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Water Supply Processes Are Responsible for Significant Nitrogen Fluxes Across the United States

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Abstract Excessive nutrient concentrations within fresh waters are a globally persistent problem. Developing effective nutrient management strategies requires improvements to nitrogen (N) mass balances, including the identification and quantification of previously unrecognized anthropogenic N fluxes. Using publicly available data, we establish that freshwater abstractions from both surface waters and groundwaters, alongside watermains leakage from public distribution networks, are responsible for significant nitrate-N (NO_3-N) fluxes across the contiguous United States. Nationally, freshwater abstraction temporarily retains 417 (min-max: 190–857) kt NO_3-N yr^{-1}, equivalent to 21% of pastureland N uptake and 2% of previous global abstraction-N flux estimates. Fluxes due to irrigation, thermoelectric power, and public water supply collectively account for 87% of this total. We find large intercounty variation in area-normalized abstraction fluxes (min-max: 0–8,267 kg NO_3-N km^{-2} yr^{-1}), with eastern regions generally associated with larger fluxes. Watermains leakage returns 7 (min-max: 6.3–7.7) kt NO_3-N yr^{-1} back to the environment, equivalent to 13% of NO_3-N initially abstracted for public supply and 1.3% of previous global leakage flux estimates. Our analyses reveal inter-county variations in area-normalized leakage fluxes (min-max: 0–576 kg NO_3-N km^{-2} yr^{-1}), with this flux exceeding other major N inputs (agricultural N fertilizer) in some urbanized and coastal counties, highlighting their importance in these areas. The local and national importance of these fluxes has implications for policy makers and water resource managers aiming to better manage the impacts of N within the environment and calls for their inclusion in both US and global N budgets.

Key Points:

- US freshwater abstractions temporarily retain 417 kt of nitrogen per year, equivalent to 57% of US river denitrification and 2% of global abstraction estimates
- Watermains leakage returns a significant mass of nitrogen to the environment across many coastal and urban US counties, particularly in the northeast
- The importance of water supply processes to nitrogen cycling is highlighted, with the methodology developed suitable for use around the globe

Supporting Information:

Supporting Information may be found in the online version of this article.

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1. Introduction

Nitrogen (N) plays a crucial role in the metabolic functioning of biological systems and thus global food production (Fields, 2004; Fowler et al., 2013a). Anthropogenic activity, in particular the unsustainable application of fertilizer, has caused global biogeochemical cycles of N to exceed safe planetary boundaries (Steffen et al., 2015). The pollution of fresh waters due to excess nitrate (NO_3) concentrations has led to significant, adverse effects on human and environmental health around the globe. These include severe eutrophication of inland and coastal waters (Bijay & Craswell, 2021; Howarth, 2008; McDowell et al., 2020) and decreases in potable water quality (Camargo & Alonso, 2006; Mekonnen & Hoekstra, 2015). Human influences on the N cycle have been pervasive across the United States (US), with the country's anthropogenic production of reactive N five times that of its natural production (EPA Science Advisory Board, 2011). Subsequent nutrient pollution is now a leading cause of degraded freshwater quality across the country (USEPA, 2015) and the effects on environmental and human health are widespread (Bouwman et al., 2013; Munn et al., 2018). Eutrophication of inland and coastal waterbodies has persisted for decades in the United States (Bricker et al., 2008; Oelsner & Stets, 2019; Oswald & Golueke, 1966), and the decrease in potable (Kozacek, 2014) and recreational water quality (World Health Organization, 1999), loss of aquatic habitats, and disruption to food chains (Munn et al., 2018) are estimated to cost billions of dollars a year (Dodds et al., 2009; Sobota et al., 2015).

Since the Clean Water Act was brought into practice (USEPA, 1972), overall improvements to US freshwater quality have been slow, and elevated N concentrations continue to contribute to persistent poor water quality across the country (Keiser et al., 2019; Manuel, 2014; USEPA, 2010). The sustainable and effective use of nutrients requires an integrated management approach, meaning that nutrient balances for a given system must identify and regularly quantify factors influencing all known inputs, transformations, and outputs (EPA Science Advisory Board, 2011; Grizzetti et al., 2015; Roy et al., 2021; Sabo et al., 2019; Wu & Ma, 2015). Delays in freshwater quality improvements reflect the response time of the system (Meals & Dressing, 2008) and associated legacy

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stores of N in the soil, vadose zone and groundwater (Ascott et al., 2017; Chang et al., 2021). However, estimates of these processes often carry large uncertainty, and gaps in our knowledge of both established and emerging anthropogenic influences on N cycling highlight the need for improved N mass balances (Baron et al., 2012; Fowler et al., 2013b; McGrane, 2016; Sobota et al., 2013).

Research has begun to highlight the potential role that water supply processes play in N cycling both in the United States and around the globe, including their effect on N retention, defined as processes that prevent or delay the delivery of N from the land to the oceans (Grizzetti et al., 2015). For example, the manipulation of water resources through dam construction has been found to have an effect on major retention processes of denitrification and burial of N in sediments both across the United States (Baron et al., 2012) and around the world (Beusen et al., 2016; Seitzinger et al., 2007; Wisser et al., 2010). Recent research has now found groundwater pumping to perturb global N cycles (Stahl, 2019), and research in England, UK, has shown that abstraction of fresh water for public supply acts as a nationally significant temporary retention mechanism for organic N (Finlay et al., 2016) and NO_3-N (Ascott, Gooddy, & Surridge, 2018), highlighting the need for the inclusion of these fluxes in future nutrient budgets.

The United States has one of the largest freshwater abstraction volumes per capita in the world (FAO, 2021). Freshwater use sectors are defined by the United States Geological Survey (USGS), and sectoral withdrawal volumes are estimated every 5 years (Dieter, Maupin, et al., 2018). Whilst approximately 14% of the country's total freshwater abstraction was for public supply (water withdrawn by both public and private suppliers), the remainder of abstractions were for nonpublic and self-supplied uses including for irrigation, thermoelectric, industry, livestock, aquaculture, domestic (i.e., private wells), and mining uses (Dieter, Maupin, et al., 2018).

Although leakage of water from the mains distribution network has long been known to return nutrients to the environment (Holman et al., 2008; Lerner et al., 1999; Wakida & Lerner, 2005), only recently has research begun to both quantify these fluxes at the scale of a given country (Ascott et al., 2016, 2018b; Gooddy et al., 2017) and investigate how to identify these fluxes isotopically (Gooddy et al., 2015). Results from a study examining public water supply in England, UK, have highlighted the significance of these fluxes in urban areas, with watermains leakage fluxes equivalent to up to 20% of all N sources in some areas (Ascott, Gooddy, & Surridge, 2018), and thus the need for their incorporation into nutrient budgets and consideration in policy decision making. The declining condition of water infrastructure across the United States means watermains leakage is a persistent yet spatially variable problem (Folkman, 2018; Rosario-Ortiz et al., 2016) and results in the loss of an average of 16% of the water initially entering the country's watermains distribution network (USEPA, 2013).

In this research, we use the United States as an exemplar to quantify NO_3-N fluxes associated with nonpublic supply abstraction for the first time globally. We also quantify public supply NO_3-N abstraction fluxes and watermains leakage NO_3-N fluxes for the first time within the US. We hypothesize that:

1. Freshwater abstraction for all uses constitutes a significant temporary NO_3-N retention process across the United States and makes a significant contribution to global abstraction NO_3-N flux estimates
2. Across the US, watermains leakage returns a spatially heterogeneous and often locally significant flux of NO_3-N back to the environment

We used publicly available abstraction and watermains leakage volumetric rate data, along with untreated (raw) and treated water NO_3-N concentrations, to quantify fluxes of NO_3-N associated with freshwater abstractions and watermains leakage across the contiguous United States. The environmental significance of these county, state, and national-scale estimates were evaluated through their comparison with other estimates for components of the N budget. We also discuss the implications of our findings in both a national and global context and make recommendations for future research.

2. Methods

2.1. Quantification of National Abstraction and Watermains Leakage NO_3-N Fluxes

County-level fluxes of NO_3-N associated with freshwater abstractions for public water supply (PWS-NO_3-N) and self-supplied irrigation (IRR-NO_3-N), thermoelectric power (THERM-NO_3-N), industry (IND-NO_3-N), mining (MINE-NO_3-N), livestock (LIVE-NO_3-N), aquaculture (AQUA-NO_3-N), and domestic (DOMESTIC-NO_3-N)

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water use sectors were estimated across the 48 states that form the contiguous United States (herein referred to simply as the United States) and aggregated to give a final abstraction flux (ABS-NO_3-N) using Equation 1.

$$\text{ABS-NO}_3\text{-N} = \left(\text{WD}_{(\text{SW})} \text{ x } \text{C}_{\text{r (SW-NO}_3\text{-N)}}\right) + \left(\text{WD}_{(\text{GW})} \text{ x } \text{C}_{\text{r (GW-NO}_3\text{-N)}}\right) \quad (1)$$

Surface water ($WD_{(SW)}$) and groundwater ($WD_{(GW)}$) withdrawal (i.e., abstraction) data, in L yr^{-1}, were provided for each water use sector within each county for the year of 2015 by the USGS (Dieter, Linsey, et al., 2018), representing the most up-to-date abstraction estimates across the US. Raw surface water NO_3-N concentrations ($C_{r\,(SW-NO_3-N)}$), in mg L^{-1}, were sourced from the USGS NWIS (USGS, 2021) and the EPA's STORET-WQX (USEPA, 2021b) databases (see Text S1 in Supporting Information S1). Raw groundwater NO_3-N concentrations ($C_{r\,(GW-NO_3-N)}$), in mg L^{-1}, were sourced from USGS NAWQA studies (Arnold et al., 2016, 2017, 2018, 2020), In order to increase the number of data points, $C_{r\,(SW-NO_3-N)}$ and $C_{r\,(GW-NO_3-N)}$ values were obtained between 2010–2020 and 2012–2016, respectively. Despite expanding C_r concentration data sets to additional years surrounding 2015, the lack of comprehensive county-level data sets meant that state-level median values of both $C_{r\,(SW-NO_3-N)}$ and $C_{r\,(GW-NO_3-N)}$ were assigned to that state's constituent counties (see Text S1 in Supporting Information S1). Sector-specific $C_{r(GW-NO_3-N)}$ concentration values were also only available for determination of PWS-NO_3-N and DOMESTIC-NO_3-N fluxes (Figures S2 and S3 in Supporting Information S1, respectively), resulting in the remaining $C_{r(GW-NO_3-N)}$ values and all $C_{r\,(SW-NO_3-N)}$ concentrations being uniform across all other sectors (Figures S4 and S1 in Supporting Information S1, respectively). These limitations are discussed further in Section 4.1. The resulting concentration data sets were non-normally distributed (Shapiro-Wilk's test $p < 0.01$), so median concentrations for each state were assigned to their comprising counties and used within Equation 1. Estimated county fluxes for each water use sector were aggregated to give total county, state, and national ABS-NO_3-N in units of metric kt NO_3-N yr^{-1}. County and state-level estimates were also normalized for their land area and expressed in units of kg NO_3-N km^{-2} yr^{-1}, allowing counties and states of contrasting area to be compared.

County-level NO_3-N fluxes due to watermains leakage (WML-NO_3-N) were derived using Equations 2 and 3. Estimated volumetric leakage rates have not been reported by the USGS since 1995 (Dieter, Maupin, et al., 2018), thus volumetric leakage rates for each county (LV), in L yr^{-1}, were calculated using Equation 2. With public supply distribution input estimates also omitted from water use reports, and given that the majority of losses are from pipes downstream of the water treatment works (Van Hecke, 2020), distribution inputs were assumed to equal the total volume of freshwater abstracted for PWS in each county ($WD_{(PWS\text{-}TOTAL)}$), in L yr^{-1}. Freshwater abstractions for PWS (assumed to be distribution inputs) are reported by the USGS (Dieter, Linsey, et al., 2018). These estimates include unaccounted for water, including system losses (i.e., leakage) and so must be corrected using a leakage factor. With the exception of California and Georgia (see Section 2.2), a lack of county-level leakage factor data meant that state-level factors, f_{state} (unitless), were obtained from various sources (see Text S1 in Supporting Information S1) and assigned to their respective counties. In the absence of an f_{state} value, the national average of 0.16 was used. Due to the absence of county-level data, average treated water NO_3-N concentrations ($C_{t-NO3-N}$) for each state between 2014 and 2019, in mg L^{-1}, sourced from the Environmental Working Group database (EWG, 2019), were assigned to all counties within that state. County fluxes were aggregated to give state and national-level WML-NO_3-N estimates and were normalized for land area and expressed in units of kg NO_3-N km^{-2} yr^{-1}.

$$\text{LV} = \text{WD}_{(\text{PWS-TOTAL})} \text{ x } f_{\text{state}} \quad (2)$$

$$\text{WML-NO}_3\text{-N} = \text{C}_{\text{t (NO}_3\text{-N)}} \text{ x LV} \quad (3)$$

2.2. ABS-NO_3-N and WML-NO_3-N Flux Method Validation

Due to greater availability of $C_{r\,(GW-NO3-N)}$ data in California (Jurgens et al., 2021), ABS-NO_3-N fluxes were calculated separately for this state, allowing for an estimate derived from data at greater spatial resolution to be compared with the national-scale method detailed in Section 2.1. Utility-level LV values for the year 2016 and their corresponding $C_{t-NO3-N}$ values were available in both California and Georgia (California Department of Water Resources, 2019; EWG, 2019; Georgia EPD, 2016), allowing utility WML-NO_3-N fluxes to be estimated and aggregated to give final county-level estimates. Derived from the most credible source of volumetric leakage rate and treated water concentration data sets, these fluxes were then compared to second state estimates made using the most assumptive method used in this paper, whereby the national average leakage factor of 0.16 was applied to estimated state-level public supply withdrawals to give LV, and C_t was assigned as the respective average state values. This allowed the effect of these assumptions upon WML-NO_3-N estimates to be evaluated.

2.3. National ABS-NO_3-N and WML-NO_3-N Flux Uncertainties

Uncertainty associated with the national ABS-NO_3-N flux estimate was determined by aggregating upper and lower state-level ABS-NO_3-N flux estimates. These were determined using Equation 1, where abstraction volumes were adjusted for ±10%, and concentrations at the 75th and 25th percentile were applied to each state. The inherent uncertainty associated with USGS abstraction data is currently not reported (National Research, 2002). Consumptive use has an estimated uncertainty of 25% (Maupin & Weakland, 2009), though we expect this represents an absolute upper-bound on water use estimate uncertainties, given that consumptive water uses are computed as residuals (National Research, 2002). Thus, we assume an uncertainty of 10% for all nonconsumptive water use (withdrawal) values. Raw surface water and groundwater concentrations at the 75th and 25th percentiles were chosen to remove outliers from uncertainty calculations. Due to a single mean $C_{t-NO3-N}$ value reported for each state by the EWG, uncertainty associated with the national WML-NO_3-N flux is represented by upper and lower estimates made solely through adjustment of leakage volumes for ±10% within Equation 3, using the same rationale as for ABS-NO_3-N uncertainties. Lower and upper uncertainty bounds surrounding national ABS-NO_3-N and WML-NO_3-N estimates are reported in parentheses after flux values.

2.4. Comparison of ABS-NO_3-N and WML-NO_3-N Fluxes With Other N Fluxes

We conceptualize national-scale ABS-NO_3-N and WML-NO_3-N fluxes to be internal within national-scale N budgets. The significance of national ABS-NO_3-N fluxes should therefore, ideally, be determined through its comparison to other national-level internal N retention flux estimates. Such estimates are scant across the US. In this research, we therefore compare ABS-NO_3-N to both a previously published estimate of a national scale internal retention flux (total pastureland N uptake (Byrnes et al., 2020)), and external N fluxes at the system boundary (total denitrification from US rivers (Baron et al., 2012)). We also compare the national-level ABS-NO_3-N estimate to an initial global ABS-NO_3-N flux estimate (Ascott, Gooddy, & Surridge, 2018).

We also conceptualize California's total ABS-NO_3-N estimate as an internal flux of N within state-scale budgets. Due to the total annual change in internal N storage within the California's surface waters and groundwaters being calculated as the difference between all major inputs and outputs of N to the state's surface waters and groundwaters by Liptzin and Dahlgren (2016), we estimate the proportional contribution that our California ABS-NO_3-N flux could make to this value. We also compare the state's ABS-NO flux to external N retention fluxes at the system boundary (surface water N exports to the ocean and total denitrification within surface waters and groundwaters (Liptzin & Dahlgren, 2016)). The state's IRR-NO_3-N flux was compared to a previous irrigation abstraction NO_3-N flux estimate (Liptzin & Dahlgren, 2016).

The relative importance of the national WML-NO_3-N flux was evaluated by its comparison to other national-level and internal N input flux estimates, including: N leached to groundwaters from septic tanks (Sobota et al., 2013), total N input to streams from point sources (Skinner & Wise, 2019), and an initial global WML-NO_3-N estimate (Ascott, Gooddy, & Surridge, 2018). National and county-level WML-NO_3-N fluxes were also compared to the amount of N applied to land as agricultural fertilizer that is then leached from the soil to groundwater. Due to the absence of a published gridded soil N leaching data set for the US, we corrected agricultural N fertilizer application values from Swaney et al. (2018a) with a fixed leaching emission factor of 0.18 (Mekonnen & Hoekstra, 2015). Assigning a single leaching emission factor, which ignores its large variability around the globe, has a large associated uncertainty (Bijay & Craswell, 2021; Goulding et al., 2000; Wang et al., 2019; Zhou & Butterbach-Bahl, 2014). California's WML-NO_3-N flux was compared to estimates of N leaching from agricultural fertilizer input (Swaney et al., 2018a), urban land, leaking sewers and wastewater to groundwater (Liptzin & Dahlgren, 2016), and N input into streams from point sources (Skinner & Wise, 2019).

Table 1
Total ABS-NO_3-N Fluxes for the United States and California, Compared to Previously Determined N Retention and Export Fluxes

Flux	Reference	Value (kt NO_3-N yr^{-1})[a]	ABS-NO_3-N/flux (%)
National			
National-level ABS-NO_3-N	This study	417 (0–8,267)	–
National-level total denitrification within US rivers	Baron et al. (2012)	730	57
National-level pastureland N uptake	Byrnes et al. (2020)	2,016	21
Global-level ABS-NO_3-N	Ascott, Gooddy, and Surridge (2018)	22,600	2.0
California			
State-level ABS-NO_3-N	This study	38	–
State-level ABS-NO_3-N	This study [b]	39	–
Total state-level N export from rivers to ocean	Liptzin and Dahlgren (2016)	39	97
Total state-level denitrification of N within SW and GW	Liptzin and Dahlgren (2016)	121	31
Change in state-level N storage within GW and SW	Liptzin and Dahlgren (2016)	331	12

[a]Values in parentheses are minimum and maximum county flux values, in units of kg km^{-2} yr^{-1}. [b]This estimate was made using the national-level data set.

3. Results

3.1. ABS-NO_3-N

The total freshwater abstraction flux (ABS-NO_3-N) for the United States is estimated as 417 (190–857) kt NO_3-N yr^{-1} (Table 1). Contributions of each water use sector to the total ABS-NO_3-N flux are 44% from irrigation, 29% from thermoelectric, 13% from public water supply, 6% from industry, 4% from domestic, and 3% combined from aquaculture, livestock, and mining (Figure 2b). Total area-normalized county fluxes range from 0 to 8,267 kg NO_3-N km^{-2} yr^{-1} (Figures 1a and 1b). Counties within coastal states such as California, southern Louisiana and the northeastern states of Massachusetts, Delaware, Maryland, and Virginia are associated with the larger fluxes, as well as counties within the interior states of Idaho and Nebraska (Figures 1 and 2a). The sectoral contributions to state-level ABS-NO_3-N fluxes vary geographically, with irrigation (IRR-NO_3-N) and thermoelectric (TE-NO_3-N) fluxes dominating from western to eastern regions, respectively, and public water supply (PWS-NO_3-N) and self-supplied domestic (DOMESTIC-NO_3-N) fluxes dominating northeastern states (Figure 2b). The ABS-NO_3-N flux estimate for the United States is equivalent to 57% of estimated total denitrification within the nation's rivers, 21% of pastureland N uptake, and 2% of the estimated global ABS-NO_3N flux (Table 1). California's total ABS-NO_3-N estimate of 38 kt NO_3-N yr^{-1} is equivalent to 97% of the export of N to the ocean by rivers, 31% of denitrification within the states surface waters and groundwaters, and 12% of total change in state-wide N storage internally within surface waters and groundwaters (Table 1). The California estimate made using the national data set is in close agreement with the California estimate made using the more comprehensive state-level data set (Table 1). Our California IRR-NO_3-N estimate is 29 kt NO_3-N yr^{-1}, equivalent to 70% of the previous estimate (41 kt NO_3-N yr^{-1}) for 2005 reported by Liptzin and Dahlgren (2016).

3.2. WML-NO_3-N

The total watermains leakage (WML-NO_3-N) flux for the United States is estimated as 7 (6.3–7.7) kt NO_3-N yr^{-1}, returning 13% of PWS-NO_3-N back to the environment (Table 2). Area-normalized county WML-NO_3-N fluxes range from 0 to 576 kg km^{-2} yr^{-1}, with a general trend of increasing fluxes from west to east and the highest values observed in many urbanized counties (Figure 3). The national WML-NO_3-N flux is equivalent to 16% of N leached from nonagricultural fertilizer application, 4% of N leached from septic tanks, 1.4% of total N from point sources, 0.3% of N leached from agricultural fertilizer input, and 1.3% of the global WML-NO_3-N flux estimate (Table 2). There is also large inter-county variability in the relative importance of WML-NO_3-N fluxes, with this flux in many northeast and western coastal counties equivalent to >10% of N inputs from leaching of agricultural fertilizer, with some county WML-NO_3-N fluxes even exceeding this input (Figure 4).

The WML-NO_3-N estimate for Georgia made using both the utility-level data set and that made by adopting state averages (see Section 2.2) are extremely close (Table 2). In contrast, California's estimate made using

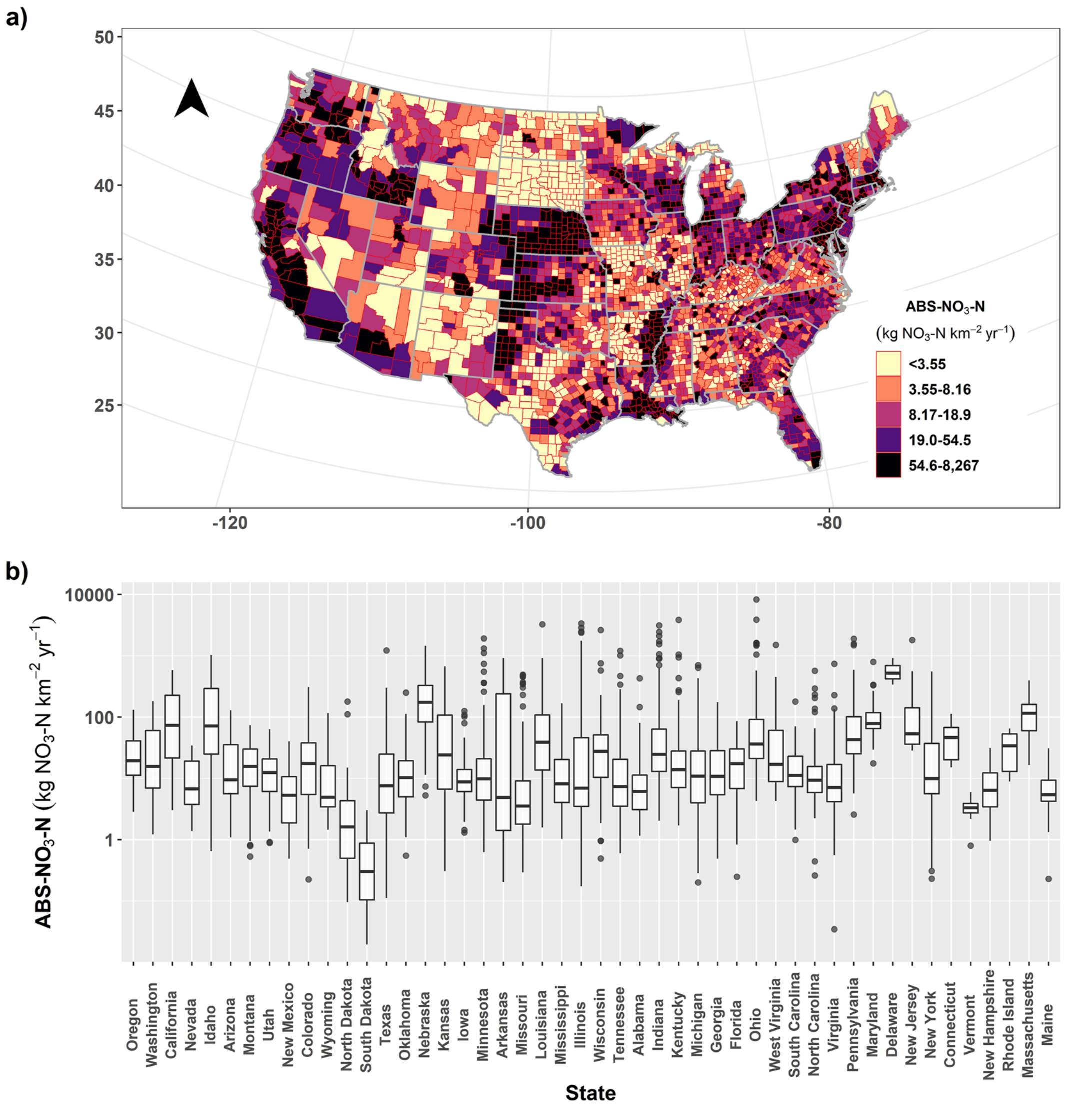

Figure 1. (a) Total area-normalized ABS-NO_3-N fluxes for each county across the United States (US). Breaks in the color scale are defined as separate quintile groups. Linework was created using the 'usmap' package on R (Di Lorenzo, 2022). (b) Boxplot showing the distribution of county-level ABS-NO_3-N flux values across each US state. States are ordered from west to east.

the state-average method is nearly three times as large as that made using the more local utility-level data set (Table 2). There is inter-county variation in area-normalized WML-NO_3-N fluxes within both states (Figure 5). Urban counties in both California and Georgia (e.g., San Francisco and Fulton, respectively) are associated with larger estimated fluxes than more rural counties (e.g., Alpine and Burke, respectively) (Figures 5a and 5b, respectively).

California's total WML-NO_3-N flux is equivalent to 76% of N leached to groundwater from urban soils, 5% of N leached to groundwater from leaking sewers, 2.8% of N leached to groundwater from treated wastewater, 2.3% of total N released to rivers from point source facilities, and 0.9% of N leached to groundwater from agricultural fertilizer inputs across the state (Table 2). There is also significant inter-county variation in the importance of these fluxes relative to leached N and PWS-NO_3-N abstraction fluxes across both states (Figure 6).

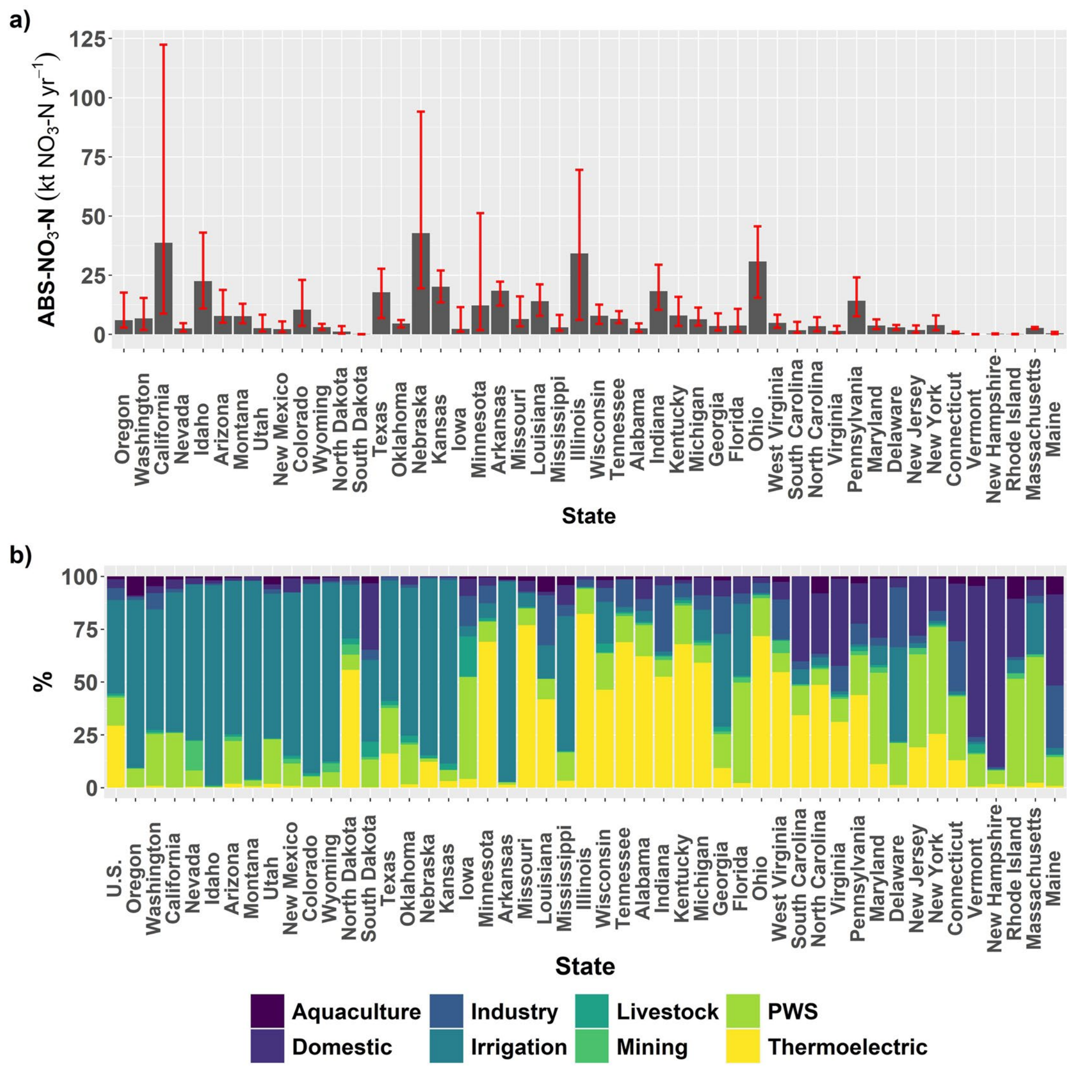

Figure 2. (a) Total state ABS-NO_3-N fluxes across the United States. States on the *x*-axis are ordered from west to east and error bars represent uncertainties (see Text S1 in Supporting Information S1) on state fluxes. (b) Percentage contribution of each water use sector to the national and state ABS-NO_3-N totals.

4. Discussion

4.1. Controls Upon ABS-NO_3-N Fluxes

Retention processes control both the amount of N within inland fresh waters and the amount exported downstream to coastal environments (Baron et al., 2012; Saunders & Kalff, 2001). Although the indirect effects of an altered hydrological cycle as a result of water abstractions have been reported (Lange et al., 2019), our research has quantified the significance of freshwater abstractions for both public and nonpublic supply uses as a NO_3-N retention mechanism for the first time. These ABS-NO_3-N fluxes have largely been neglected in N cycling research, and this research begins to address specific calls for an increased understanding of these fluxes globally (Ascott, Gooddy, & Surridge, 2018; Stahl, 2019). Whilst freshwater withdrawals in the United States account for approximately 10% of global freshwater withdrawals (FAO, 2021), our ABS-NO_3-N estimate accounts for only 2% of initial global ABS-NO_3-N estimates (Table 1). This is likely due to relatively low NO_3-N concentrations within raw surface waters and groundwaters across the United States when compared to other countries that make globally significant freshwater withdrawals, such as India (FAO, 2021; Zhou, 2015). Despite the small contribution from the United States to global ABS-NO_3-N estimates, the transferrable methodology presented here will allow this flux to be estimated in other countries, thus further resolving the global ABS-NO_3-N estimate.

Table 2
Total WML-NO_3-N Fluxes Compared to Estimates of PWS-NO_3-N, Global WML-NO_3-N and Other N Sources for the United States and States of California and Georgia

Flux	Reference	Value (kt NO_3-N yr^{-1})[a]	WML-NO_3-N/flux (%)
National			
National-level WML-NO_3-N	This study	7.0 (0–576)	–
National-level PWS-NO_3-N	This study	55.5	13
National-level N leached from nonagricultural fertilizer input	Swaney et al. (2018a) [b]	44.6	16
National-level N leached from septic tanks	Sobota et al. (2013)	200	4.0
National-level N from point source facilities	Skinner and Wise (2019)	503	1.4
National-level N leached from agricultural fertilizer input	Swaney et al. (2018a) [b]	2,155	0.3
Global-level WML-NO_3-N	Ascott, Gooddy, and Surridge (2018)	525	1.3
California			
State-level WML-NO_3-N	This study	0.758 (1×10^{-4}–65)	–
State-level WML-NO_3-N[c]	This study	2.1	36
State-level N leached to groundwater from urban land	Liptzin and Dahlgren (2016)	1.0	76
State-level N to groundwater from leaking sewers	Liptzin and Dahlgren (2016)	15	5.0
State-level N leached to groundwater from treated wastewater	Liptzin and Dahlgren (2016)	27	2.8
State-level N to rivers from point source facilities	Skinner and Wise (2019)	33.3	2.3
State-level N leached to groundwater from agricultural fertilizer input	Swaney et al. (2018a) [a]	88	0.9
Georgia			
State-level WML-NO_3-N	This study	0.110	–
State-level WML-NO_3-N[c]	This study	0.108	–

[a]Values in parentheses are minimum and maximum county flux values, in units of kg km^{-2} yr^{-1}. [b]Input of N from agricultural and nonagricultural fertilizer were for corrected using a leaching emission factor of 0.18, sourced from Mekonnen and Hoekstra (2015). [c]These estimates were made using national average leakage factor (0.16) and a state average C_t.

Contributions from different water use sectors to the national ABS-NO_3-N flux largely reflect sectoral water abstraction volumes, with freshwater abstractions for irrigation, thermoelectric power, and public supply collectively accounting for 90% of the country's annual total water abstraction volume (Dieter, Maupin, et al., 2018) and 89% of the total ABS-NO_3-N flux (Figure 2b). Variations between county ABS-NO_3-N fluxes (Figure 1) and the sectoral contributions to state-totals (Figure 2) also reflect strong regional differences in water resource use and thus abstraction volumes. For example, IRR-NO_3-N fluxes dominate in western states, whereas this flux only accounts for 37% of the national ABS-NO_3-N flux. Conversely, TE-NO_3-N fluxes dominate in eastern states, facilitating 70% of net power generation in the United States (Dieter, Maupin, et al., 2018), despite collectively only accounting for 27% of the national ABS-NO_3-N flux.

The influence of raw water NO_3-N concentrations upon final ABS-NO_3-N fluxes is illustrated by the fact that although freshwater withdrawals for public supply made up 46% of New York's total freshwater withdrawals, they are responsible for 51% of the state's ABS-NO_3-N flux (Figure 2b). Across New York, freshwater withdrawals for public supply are 25% from groundwater and 75% from surface water (Dieter, Linsey, et al., 2018). However, due to the fact that groundwater NO_3-N concentrations are over four times that of surface water (Figures S1 and S2 in Supporting Information S1), these abstractions contribute 60% and 40% to the state's total PWS-NO_3-N flux (Figure 2b). Counties and states whose water use sectors have a larger dependency on groundwater abstractions are likely to have abstraction fluxes relatively larger than surface water abstractions of the same volume would have, due to the higher NO_3-N concentrations generally found within groundwaters compared to surface waters (Figures S1–S4 in Supporting Information S1) (Pennino et al., 2017). The adoption of a median raw water NO_3-N concentration for each county (see Text S1 in Supporting Information S1) results in high levels of uncertainty surrounding state-level ABS-NO_3-N fluxes (Figure 2a). State-level ABS-NO_3-N flux estimates with higher levels of uncertainty, such as for California (Figure 2a), reflect the greater inter-quartile range of raw water NO_3-N concentrations used to derive median values (see Section 2.3). In reality, these concentrations are highly spatially

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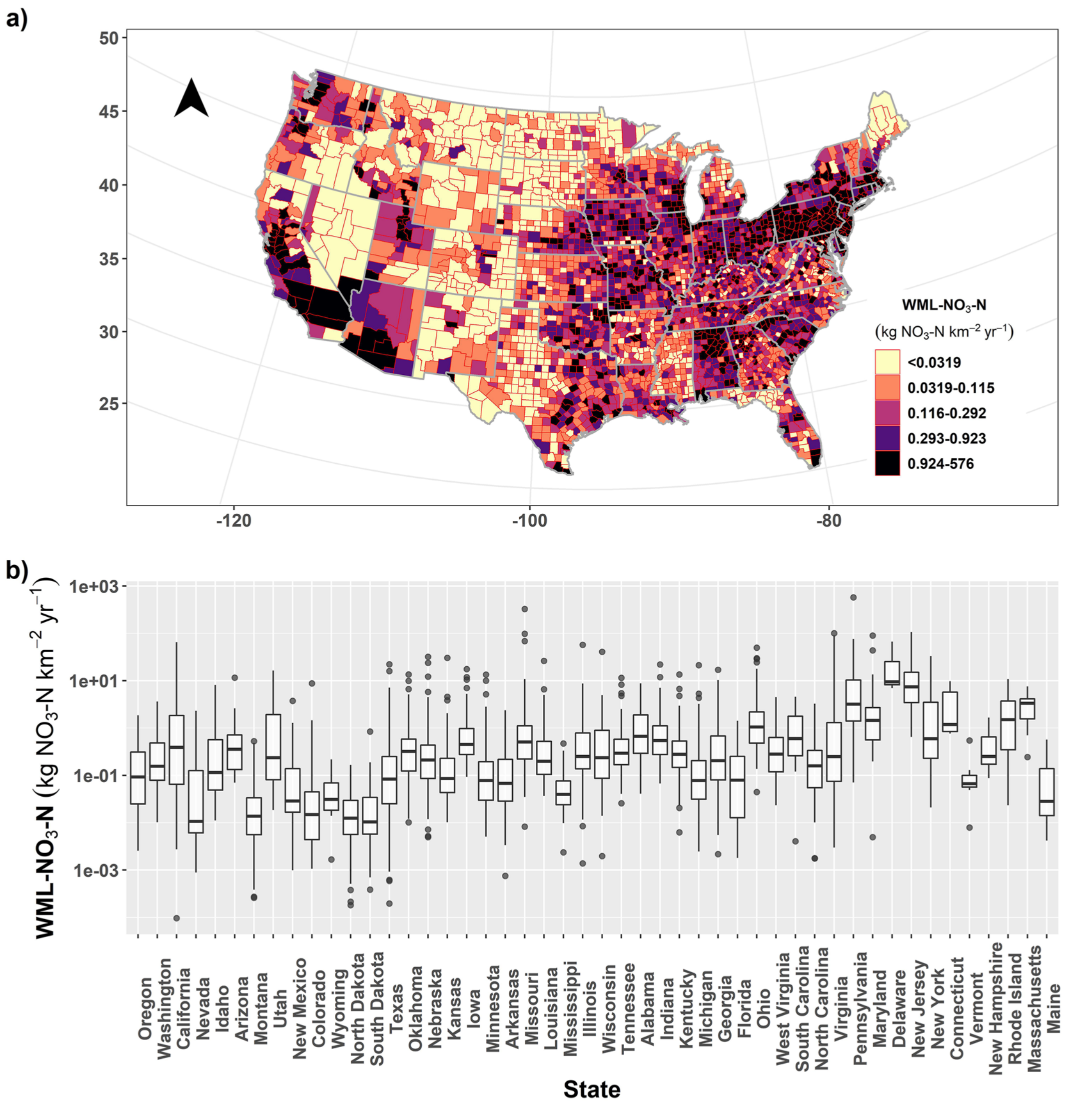

Figure 3. (a) Total area-normalized WML-NO$_3$-N fluxes for each county across the United States (US). Breaks in the color scale are defined as separate quintile groups. Linework was created using the 'usmap' package on R (Di Lorenzo, 2022). (b) Boxplot showing the distribution of county-level WML-NO$_3$-N flux values across each US state. States are ordered from west to east.

variable, even within individual counties (Figures S5–S8 in Supporting Information S1). Minimizing the uncertainties associated with ABS-NO$_3$-N flux estimates should be a future priority, particularly if the spatial distribution of concentration data increases.

This effect of elevated NO$_3$-N concentrations in groundwaters on abstraction fluxes is even stronger for self-supplied domestic withdrawals. Although self-supplied domestic withdrawals account for 3.5% of New York's total withdrawals (Dieter, Linsey, et al., 2018), the state's DOMESTIC-NO$_3$-N flux constitutes 15% of its total ABS-NO$_3$-N flux (Figure 2b). NO$_3$-N concentrations are often higher within domestic supply wells than public supply wells, due to the fact that these wells are screened at shallower depths and are often located in rural areas heavily influenced by agricultural fertilizer practices, and thus closer to anthropogenic sources of NO$_3$ contamination (Desimone et al., 2009; Johnson & Belitz, 2015). With irrigation withdrawals only decreasing by 23% since 2005, the 40% difference between the IRR-NO$_3$-N estimate made for California in 2005 by Liptzin and Dahlgren (2016) compared to that in this paper (Table 1) is only partially explained by changing withdrawal volumes. We suggest that changes to raw water NO$_3$-N concentrations will also control the spatial and temporal variations observed in ABS-NO$_3$-N estimates and that geographic areas and sectors withdrawing water with

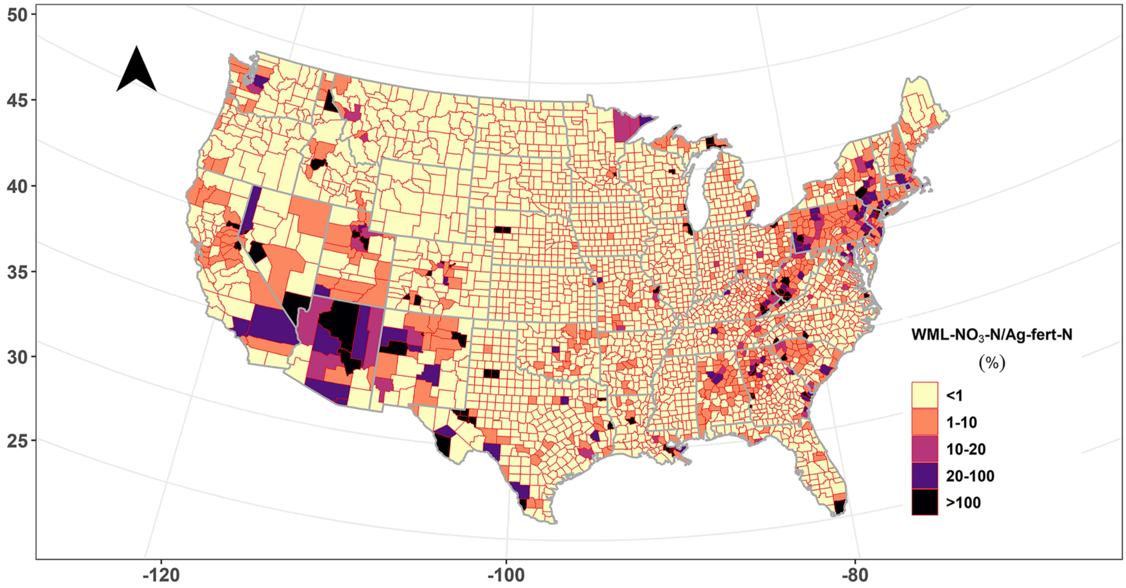

Figure 4. County-level WML-NO_3-N fluxes as percentage equivalent of N leached to groundwater from agricultural fertilizer application. Linework was created using the 'usmap' package on R (Di Lorenzo, 2022).

elevated NO_3-N concentrations can have disproportionate impacts on ABS-NO_3-N fluxes (Stahl, 2019). Whilst data availability allowed specific public and domestic supply raw groundwater NO_3-N concentration (C_r) data sets to be used in estimating PWS-NO_3-N and DOMESTIC-NO_3-N abstraction fluxes, the surface water and groundwater C_r data sets for other water uses do not distinguish values between each sector. Future research should investigate the potential differences in C_r NO_3-N values between each water use sector and utilize these sector specific values to further resolve abstraction flux estimates.

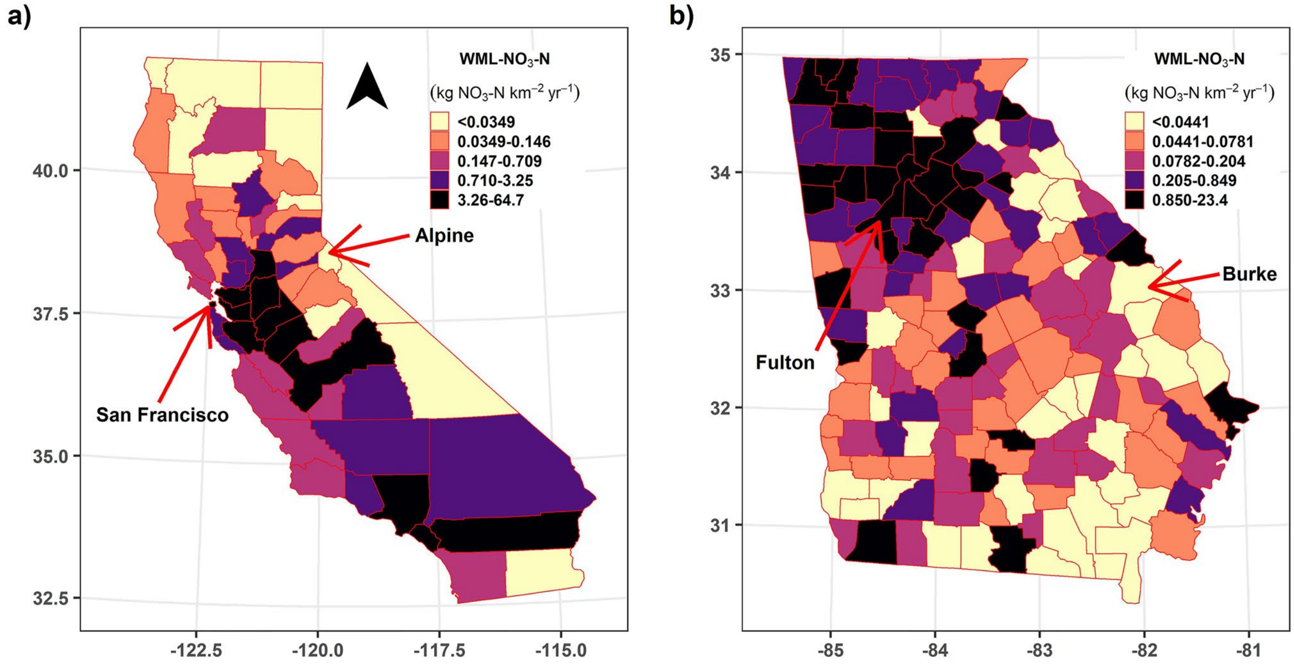

Figure 5. Estimated area-normalized WML-NO_3-N fluxes for each county within the states, calculated using utility-level audit data of (a) California and (b) Georgia. Breaks in color scales are defined as separate quintile groups. Linework was created using the 'usmap' package on R (Di Lorenzo, 2022).

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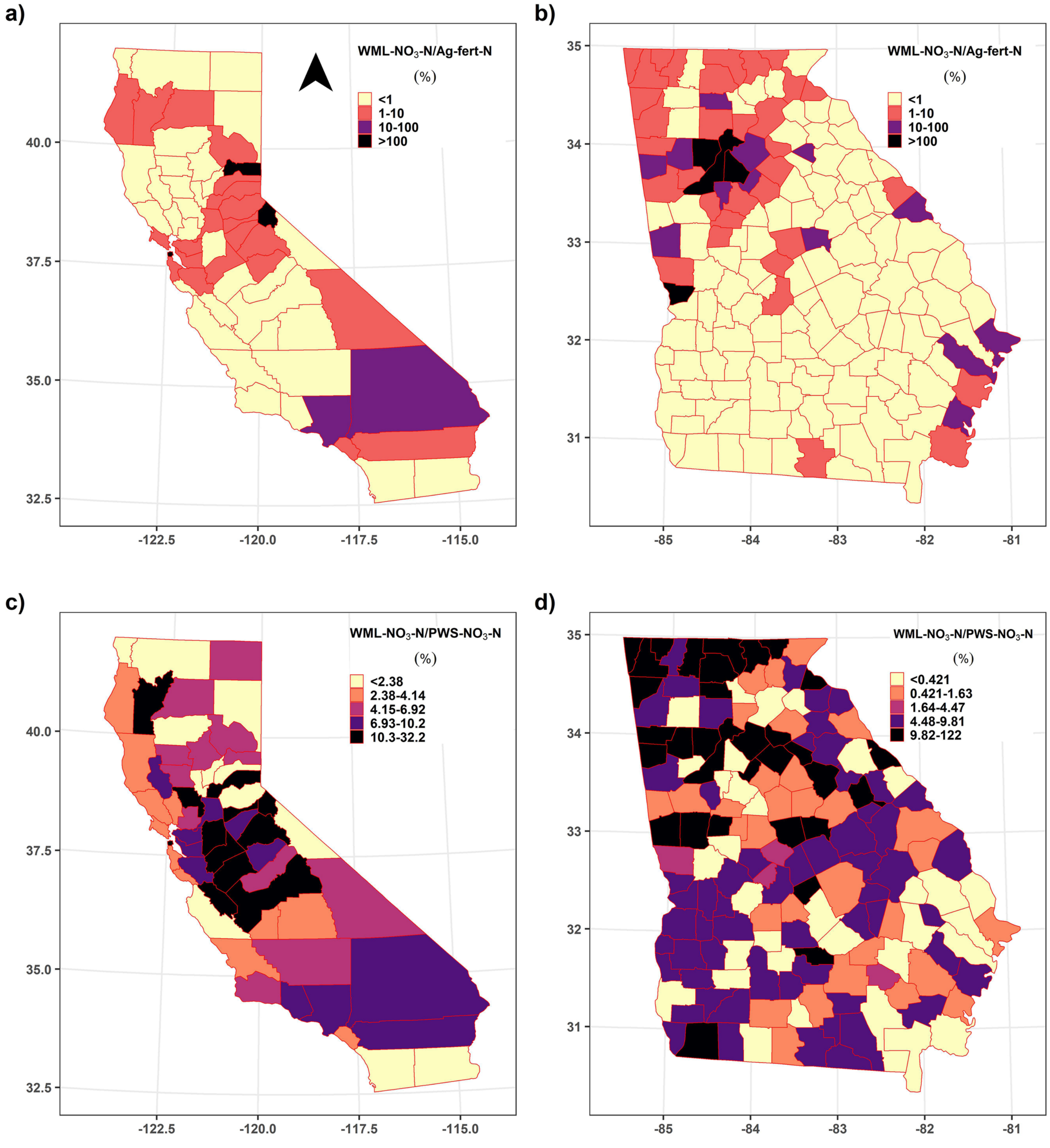

Figure 6. County-level WML-NO_3-N fluxes in California and Georgia as percentage equivalents of (a) and (b) leached N from agricultural fertilizer application, (c) and (d) PWS-NO_3-N. Breaks in color scales for figures c and d are defined as separate quintile groups. Linework was created using the 'usmap' package on R (Di Lorenzo, 2022).

4.2. ABS-NO_3-N as a N Retention Mechanism of Local, National and Global Importance

Whilst denitrification acts as mechanism that removes N from fresh waters on longer timescales and across system boundaries (Grizzetti et al., 2015), abstraction fluxes instead temporarily reroute NO_3-N internally within the defined system, before being returned to the environment. Such a distinction reflects that made by Liptzin and

Dahlgren (2016), in which the irrigation abstraction N flux estimate was represented as an internal flux between different subsystems, as opposed to a permanent removal from the state-level system. Whilst we acknowledge the limitation of comparing temporary and internal N fluxes with longer-term external boundary fluxes (see Section 2.4), the 57% and 97% equivalence of our national-level and California state-level ABS-NO_3-N fluxes (respectively) to total denitrification (Table 1) provides an initial insight into the potential importance of these internal fluxes.

Contrasting patterns in transport, consumption, and fate of freshwaters abstracted for different uses will likely have varying effects upon N cycling. Deducing the implications of these abstraction fluxes thus requires a disaggregation of total ABS-NO_3-N values into their constituent sectors. With regards to PWS-NO_3-N fluxes, the average residence time for water in the US public supply distribution system is estimated at 1.3 days (USEPA, 2002), we could expect WML-NO_3-N fluxes to return some of the NO_3-N within a similar timeframe. Despite this, denitrification can be used as an intentional treatment option either presupply (Hunter, 2008), or during the wastewater treatment process, and will result in the long-term removal of NO_3-N from water and a release of N into the atmosphere (USEPA, 2007). In addition, water that is abstracted and transported across catchment boundaries, often for agricultural and municipal use (Dickson & Dzombak, 2017; Young & Brozovik, 2019), can be thought of as a permanent removal from the abstraction catchment, and as an input into the receiving catchment, thus invalidating the assumption that abstraction fluxes merely occur internally between subsystems and instead contribute to net anthropogenic inputs of N (Hong et al., 2011; Swaney et al., 2018a). The exceedance of WML-NO_3-N fluxes compared to PWS-NO_3-N fluxes in some counties (Figure 6) is potentially attributable to these freshwater transfers across county boundaries (Dickson & Dzombak, 2017), where low PWS-NO_3-N fluxes are a result of water imports offsetting the need for freshwater abstractions. Many urbanized areas across the United States are particularly reliant on such transfers, such as in San Francisco and counties in the San Joaquin Valley (e.g., Fresno, Stanislaus, and San Joaquin) and Greater Sacramento areas (Placer and Yolo), where a significant source of fresh water for public use is via imports from the State Water and Central Valley Projects (Feinstein & Thebo, 2021; USBR, 2021). Similarly, counties in northwest Georgia where WML-NO_3-N fluxes return a large proportion of the PWS-NO_3-N flux (Figure 6), receive drinking water from interbasin transfers (Metropolitan North Georgia Water Planning District, 2017). Under predicted future water stress (Brown et al., 2019), the influence of increasing water transfers upon N fluxes across hydrological and administrative boundaries will add nuance to many of the existing input-output anthropogenic N budget methodologies (Byrnes et al., 2020; Swaney et al., 2018a), and represents an important priority for future research.

The timeframe and spatial distribution of return fluxes and the overall effect of abstractions for nonpublic water use sectors upon N cycling remain largely unknown. Unlike public supply abstractions, where water is often distributed large distances via distribution networks, irrigation abstractions responsible for IRR-NO_3-N fluxes are often done on-site or near to the location of irrigation (Young & Brozovik, 2019). Whilst some of the water abstracted for irrigation use is returned to the environment, including via seepage from irrigation canals (Hrozencik et al., 2021), more than 50% of irrigation withdrawals are consumptive (Dieter, Linsey, et al., 2018). The consumptive use of water for irrigation could act as a mechanism that retains substantial masses of N from freshwater systems on timeframes relevant to nutrient budgets that inform nutrient management plans (Zhang et al., 2020). With over half of the nation-wide irrigation water withdrawals being from groundwater, this flux also represents substantial movement of NO_3-N from the subsurface to surface environment, where different environmental conditions will significantly affect the behavior of NO_3 (Winter et al., 1998). Once applied on land, the NO_3-N deposited on agricultural soils may become significantly concentrated when the large volumes of water applied are subjected to evapotranspiration (Dieter, Maupin, et al., 2018). This N can either accumulate in the soil, be taken up by plants, be lost to the atmosphere, or leached back to groundwater—meaning that return fluxes of IRR-NO_3-N will operate over a range of timescales (Galloway et al., 2003).

Thermoelectric water abstractions are also typically self-supplied, with withdrawals often undertaken on-site and predominantly taken from (and returned to) surface water. On a national-level, water use for the thermoelectric power generation is largely non consumptive, with 64% of the total freshwater withdrawn for this sector returned to its source (Dieter, Maupin, et al., 2018). Whilst the elevated temperatures of return flows are known to impact the quality of source water bodies, NO_3-N concentrations within these waters may be more concentrated as a result of water evaporation during the cooling process (Petrakopoulou, 2021). Whilst low on a national-level, consumptive water use for the generation of thermoelectric power is proportionally higher across certain regions.

Due to the lower availability of fresh water, the majority of thermoelectric power plants across western US states continually circulate water through heat exchangers, which leads to a larger proportion of water being consumed. In contrast, the higher availability of fresh water across eastern states means that once water has passed through heat exchangers, it is then returned to its source, thus consuming less water (Lee et al., 2018). Although THERM-NO_3-N fluxes make a smaller relative contribution to overall ABS-NO_3-N fluxes in western states (Figure 2b), the higher proportion of water consumption in these areas results in these fluxes acting as a temporary anthropogenic store of N. Whilst the timescale and magnitude of N retention associated with consumptive withdrawals may be transient when compared with more permanent removals such as denitrification and burial in sediments (Baron et al., 2012), they may become relevant when considered in relation to nutrient balances and management decisions related to smaller subnational (e.g., catchment-scale) systems (Ator & Denver, 2015). With Europe also dedicating large volumes of water to thermoelectric power generation (Magagna et al., 2018), the potential for THERM-NO_3-N fluxes to retain N across the United States warrants their quantification elsewhere around the globe.

Comprehensive consumptive use data for the remaining water use sectors (industry, domestic, aquaculture, livestock, and mining) are unavailable across the US; however, regional assessments have been made (Shaffer, 2008; Shaffer & Runkle, 2007). In addition, consumptive use volumes of fresh water for thermoelectric use exceed total fresh water thermoelectric withdrawals in Arizona, California, and Oregon, potentially due to this sector receiving water transfers in these states (see previous discussion on the effects of transfers). The amount of water consumed by these water use sectors depends on a wide range of processes that vary geographically across the US, including evapotranspiration, product incorporation, as well as livestock and human consumption (Shaffer & Runkle, 2007).

Improving understanding of the different effects that sectoral abstraction fluxes have upon N cycling across the United States is imperative in order to further resolve nuances within nutrient budget methodologies and may have implications within the nutrient spiraling paradigm (Ensign & Doyle, 2006). For example, the change in storage of N internally within surface waters and groundwaters is a component of the California Nitrogen Assessment (Liptzin & Dahlgren, 2016). This value is quantified as the difference between the other input and output N fluxes defined within the budget to these subsystems. The temporary anthropogenic store of N that our ABS-NO_3-N flux represents may thus account for 12% of the internal storage of N within California's surface waters and groundwaters (Table 1), thus emphasizing the importance of its inclusion within future state-level N balances.

The annual ABS-NO_3-N estimates presented here neglect seasonal fluctuations in both volume and quality of abstracted water (Bexfield & Jurgens, 2014; Josset et al., 2019; Lee et al., 2020; Ornelas Van Horne et al., 2019; Wiener et al., 2020). As more temporally and spatially resolved water abstraction volume and quality data becomes available, future research could seek to assess the seasonality in, and reduce uncertainties associated with, ABS-NO_3-N flux estimates. Anticipated increases in human population and a changing climate will also affect future ABS-NO_3-N fluxes in the United States and around the world (Brown et al., 2013; Harris & Diehl, 2019; McDonald & Girvetz, 2013; Pickard et al., 2017; Wada & Bierkens, 2014), and remains an important research gap.

4.3. WML-NO_3-N Returns Variable Amounts of N Back to the Environment

The national WML-NO_3-N estimate returns a relatively small flux of NO_3-N to the environment when compared to other global and national N input and return fluxes (Table 2). Despite the volume of leaked water within the United States accounting for around 11% of the estimated global total reported by Wyatt and Liemberger (2019), our WML-NO_3-N estimate accounts for only 1.3% of the global WML-NO_3-N estimate. This potentially reflects the relatively low NO_3-N concentrations of treated water in the United States as a result of high levels of drinking water treatment (EWG, 2019). The ratio of estimates for national-scale WML-NO_3-N compared to the PWS-NO_3-N that we report (Table 2) is also similar to that in England, UK (Ascott, Gooddy, & Surridge, 2018). As developed countries, both the United States and England have relatively low average fractional leakage rates when compared to many developing countries, such as Vietnam (Kingdom et al., 2006), where along with higher concentrations of NO_3-N in drinking water (Hung et al., 2020), we may expect to find higher area-normalized WML-NO_3-N fluxes than in developed countries, such as the US.

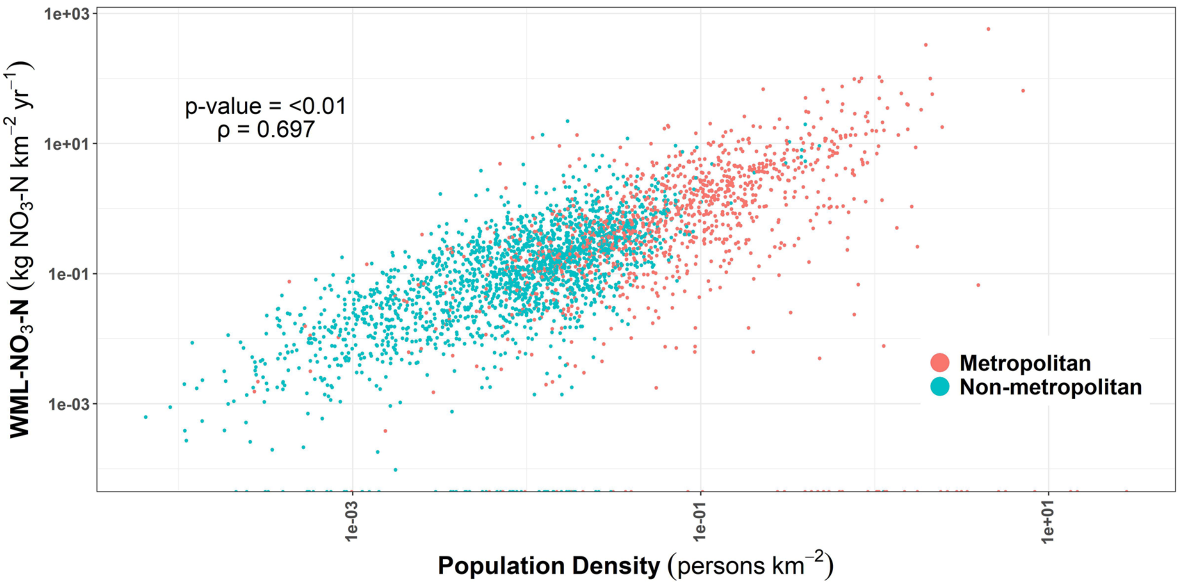

Figure 7. Plot showing the relationship between watermains leakage fluxes and population density for urban and nonurban counties across the United States (US), as defined by the US Department of Agriculture. Spearman's rank correlation test returned a p-value < 0.01 and a positive rank correlation ($\rho = 0.697$), suggesting a strong monotonic relationship between watermains leakage fluxes and population density.

This national WML-NO_3-N estimate masks the localized importance of this flux across many urban and coastal counties (Figure 3a). Despite the predominantly low NO_3-N concentrations in treated tap water across the country, leakage from water distribution networks can still lead to large overall nutrient loads due to the large volume of water released into the environment. Higher area-normalized county WML-NO_3-N fluxes largely reflect higher population densities of more urbanized counties (Figure 7), often in coastal areas (Figure S9 in Supporting Information S1). These areas have larger volumes of water input into their distribution network and a higher density of watermains pipes with the potential to leak, per unit of land area. The ages of water distribution networks also vary regionally across the US, with older pipes more susceptible to leakage largely found in the older cities in eastern states (Speight, 2015). Although limited, the fractional leakage rate data set shows larger f_{state} values to be concentrated in eastern states (see Text S1 in Supporting Information S1), where together with their larger population densities, we observe larger WML-NO_3-N fluxes (Figure 7).

Along with the absolute magnitude of the WML-NO_3-N flux, the relative importance of this flux to county-level N cycling is largely determined by the importance of other N retention and input mechanisms in these areas. We find that urban counties, as defined by US Department of Agriculture (USDA, 2020), account for 96% of the total number of counties whose WML-NO_3-N flux exceeds the median ABS-NO_3-N flux for all counties.

Whilst N fertilizer application is the largest anthropogenic N input on a national scale, N inputs in many localized areas are dominated by other natural and anthropogenic processes (Sabo et al., 2019; Swaney et al., 2018a). Our research highlights water leakage from mains distribution networks as an important, yet previously largely overlooked, localized input of N. For example, although California and Georgia have relatively small state-wide WML-NO_3-N estimates, the counties of San Francisco and Fulton have WML-NO_3-N fluxes that exceed the amount of N leached from agricultural fertilizer (Figure 6), as a result of both urbanization and low agricultural activity. There is a decreasing trend in agricultural N fertilizer inputs across many urban counties of the US, particularly in the northeast (Sabo et al., 2019), as well as reports of zero agricultural N fertilizer application across many interior and southeastern states (Swaney et al., 2018a). We find that urban counties account for 84% of total counties where WML-NO_3-N fluxes exceed N leached from agricultural fertilizer, highlighting the significance of WML-NO_3-N fluxes as a component of overall anthropogenic N cycles in these areas (Figure 4). Conversely, in areas with intensive agricultural activity, such as counties across the midwestern states and California's Central Valley region, N fertilizer application is the largest terrestrial N input (Sabo et al., 2019) and the main cause of elevated groundwater NO_3-N concentrations (Exner et al., 2014; Harter et al., 2017), meaning that WML-NO_3-N fluxes make a relatively small contribution to local anthropogenic N cycles (Figure 4).

Along with contributions of leaky sewers to N within the country's urban streams (Divers et al., 2013; Pennino et al., 2016; Viers et al., 2012), these newly quantified WML-NO_3-N fluxes further our understanding of how urban development acts as an agent of environmental change (McGrane, 2016). These results not only contribute to the particular challenge of understanding nonpoint source pollution in urban watersheds (Cappiella et al., 2012; Hobbie et al., 2017; Pennino et al., 2016) but also our understanding of the risks to the wider environment. For example, the proximity of many urbanized areas to coastal environments (Figure S9 in Supporting Information S1) means that nutrients delivered to urban fresh waters may be more likely to affect coastal water quality and ecosystems (Sawyer et al., 2016).

In the face of increasing water scarcity, the need to reduce leakage from distribution networks has been well-established (Speight, 2015; Xu et al., 2014). Although WML-NO_3-N fluxes have associated environmental costs, largely due to their contribution to NO_3-N in groundwaters and other receiving waters downstream, the relatively low NO_3-N concentrations within leaked treated water in comparison to those from nearby sewage network leakage means that WML-NO_3-N fluxes may dilute concentrations of N in groundwater (Yates et al., 1990). Understanding the trade-off between these negative and positive impacts as well as the fates of WML-NO_3-N fluxes once released into the environment will be important in future evaluations of environmental impacts and policy surrounding leakage control (Ascott, Gooddy, & Surridge, 2018; Xu et al., 2014).

The consistency of WML-NO_3-N flux estimates in California and Georgia using both methods outlined in Section 2.2 indicates that whilst using utility-level data will likely carry less uncertainty, the suite of assumptions adopted for many states due to the absence of data may still provide a reasonable first estimate of WML-NO_3-N fluxes. With some states planning to legally require validated audits that disclose utility-level volumetric leakage rates (e.g., New Jersey Department of Environmental Protection (2017)), these new data sets will allow WML-NO_3-N fluxes to be determined with lower uncertainties and at finer spatial resolution in the future, such as for individual public water systems or watershed catchments.

Extreme weather events across the United States may lead to seasonal WML-NO_3-N fluxes (Folkman, 2018; Healey et al., 2021; Miller, 2021), similar to those observed in England, UK, as a result of winter pipe burst events (Ascott, Gooddy, Lapworth, et al., 2018). Watermains leakage has been found to make significant contributions to baseflow in urban streams during summer months (Fillo et al., 2021), suggesting that the relative importance of WML-NO_3-N fluxes may also change seasonally. Investigating how these fluxes change throughout the year, as well as with anticipated aging water infrastructure replacement (USEPA, 2021a), should be a focus of future research. Watermains leakage is also an important return mechanism of phosphate in urban areas across England, UK (Ascott et al., 2016; Gooddy et al., 2017; Holman et al., 2008; Wakida & Lerner, 2005). The widespread phosphate dosing of tap water across the United States for the purpose of corrosion control (McNeill & Edwards, 2002) and the known contribution of phosphate to freshwater eutrophication across the US, particularly within urban catchments (Haque, 2021; Hejna & Cutright, 2021; Metson et al., 2017; Watson et al., 2016), warrants estimation of watermains leakage phosphate fluxes.

4.4. Policy Implications

Whilst we do not aim to make prescriptive policy suggestions here, the advancement in understanding from this research could contribute to the formulation of more effective and integrated nutrient management strategies (Ator & Denver, 2015; EPA Science Advisory Board, 2011; Grizzetti et al., 2015; Tomich et al., 2016). Now revealed to be a significant retention mechanism on a range of spatial scales, the absence of ABS-NO_3-N and variable return times for this flux in N balances may invalidate many models and budgets used by policymakers in their attempt to manage N (Ascott et al., 2021). For instance, increased understanding of IRR-NO_3-N fluxes will aid the development of optimum N fertilizer application and crop production recommendations, as the movement of N associated with water abstractions has implications for the amount of N required to be added to soils. The potential impact of water transfers on the balance between PWS-NO_3-N and WML-NO_3-N fluxes could also influence the future regulation of water withdrawal permits and transfers both in the United States and around the world (Shumilova et al., 2018). The national WML-NO_3-N estimate will facilitate international comparisons (Swaney et al., 2018b), and its significance on local scales should help resolve urban watershed scale N budgets (Winiwarter et al., 2020) and inform local and state optimum leakage control policy (Xu et al., 2014).

5. Conclusions

Understanding anthropogenic controls on the N cycle is imperative for mitigating the effects of human activity on nutrient pollution in fresh water. In this research, we quantify for the first time how processes associated with water supply drive changes in the N cycle across the US. The abstraction of fresh water for both public and nonpublic supply has a potentially significant influence on N cycling across the US. We find that consumptive water use may act as a relatively significant temporary internal store of N on a national level, and that post-withdrawal water transfers may be a mechanism importing and exporting N across system boundaries on a subnational level. These newly quantified processes may occur on timescales relevant to nutrient management and thus be of interest to stakeholders involved in developing more effective nutrient management strategies. Their significance in the context of other N fluxes suggests that internal fluxes may be a necessary nuance to be considered within future N budget methodologies. Watermains leakage is estimated to return 7.0 kt NO_3-N yr^{-1} back to the environment across the US. Despite a small flux on a national level when compared to other major N inputs, this estimate masks greater relative importance of watermains leakage fluxes of N on more localized scales, with this flux exceeding the amount of N leached from agricultural fertilizer input in some urbanized counties. The results and transferrable methodology we report here, using the United States as an exemplar, should support future research to quantify similar fluxes for other locations around the globe as more data becomes available.

Data Availability Statement

All data used within this research are publicly available. Withdrawal data are available from Dieter, Linsey, et al. (2018), raw surface water concentrations were available from the Water Quality Portal (2021), and obtained using the 'dataRetrieval' package in R (De Cicco et al., 2018). Raw groundwater concentrations were sourced from the USGS Groundwater Data Releases as part of the National Water-Quality Assessment (https://www.sciencebase.gov/catalog/item/57f7f703e4b0bc0bec0a1ba8). Treated water concentrations are available from the Environmental Working Group's Tap Water Database via their website (EWG, 2019) and volumetric leakage rate data are available from a variety of sources, see Text S1 in Supporting Information S1. Linework for map figures was created using the 'usmap' package on R (Di Lorenzo, 2022).

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