recycled back to the system includes N recycled back to the system as fertilizer to crops from crop processing and manure wastes (purple arrows), and N recycled back to the system as animal feed from live animal and animal products processing wastes (red arrows).

3.1.1. Watershed-Scale NFCBF Components. We found that from the total nitrogen input to the crops in the watershed (526 ktons), about 70% is from fertilizer N (including new and recycled), and 30% is from BNF of legumes (Figure 2). New fertilizer N accounts for 70% of the total fertilizer N, and the recycled fertilizer N (purple arrows) for the remaining 30%. The recycled fertilizer N comes 28% from crop residues and 72% from animal manure. Our model reveals that corn grain and soybean not only contribute the most to domestic N production in the watershed, but they also have the largest share of N import to the watershed among commodities (Figure 2).

Table 2. Coefficient of Variation of Nitrogen Loss in the Chesapeake Bay Watershed and across States (Year: 2012)^a

state	LN1	LN2	LN3	LN4	LN5	LN6	LN7	TLN
watershed	32.90	2.87	22.35	28.79	34.61	24.49	8.82	13.34
Delaware (DE)	45.79	6.14	25.59	33.86	54.80	45.41	33.76	18.80
Maryland (MD)	37.50	3.39	26.30	34.78	44.19	22.52	11.52	13.31
New York (NY)	56.69	8.60	64.64	22.44	46.21	42.01	10.26	24.54
Pennsylvania (PA)	40.77	2.89	32.18	22.41	31.25	33.85	9.23	17.81
Virginia (VA)	22.63	3.31	18.68	29.64	42.35	18.39	10.52	10.63
West Virginia (WV)	21.10	5.25	20.77	31.54	47.37	22.57	14.25	13.07

^aLN1: loss from crop production; LN2: loss from crop processing; LN3: loss from live animal production; LN4: loss from animal slaughtering/milking/laying; LN5: loss from animal product processing; LN6: loss from animal product preparation; LN7: loss from animal product consumption; TLN: total N Loss.

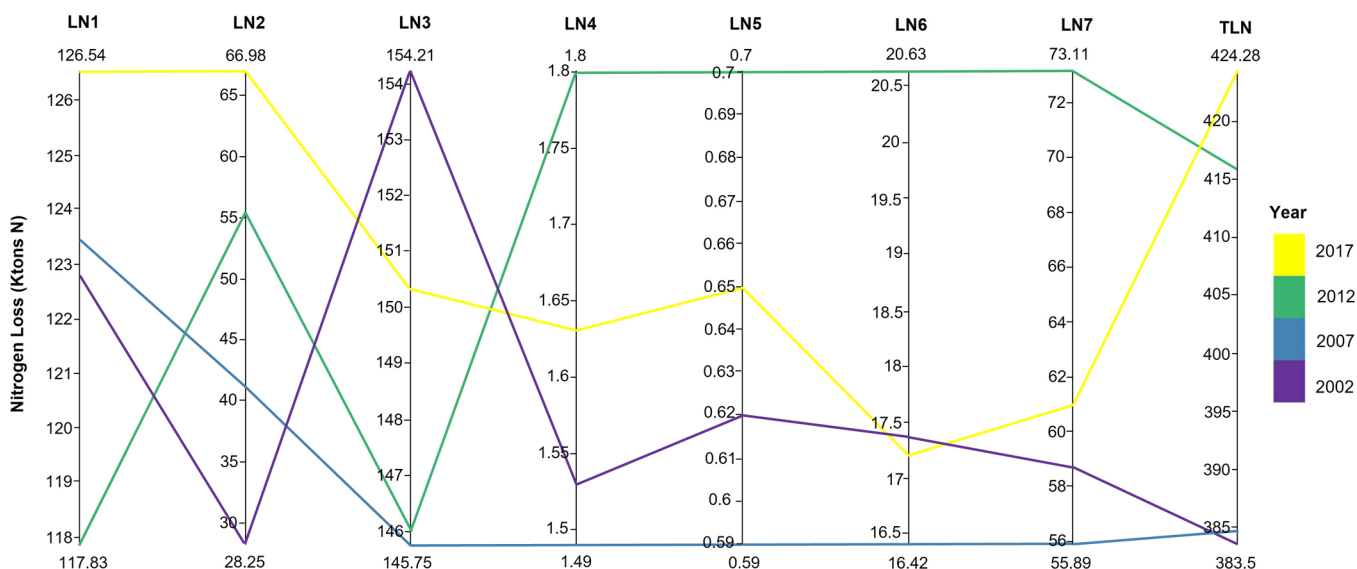


Figure 4. Temporal variation of nitrogen loss (LN) from the food chain in the Chesapeake Bay Watershed. Axes do not begin at 0. Each axis varies across the food chain in terms of magnitude. LN1: loss from crop production; LN2: loss from crop processing; LN3: loss from live animal production; LN4: loss from animal slaughtering/milking/laying; LN5: loss from animal product processing; LN6: loss from animal product preparation; LN7: loss from animal product consumption; TLN: total N Loss.

Among crops, corn grain and wheat grain production contributes the most in releasing N loss to the watershed environment, respectively (Figure 2). Although wheat grain is the second-largest source of crop production N loss in the watershed, it has the least domestic N production among crops. In contrast, soybean, the primary source of domestic crop N production (N within the harvested crop: SbP N Box in Figure 2) in the watershed, has the second least crop production N loss. This is not unexpected since soybeans are nitrogen-fixing legumes with very limited N applied in their production process.

Among animals, poultry-broiler and cow milk contribute the most to the domestic live animals and animal products N production and N loss in the watershed (Figure 2). Our results also indicate that the animal feed conversion ratio is one of the most significant values in determining nitrogen loss and nitrogen use efficiency. There are several regional trends in animal production captured throughout our model as well (Section 3.1.2).

In terms of trade, for the whole commodities in the watershed, N import (Figure 2, orange boxes) is 3% higher than N export (Figure 2, blue boxes); however, this is not indicative of net N changes because the import and export increase N losses within the Bay depending on where and when they are added to the system and which crop or animal product is being discussed. Crops contribute the most in both N import (78%) and N

export (78%). Live animals and animal products share a similar ratio in the remaining 22% for both N import and N export. Among all commodities, soybean has the largest net export, and alfalfa hay has the greatest net import.

Total nitrogen loss (TLN) shows a large variation band (177–456 ktons N) and a relatively high coefficient of variation (CV) (Figure 3 and Table 2). Among all stages, nitrogen loss at the first stage (crop production) and the third stage (live animal feeding) shows the largest variability (ktons N) and high CV. This is in line with our sensitivity analysis of the model that shows the most sensitive variables of the model as production quantity for both crops and animals, crops' fertilizer application rates, crops' yield N content, live animals' weight gain, and feed conversion ratio which are the main variables in estimating LN1 and LN3. Although LN4, LN5, and LN6 have high CVs, due to their low magnitude and variation bands compared to LN1 and LN3 will not be very effective on the total N loss variability (Figure 3 and Table 2). Crop processing loss (LN2) shows the lowest CV and a relatively small variation band, making it less effective on the TLN variation. On the other hand, LN7 with a relatively low CV has a relatively large variation band which makes it somehow effective on the TLN variation.

We also compared the uncertainty around nitrogen loss among the states located within the watershed (Table 2). The state-level CVs follow the same pattern as the watershed, with

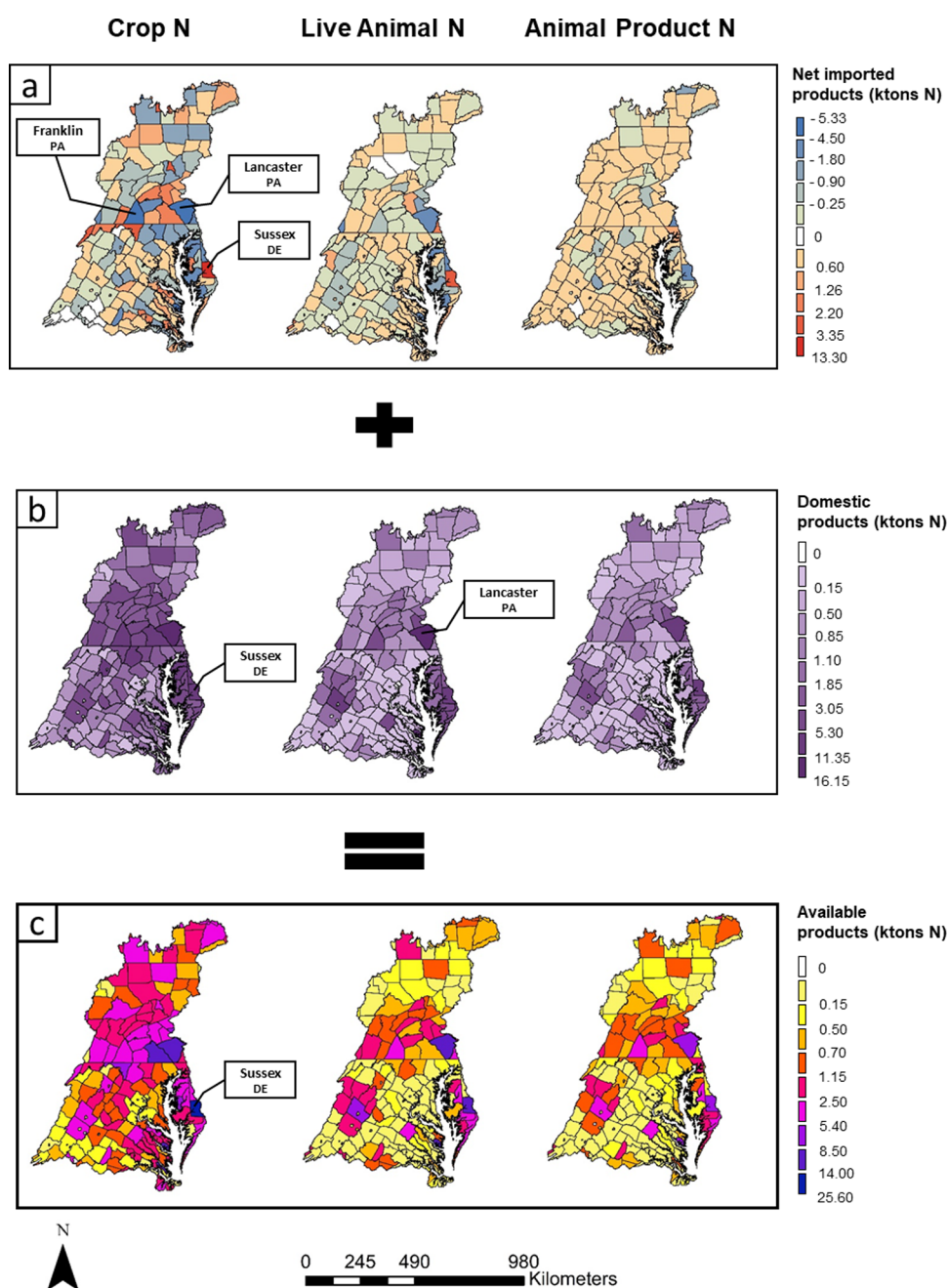


Figure 5. Regional variation of N embedded in (a) net imported products, (b) domestic products, and (c) available products for consumption in the Chesapeake Bay Watershed (year:2012).

the least amounts for LN2 and the highest amounts for LN5, except for New York and Pennsylvania, with the highest CV related to LN3 and LN1, respectively. In Delaware and New York, LN1 is very high, which is due to multiple factors, including the high amounts and high coefficient of variation for the crop production data and relatively high CV for crop fertilizer application rates and nitrogen content. Also, in New York, the CV for LN3 is very high, which is mostly affected by the high CV within the production quantity of live animals, specifically pigs, and the high CV of feed efficiency ratio for cow milk.

By developing the model for 2002, 2007, 2012, and 2017, we determined the temporal variation of nitrogen loss at every seven stages of the NFCBF model (LN1 to LN7) and the TLN (Figure 4). The year 2017 shows the highest nitrogen loss within the first

two stages (crop production and processing), influenced by a big jump in corn grain production compared to the previous years.

Despite the increase of corn grain and wheat grain in 2012 compared to the previous years, NL1 is the least in 2012, which is as a result of a large increase in soybean, which decreased the fertilizer need (Figure 4). Also, this production increase in 2012 for these three crops as the main sources of crop processing loss caused an increase in LN2 in 2012 compared to the previous years. Live animal production for all animal types except for poultry layers had the least values for 2012, which resulted in a significant reduction of LN3 (animal feeding loss) compared to 2002 and 2017. Despite the low domestic production of live animals in 2012 among years, this year became the largest source of nitrogen loss due to the live animal import, especially poultry broilers, to the watershed. Overall, the total N loss (TLN)

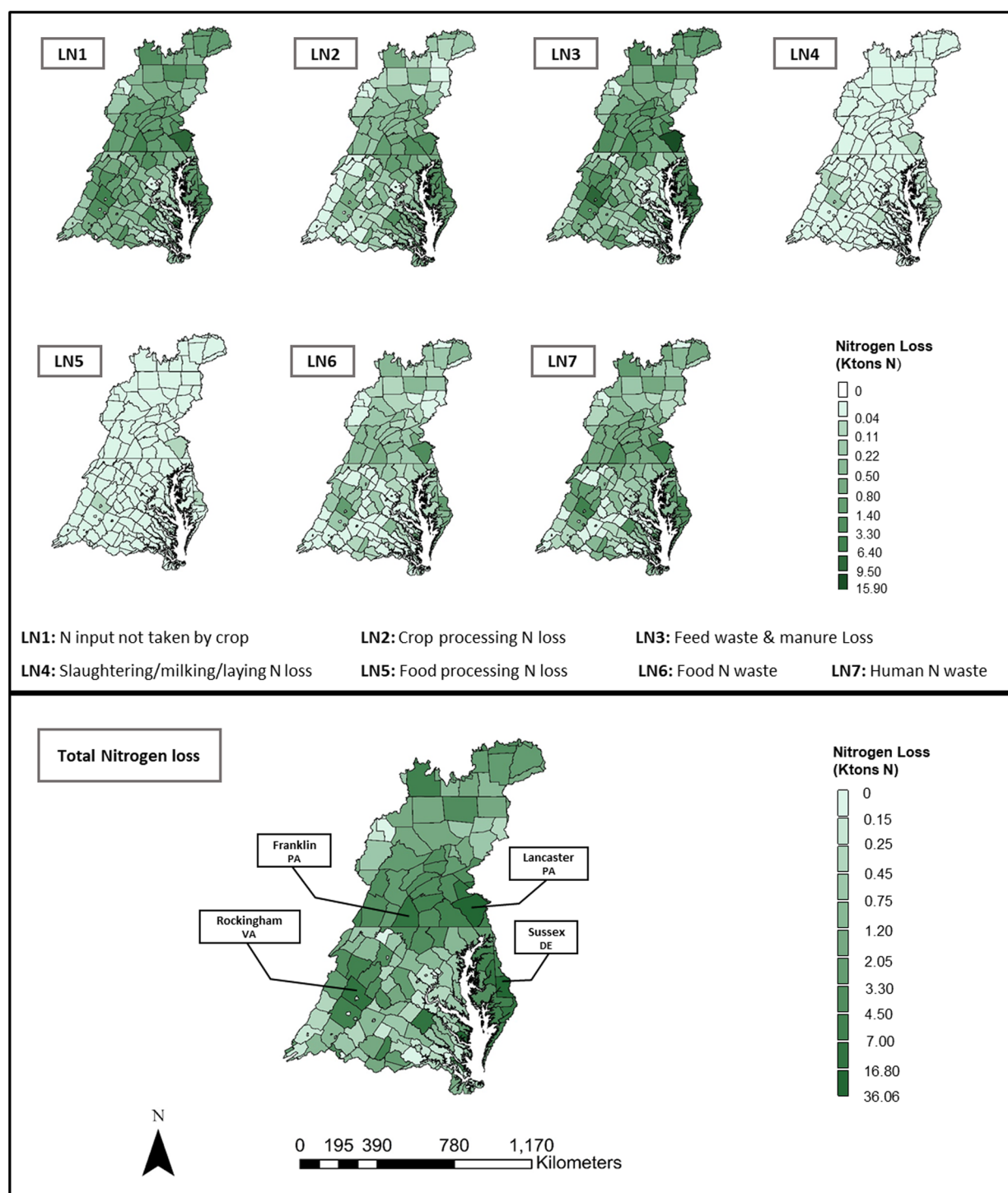


Figure 6. Regional variation of nitrogen loss (LN) from the food chain in the Chesapeake Bay Watershed (year:2012).

increased over the years despite not having a consistent and similar pattern for N loss in different stages over the years.

3.1.2. Regional Variation of NFCBF Components. We found the regional variation of net imported N (embedded in imported products), domestic production, and available N for consumption (embedded in available products) for crops, live animals, and animal products (Figure 5). Net imported N (import N minus export N) is positive when N embedded in the imported product is greater than N embedded in the exported product, and it is negative in the opposite situation. Although net imported N from crops and animal products are positive for the

whole watershed, there are several counties in the watershed with negative amounts (Figure 5a). We found an opposite trend for live animals, where the Chesapeake Bay region is a net exporter, with several counties still serving as importers.

Through this section, we will refer to the regions with positive amounts of net imported N as net-importing regions and the ones with negative amounts as net-exporting regions. Both highest regional net-exporting and net-importing N are related to crop trade at Franklin County (PA) and Sussex County (DE), respectively, (Figure 5a). In contrast to the watershed, alfalfa hay

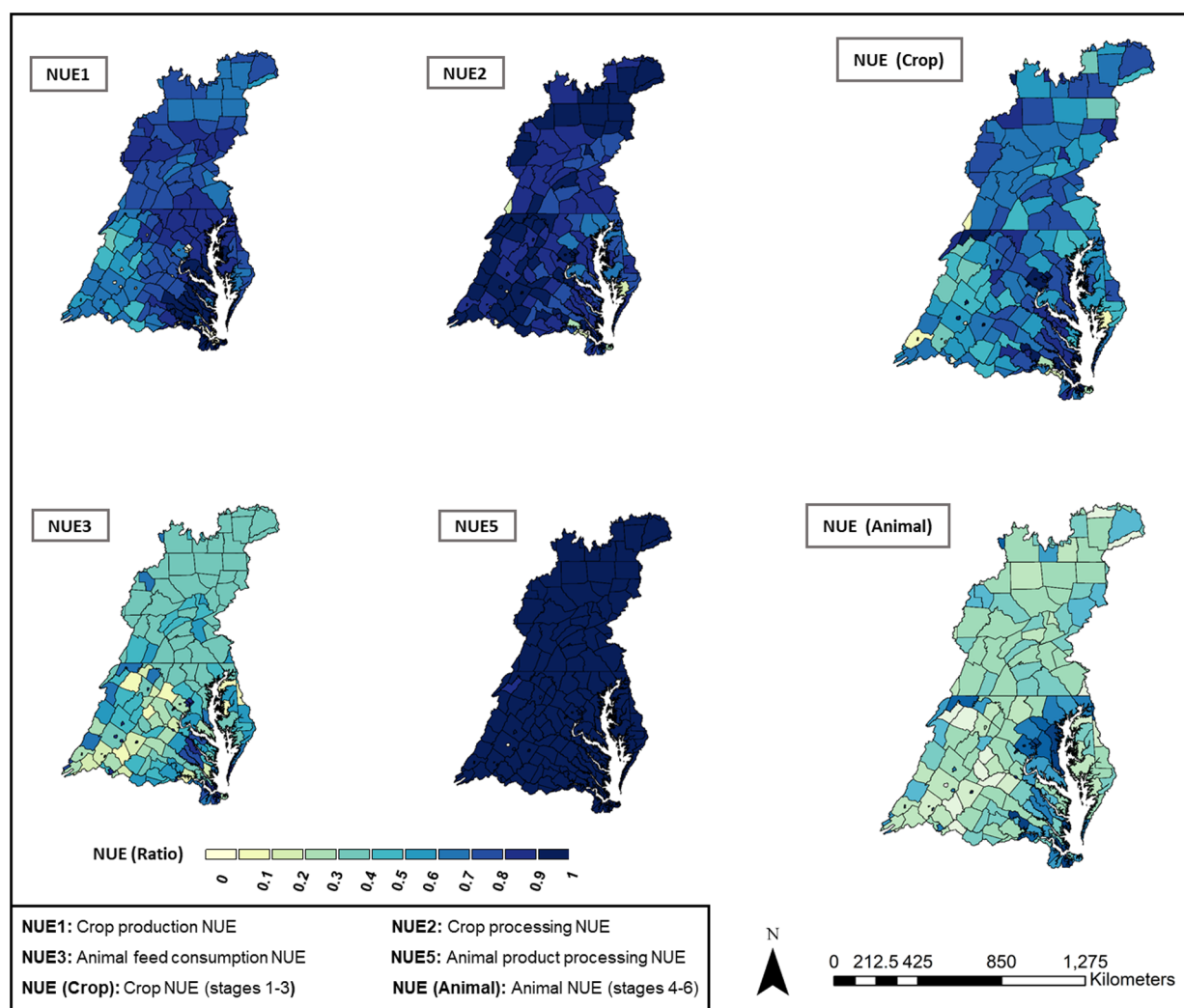


Figure 7. Spatial variation of food chain NUE in the Chesapeake Bay Watershed (year: 2012).

has the highest regional net export N (in Franklin County (PA)).

Consistent with the watershed results, crops are the primary source of domestic N production (i.e., the N mass within the domestically produced products) in most regions (Figure 5b). Lancaster County (PA) and Sussex County (DE) have the highest domestic N production amounts in all categories of crops, live animals, and animal products. Regional domestic N production for live animals and animal products follows the same pattern except for the net-exporting regions of live animals (e.g., Lancaster County (PA)), where N left the region due to export reduces available live animal N for producing animal products. Our model also accurately captures the largest broiler county in the country, Sussex County (DE),⁴⁹ which is shown to have the highest net-importing of crops and live animals and expectantly is a net-exporting region for those animal products (Figure 5a). In contrast, Franklin County (PA) and Lancaster County (PA), with the highest net exporting of crops and live animals, are net-importing regions of animal products showcasing their field crop and livestock commodity production focus (Figure 5a).

Available product N determines total N available for consumption in a region and is equal to the summation of local (domestic product N) and nonlocal (net imported N) sources. For regional available products N, live animals and

animal products follow the same pattern (influenced by domestic production) with lower amounts than the crops (Figure 5c). The highest amount of regional available product N for consumption is related to Sussex County (DE) crops with the highest regional net-importing N and domestic N production. The huge amounts of available crop N for feeding live animals in Sussex County (DE) (the largest poultry broiler county in the US) were expected. It is interesting that, although Lancaster County (PA) has the highest net-exporting of crops and live animals, it still is one of the main sources of available N for consumption due to the high amount of domestic N production.

In analyzing the regional variation of N loss, we found that the regional statistics for N loss follow the same pattern as watershed-level results, with the highest N loss in stages 3 and 1 and the lowest amounts in stages 5 and 4 (Figure 6, Table S9). The 10 most highly affected regions for the total N loss show similar patterns in almost all stages (Figure 6, Table S9). Compared to previous work, our crop production N loss results (LN1 and LN2) show a similar pattern with the highest amounts in the east center (Lancaster and Franklin counties of PA) and southwest of the watershed (Rockingham in VA) and the eastern shore of the Bay (Sussex in DE).^{19,50,51}

3.2. Nitrogen Pollution Externalities in NFCBF. The results of production-based nitrogen pollution externality

(ExLNP), consumption-based nitrogen pollution externality (ExLNC), and total nitrogen pollution externality (ExLN) for the watershed are derived as $-35,885$, -3476 , and $-39,361$ ktN, respectively, indicating that food trade reduces the food chain N loss in the watershed by about 40 million tons. To say it in a different way, (1) if citizens in the Chesapeake Bay Watershed were to consume all of their products' demands from the domestic products, we would have an even higher N loss in the environment; and (2) if all the food commodities produced in the Bay were supposed to consume locally, it would cause more N loss as well. Although for the whole watershed, N import and export amounts are pretty similar, the regional variability of trade in the watershed (Figure 5a) is responsible for the great amounts of N loss reduction.

We also determined a regional variation of ExLNP (Figure S4a), ExLNC (Figure S4b), and ExLN (Figure S4c) for the Chesapeake Bay Watershed. The spatial variation of ExLN shows a higher difference for live animals compared to crops and animal products. It also showcases that the nitrogen pollution externality is positive in most regions in the watershed for animal products which is influenced by consumption-based N loss because regional net import is mostly positive for animal products (Figures S4c and 5a). However, for crops, ExLN is negative in most regions where they are net-exporter of crop N. This means that in contrast to animal products, crop trade helped decrease the regional N loss in the watershed.

3.3. Nitrogen Use Efficiency of NFCBF. The results for NUE of each commodity at each stage (Table S10) show the lowest efficiency for stage 3 and the highest for stages 5 and 4, respectively, confirming the findings from N loss where stage 3 had the greatest amount and stages 5 and 4 the least. While NUE in stages 4 and 5 are very high, NUE (Crop) is considerably higher than NUE (Animal) due to the significant contribution of the third stage (manure) in the animal-related production processes. Also, the results for the NUE (total) for each commodity (last column, Table S10) show that alfalfa hay has the highest and the poultry layer has the least N use efficiency among all commodities in the watershed.

Considering recycled N and trade in the estimation of NUE (Figure S5) revealed that NUE has increased considerably for the watershed in the stages where there is N recycling (stages 2, 3, 4, and 5). This showcases that although trade enhanced the crop NUE slightly for the watershed, the highest contributing factor in increasing the NUE at the watershed scale was N recycling. On the other hand, regional variation of NUE in the watershed is mainly affected by food trade (Figure 7). For example, the net importing regions of crop N (Figure 5a) have higher amounts of NUE2 (crop processing) (Figure 7) because crop import increased the total nitrogen available for consumption in the region, while the crop is processed in another region. Likewise, the regional variation of NUE5 (animal product processing) (Figure 7) follows the same pattern as net imported animal product N (Figure 5a); both showcase a uniform regional variation.

Generally, the regional statistics for NUE follow the same spatial distribution pattern as watershed-level results, with the highest amounts in stages 5 and 2 and the lowest amounts in stage 3 and NUE-Animal (Figure 7). Also, the regional distribution of NUE (Animal) shows the highest counties concentrated on the west shore of the watershed, but the counties with the highest Crop NUE are scattered in the watershed. For both cases, the highest amounts are related to the net-importing counties with low domestic production.

Overall, the NUE spatial distribution in the watershed shows that the regional variation of NUE for different commodity types and different stages in the NFCBF model is mainly controlled by both commodity trade (NUE2 and NUE5) and the recycled input (NUE1 and NUE3). The influence of recycled input is quite expected given the impact of manure management^{52,53} however, showcasing the influence of trade in stages 2 and 5 exemplifies the importance of considering entire production-consumption chains instead of simply estimating production values for a given watershed.

4. DISCUSSION, LIMITATIONS, AND APPLICATION OF THE NFCBF MODEL

We built the NFCBF based upon previous work on both nitrogen footprint and nitrogen budget, advancing the previous nitrogen footprint models to include multiple aspects of a food production chain beyond consumption. By switching the focus from consumption to production and trade, we enabled the model to determine where along the food production chain the N loss occurs and estimate the N loss externality due to food trade. This multiaspect model is able to quantify the effects of various factors, including trade, dietary choices, production patterns, and agricultural practices, on the watershed environmental loss and could be utilized to evaluate environmental protection scenarios. Although we created the model at watershed and county scales on an annual basis, the model is developed in a way to be applicable and implementable at any spatial and temporal scales that the data are available.

Even though several studies have investigated nitrogen flow in the Chesapeake Bay watershed^{36,38,54} at various scales thus far, they have limited consideration for the effects of food import-export throughout the watershed on the nitrogen loss. We show that without considering the food trade, we end up overestimating the N loss in some regions and underestimating it in some others (Figure S4). Likewise, the results of our study depict that the total nitrogen loss for the whole watershed and at the regional level has decreased when considering commodity trade. Our model's ability to distinguish between nitrogen loss from local (domestic production and consumption) and nonlocal (import and export) sources, making it a potential tool to optimize regional domestic production and trade to meet local watershed's human protein needs while minimizing the resulting nitrogen loss. Beyond the same spatial trends however, our model further incorporates the resolution of showing the surplus variation spatially across each stage of the crop-animal production chain. In addition to considering commodity trade, our model advanced available county-level N budget models^{19,51} by shifting the focus from croplands to the crop-animal system boundary inclusive of trade.

In addition, we have calculated the model for the years 2002, 2007, 2012, and 2017, thus considering time series variability in production, consumption, and trade due to multiple factors like land-use change and production quantity on the temporal differences in N loss. The temporal analysis of the model, along with the spatial analysis, enables us to explore the spatiotemporal drivers of the nitrogen loss. Despite decreases in atmospheric nitrogen deposition onto agricultural land, recent mass balance work has suggested that promising long-term declines in agricultural surplus and improvements in nutrient use efficiency have stagnated or begun to reverse after the mid-2000s, yet it is unclear why the sudden reversal has occurred.⁴⁵ Insights from NFCBF suggest that increased corn and wheat acreage in 2017 (leading to greater fertilizer use) and steadily increasing

livestock/poultry production after 2002 may have led to the reversal/stagnation in decreasing N loss trends from agricultural production observed over the past two decades (Figure 4). Our uncertainty analysis of the model showcases a range of variability of potential values for nitrogen loss due to uncertainty in the model inputs, which has the potential to contribute to understanding tradeoffs in decision making. Overall, our results provide a more intricate view of nitrogen flow within a watershed to show where in the food chain Nr loss is increasing and NUE degrading.

We found that corn grain, wheat grain, poultry-broiler, and milk are the commodities that contribute the most to releasing N loss to the watershed environment. This aligns with expectations since there was a big increase in corn and wheat grain production in 2012 compared to the previous years.⁴⁶ All states in the Chesapeake Bay Watershed, except New York, are also among the top 20 states for producing poultry broilers in the United States.⁵⁵ Specifically, Sussex County (DE) produces the most broiler chickens in the nation by county.⁴⁹ In our uncertainty analysis, we also found that animal feed conversion rates impact nitrogen loss in very significant ways. If the Chesapeake Bay continues to be a large producer of poultry, continued trends in more efficient feed conversion ratios will have a large impact on nitrogen surplus. Also, New York and Pennsylvania are among the top 10 states of cow-milk production in the United States.^{56,57}

This study shows that the NFCBF model is applicable at multiple scales and has the ability to quantify nitrogen flow at each phase of the food production system, and our results allow for determining the regional and temporal variation of nitrogen flow components, nitrogen loss hotspots, spatiotemporal drivers of nitrogen loss, and nitrogen pollution externalities due to trade. The first limitation of this work is that the model is developed for the major commodities in the Bay and not all the commodities. For example, vegetable production is not included in the model. Although we do not expect a great change,⁶ without considering the vegetable production in the model, we expect that the model will underestimate the regional Nr loss. Also, the nitrogen loss estimated in this model is the total nitrogen loss released to the environment, including to the water, air, and soil thus we do not incorporate fate and transport estimates in this work. A potential opportunity for future work is to explore our results to inform nitrogen management and policy and hence restoration of the Bay and its tributaries via determining pathways of Nr loss and evaluating the N loss runoff in the context of the Chesapeake Bay TMDL goals. Also, other potential future work using our model could include running scenario analyses to indicate how policy or land management decisions may influence these flows of embedded nitrogen.

This study's findings showcase that our nitrogen flow model can quantify the nitrogen loss and nitrogen use efficiency in a food chain incorporating trade flux at several scales. The nitrogen model determined the key factors and sources controlling the Nr loss values and regional variation and identified the most affected regions. The model identified that the primary sources of nitrogen pollution to the watershed environment are manure loss and nitrogen fertilizer loss, which contribute the most to releasing nitrogen loss to the watershed. Based on the results, in terms of total nitrogen loss, the most highly affected regions are Lancaster County (PA), Sussex County (DE), and Rockingham County (VA), respectively. The findings also revealed the significant contribution of trade to N pollution reduction in the watershed (40 million tons N). The

noticeable impacts of food trade on the regional nitrogen flow confirm the importance of integrating trade data in our model to inform understanding around reactive nitrogen impacts from local and nonlocal agricultural commodities and food products. Our developed model (NFCBF) could be used as a tool to quantify the effect of the food chain changes on a watershed or a region's environmental pollution and a metric to evaluate environmental protection scenarios.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.est.2c07391>.

Data preparation for the NFCBF model; NFCBF model assumptions; variables, equations, and datasets in the NFCBF model; and NFCBF model results (PDF)

NFCBF model data package including input data and N flow component results (XLSX)

■ AUTHOR INFORMATION

Corresponding Author

Caitlin Grady – *Engineering Management and Systems Engineering, The George Washington University, Washington, District of Columbia 20052, United States*; orcid.org/0000-0002-9151-6664; Email: caitlin.grady@gwu.edu

Author

Paniz Mohammadpour – *Department of Civil and Environmental Engineering and LandscapeU NSF National Research Traineeship (NRT), The Pennsylvania State University, University Park, Pennsylvania 16802, United States*

Complete contact information is available at:

<https://pubs.acs.org/10.1021/acs.est.2c07391>

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We would like to thank Dr. Robert Sabo and the other anonymous reviewers for their detailed critique and feedback of previous drafts of this manuscript. We would also like to thank Drs. Jason Kaye, Cibin Raj, Lisa Wagner, and David Abler for their feedback on this work. This work was supported by U.S. Department of Agriculture NIFA Grant 2019-68012-29904. The views expressed in this article are those of the authors and do not necessarily represent the views or policies of the U.S. Department of Agriculture.

■ REFERENCES

- (1) Dukes, E. S. M.; Galloway, J. N.; Band, L. E.; Cattaneo, L. R.; Groffman, P. M.; Leach, A. M.; Castner, E. A. A Community Nitrogen Footprint Analysis of Baltimore City, Maryland. *Environ. Res. Lett.* **2020**, *15*, No. 075007.
- (2) Fowler, D.; Coyle, M.; Skiba, U.; Sutton, M. A.; Cape, J. N.; Reis, S.; Sheppard, L. J.; Jenkins, A.; Grizzetti, B.; Galloway, J. N.; Vitousek, P.; Leach, A.; Bouwman, A. F.; Butterbach-Bahl, K.; Dentener, F.; Stevenson, D.; Amann, M.; Voss, M. The Global Nitrogen Cycle in the Twenty-First Century. *Phil. Trans. R. Soc. B* **2013**, *368*, No. 20130164.
- (3) Galloway, J. N.; Townsend, A. R.; Erisman, J. W.; Bekunda, M.; Cai, Z.; Freney, J. R.; Martinelli, L. A.; Seitzinger, S. P.; Sutton, M. A. Transformation of the Nitrogen Cycle: Recent Trends, Questions, and Potential Solutions. *Science* **2008**, *320*, 889–892.

- (4) Guo, M.; Chen, X.; Bai, Z.; Jiang, R.; Galloway, J. N.; Leach, A. M.; Cattaneo, L. R.; Oenema, O.; Ma, L.; Zhang, F. How China's Nitrogen Footprint of Food Has Changed from 1961 to 2010. *Environ. Res. Lett.* **2017**, *12*, 104006.
- (5) Pierer, M.; Winiwarter, W.; Leach, A. M.; Galloway, J. N. The Nitrogen Footprint of Food Products and General Consumption Patterns in Austria. *Food Policy* **2014**, *49*, 128–136.
- (6) Leach, A. M.; Galloway, J. N.; Bleeker, A.; Erisman, J. W.; Kohn, R.; Kitzes, J. A Nitrogen Footprint Model to Help Consumers Understand Their Role in Nitrogen Losses to the Environment. *Environ. Dev.* **2012**, *1*, 40–66.
- (7) Lehnert, N.; Dong, H. T.; Harland, J. B.; Hunt, A. P.; White, C. J. Reversing Nitrogen Fixation. *Nat. Rev. Chem.* **2018**, *2*, 278–289.
- (8) Liang, X.; Leach, A. M.; Galloway, J. N.; Gu, B.; Lam, S. K.; Chen, D. Beef and Coal Are Key Drivers of Australia's High Nitrogen Footprint. *Sci. Rep.* **2016**, *6*, 39644.
- (9) Chesapeake Bay Foundation. *Chesapeake Bay Foundation*. <https://www.cbf.org/issues/agriculture/nitrogen-phosphorus.html> (accessed 2021-03-22).
- (10) Erisman, J.; Leach, A.; Bleeker, A.; Atwell, B.; Cattaneo, L.; Galloway, J. An Integrated Approach to a Nitrogen Use Efficiency (NUE) Indicator for the Food Production–Consumption Chain. *Sustainability* **2018**, *10*, 925.
- (11) Daughtry, C. S. T.; Gish, T. J.; Dulaney, W. P.; Walthall, C. L.; Kung, K. J. S.; McCarty, G. W.; Angier, J. T.; Buss, P. Surface and Subsurface Nitrate Flow Pathways on a Watershed Scale. *Sci. World J.* **2001**, *1*, 155–162.
- (12) *Nitrogen Cycling in the North Atlantic Ocean and Its Watersheds*; Howarth, R. W., Ed.; Springer Netherlands: Dordrecht, 1996.
- (13) Westhoek, H.; Lesschen, J. P.; Leip, A.; Rood, T.; Wagner, S.; De Marco, A.; Murphy-Bokern, D.; Pallière, C.; Howard, C. M.; Oenema, O.; Sutton, M. A. *Nitrogen on the Table: The Influence of Food Choices on Nitrogen Emissions and the European environment*; NERC/Centre for Ecology & Hydrology: Edinburgh, UK, 2015.
- (14) Zhang, W.; Li, H.; Li, Y. Spatio-Temporal Dynamics of Nitrogen and Phosphorus Input Budgets in a Global Hotspot of Anthropogenic Inputs. *Sci. Total Environ.* **2019**, *656*, 1108–1120.
- (15) Shibata, H.; Galloway, J. N.; Leach, A. M.; Cattaneo, L. R.; Cattell Noll, L.; Erisman, J. W.; Gu, B.; Liang, X.; Hayashi, K.; Ma, L.; Dalgaard, T.; Graversgaard, M.; Chen, D.; Nansai, K.; Shindo, J.; Matsubae, K.; Oita, A.; Su, M.-C.; Mishima, S.-I.; Bleeker, A. Nitrogen Footprints: Regional Realities and Options to Reduce Nitrogen Loss to the Environment. *Ambio* **2017**, *46*, 129–142.
- (16) Lassaletta, L.; Billen, G.; Romero, E.; Garnier, J.; Aguilera, E. How Changes in Diet and Trade Patterns Have Shaped the N Cycle at the National Scale: Spain (1961–2009). *Reg. Environ. Change* **2014**, *14*, 785–797.
- (17) Lassaletta, L.; Billen, G.; Grizzetti, B.; Garnier, J.; Leach, A. M.; Galloway, J. N. Food and Feed Trade as a Driver in the Global Nitrogen Cycle: 50-Year Trends. *Biogeochemistry* **2014**, *118*, 225–241.
- (18) Oita, A.; Malik, A.; Kanemoto, K.; Geschke, A.; Nishijima, S.; Lenzen, M. Erratum: Substantial Nitrogen Pollution Embedded in International Trade. *Nat. Geosci.* **2016**, *9*, 260–260.
- (19) Byrnes, D. K.; Van Meter, K. J.; Basu, N. B. Long-Term Shifts in U.S. Nitrogen Sources and Sinks Revealed by the New TREND-Nitrogen Data Set (1930–2017). *Global Biogeochem. Cycles* **2020**, *34*, No. e2020GB006626.
- (20) Sabo, R. D.; Clark, C. M.; Bash, J.; Sobota, D.; Cooter, E.; Dobrowolski, J. P.; Houlton, B. Z.; Rea, A.; Schwede, D.; Morford, S. L.; Compton, J. E. Decadal Shift in Nitrogen Inputs and Fluxes across the Contiguous United States: 2002–2012. *J. Geophys. Res.: Biogeosci.* **2019**, *124*, 3104–3124.
- (21) Swaney, D. P.; Howarth, R. W.; Hong, B. County, Subregional and Regional Nitrogen Data Derived from the Net Anthropogenic Nitrogen Inputs (NANI) Toolbox. *Data Br.* **2018**, *18*, 1877–1888.
- (22) Liang, X.; Lam, S. K.; Gu, B.; Galloway, J. N.; Leach, A. M.; Chen, D. Reactive Nitrogen Spatial Intensity (NrSI): A New Indicator for Environmental Sustainability. *Global Environ. Change* **2018**, *52*, 101–107.
- (23) Asmala, E.; Saikku, L.; Vienonen, S. Import–Export Balance of Nitrogen and Phosphorus in Food, Fodder and Fertilizers in the Baltic Sea Drainage Area. *Sci. Total Environ.* **2011**, *409*, 4917–4922.
- (24) Cameira, M. R.; Rolim, J.; Valente, F.; Faro, A.; Dragosits, U.; Cordovil, C. M. d. S. Spatial Distribution and Uncertainties of Nitrogen Budgets for Agriculture in the Tagus River Basin in Portugal – Implications for Effectiveness of Mitigation Measures. *Land Use Policy* **2019**, *84*, 278–293.
- (25) Dalgaard, T.; Bienkowski, J. F.; Bleeker, A.; Dragosits, U.; Drouet, J. L.; Durand, P.; Frumau, A.; Hutchings, N. J.; Kedziora, A.; Magliulo, V.; Olesen, J. E.; Theobald, M. R.; Maury, O.; Akkal, N.; Cellier, P. Farm Nitrogen Balances in Six European Landscapes as an Indicator for Nitrogen Losses and Basis for Improved Management. *Biogeosciences* **2012**, *9*, 5303–5321.
- (26) Elrys, A. S.; Raza, S.; Abdo, A. I.; Liu, Z.; Chen, Z.; Zhou, J. Budgeting Nitrogen Flows and the Food Nitrogen Footprint of Egypt during the Past Half Century: Challenges and Opportunities. *Environ. Int.* **2019**, *130*, 104895.
- (27) Leip, A.; Britz, W.; Weiss, F.; de Vries, W. Farm, Land, and Soil Nitrogen Budgets for Agriculture in Europe Calculated with CAPRI. *Environ. Pollut.* **2011**, *159*, 3243–3253.
- (28) Oenema, O. Nitrogen Budgets and Losses in Livestock Systems. *Int. Cong. Ser.* **2006**, *1293*, 262–271.
- (29) Lassaletta, L.; Romero, E.; Billen, G.; Garnier, J.; García-Gómez, H.; Rovira, J. V. Spatialized N Budgets in a Large Agricultural Mediterranean Watershed: High Loading and Low Transfer. *Biogeosciences* **2012**, *9*, 57–70.
- (30) Gu, B.; Leach, A. M.; Ma, L.; Galloway, J. N.; Chang, S. X.; Ge, Y.; Chang, J. Nitrogen Footprint in China: Food, Energy, and Nonfood Goods. *Environ. Sci. Technol.* **2013**, *47*, 9217–9224.
- (31) Serra, J.; Cordovil, C. M. d. S.; Cruz, S.; Cameira, M. R.; Hutchings, N. J. Challenges and Solutions in Identifying Agricultural Pollution Hotspots Using Gross Nitrogen Balances. *Agric. Ecosyst. Environ.* **2019**, *283*, No. 106568.
- (32) Gao, B.; Wang, L.; Cai, Z.; Huang, W.; Huang, Y.; Cui, S. Spatio-Temporal Dynamics of Nitrogen Use Efficiencies in the Chinese Food System, 1990–2017. *Sci. Total Environ.* **2020**, *717*, No. 134861.
- (33) Godinot, O.; Leterme, P.; Vertès, F.; Carof, M. Indicators to Evaluate Agricultural Nitrogen Efficiency of the 27 Member States of the European Union. *Ecol. Indic.* **2016**, *66*, 612–622.
- (34) Ma, L.; Ma, W. Q.; Velthof, G. L.; Wang, F. H.; Qin, W.; Zhang, F. S.; Oenema, O. Modeling Nutrient Flows in the Food Chain of China. *J. Environ. Qual.* **2010**, *39*, 1279–1289.
- (35) Wang, F.; Wang, Y.; Cai, Z.; Chen, X. Environmental Losses and Driving Forces of Nitrogen Flow in Two Agricultural Towns of Hebei Province during 1997–2017. *Environ. Pollut.* **2020**, *264*, No. 114636.
- (36) Ator, S. W.; García, A. M.; Schwarz, G. E.; Blomquist, J. D.; Sekellick, A. J. Toward Explaining Nitrogen and Phosphorus Trends in Chesapeake Bay Tributaries, 1992–2012. *J. Am. Water Resour. Assoc.* **2019**, *55*, 1149–1168.
- (37) Fox, R. H.; Zhu, Y.; Toth, J. D.; Jemison, J. M.; Jabro, J. D. Nitrogen Fertilizer Rate and Crop Management Effects on Nitrate Leaching from an Agricultural Field in Central Pennsylvania. *Sci. World J.* **2001**, *1*, 181–186.
- (38) Kaufman, Z.; Abler, D.; Shortle, J.; Harper, J.; Hamlett, J.; Feather, P. Agricultural Costs of the Chesapeake Bay Total Maximum Daily Load. *Environ. Sci. Technol.* **2014**, *48*, 14131–14138.
- (39) Tuppad, P.; Kannan, N.; Srinivasan, R.; Rossi, C. G.; Arnold, J. G. Simulation of Agricultural Management Alternatives for Watershed Protection. *Water Resour. Manage.* **2010**, *24*, 3115–3144.
- (40) Cherry, K. A.; Shepherd, M.; Withers, P. J. A.; Mooney, S. J. Assessing the Effectiveness of Actions to Mitigate Nutrient Loss from Agriculture: A Review of Methods. *Sci. Total Environ.* **2008**, *406*, 1–23.
- (41) McDowell, R. W.; Withers, P. J.; van der Weerden, T. J. The Environmental Impact of Fertiliser Nutrients on Freshwater. In *Agricultural Chemicals and the Environment*; Royal Society of Chemistry, 2016; pp. 20–44.
- (42) Patterson, J. J.; Smith, C.; Bellamy, J. Understanding Enabling Capacities for Managing the ‘Wicked Problem’ of Nonpoint Source

Water Pollution in Catchments: A Conceptual Framework. *J. Environ. Manage.* **2013**, *128*, 441–452.

(43) Schoumans, O. F.; Chardon, W. J.; Bechmann, M. E.; Gascuel-Oudou, C.; Hofman, G.; Kronvang, B.; Rubæk, G. H.; Ulén, B.; Dorioz, J.-M. Mitigation Options to Reduce Phosphorus Losses from the Agricultural Sector and Improve Surface Water Quality: A Review. *Sci. Total Environ.* **2014**, *468-469*, 1255–1266.

(44) EPA Chapter 2. Agriculture. In *Guidance for Federal Land Management in the Chesapeake Bay Watershed*; EPA: 2010, p. 247.

(45) Sabo, R. D.; Sullivan, B.; Wu, C.; Trentacoste, E.; Zhang, Q.; Shenk, G. W.; Bhatt, G.; Linker, L. C. Major Point and Nonpoint Sources of Nutrient Pollution to Surface Water Have Declined throughout the Chesapeake Bay Watershed. *Environ. Res. Commun.* **2022**, *4*, No. 045012.

(46) USDA-NASS. *USDA/NASS QuickStats Ad-hoc Query Tool*. <https://quickstats.nass.usda.gov/> (accessed 2021-05-18).

(47) Hwang, H. L.; Hargrove, S.; Chin, S. M.; Wilson, D.; Lim, H.; Chen, J.; Taylor, R.; Peterson, B.; Davidson, D. *The Freight Analysis Framework Version 4 (FAF4) - Building the FAF4 Regional Database: Data Sources and Estimation Methodologies (Technical Report)* OSTI.GOV. <https://www.osti.gov/biblio/1325489> (accessed 2021-04-19).

(48) Lin, X.; Ruess, P. J.; Marston, L.; Konar, M. Food Flows between Counties in the United States. *Environ. Res. Lett.* **2019**, *14*, No. 084011.

(49) USDA-NASS. *COA County Profile: Sussex County Delaware*. https://www.nass.usda.gov/Publications/AgCensus/2017/Online_Resources/County_Profiles/Delaware/cp10005.pdf (accessed 2021-08-09).

(50) Hong, B.; Swaney, D. P.; Howarth, R. W. A Toolbox for Calculating Net Anthropogenic Nitrogen Inputs (NANI). *Environ. Model. Softw.* **2011**, *26*, 623–633.

(51) MAWP. *Mid-Atlantic Water Program. 2007. Nutrient budgets for the Mid-Atlantic states*. <http://www.mawaterquality.agecon.vt.edu/> (accessed 2010-02-21).

(52) Kast, J. B.; Long, C. M.; Muenich, R. L.; Martin, J. F.; Kalcic, M. M. Manure Management at Ohio Confined Animal Feeding Facilities in the Maumee River Watershed. *J. Great Lakes Res.* **2019**, *45*, 1162–1170.

(53) Savage, J. A.; Ribaud, M. O. Impact of Environmental Policies on the Adoption of Manure Management Practices in the Chesapeake Bay Watershed. *J. Environ. Manage.* **2013**, *129*, 143–148.

(54) Russell, K. M.; Galloway, J. N.; Macko, S. A.; Moody, J. L.; Scudlark, J. R. Sources of Nitrogen in Wet Deposition to the Chesapeake Bay Region. *Atmos. Environ.* **1998**, *32*, 2453–2465.

(55) USDA-NASS. *USDA - National Agricultural Statistics Service - Charts and Maps - Broilers: Inventory by State, US*. https://www.nass.usda.gov/Charts_and_Maps/Poultry/brlmap.php (accessed 2021-10-04).

(56) USDA-NASS. *COA State Profile: New York*. https://www.nass.usda.gov/Publications/AgCensus/2017/Online_Resources/County_Profiles/New_York/cp99036.pdf (accessed 2021-10-04).

(57) USDA-NASS. *COA State Profile: Pennsylvania*. https://www.nass.usda.gov/Publications/AgCensus/2017/Online_Resources/County_Profiles/Pennsylvania/cp99042.pdf (accessed 2021-10-04).