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Seasonal disconnect between streamflow and retention shapes riverine nitrogen export in the Willamette River Basin, Oregon

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Abstract

Watershed nutrient balance studies traditionally focus on annual fluxes. In areas with strongly seasonal, Mediterranean-type climate regimes, riverine nutrient export may be greater during wet seasons when hydrologic forcing overwhelms or bypasses retention mechanisms. By combining data on riverine export with spatially detailed nutrient inputs, we examine how nitrogen (N) supply, retention, and streamflow shape annual and seasonal riverine N export in Oregon's Willamette River Basin (WRB). The WRB has pronounced dry summers and wet winters, and the distribution of farmland, cities and forests create significant spatial variations in N inputs. Local data on N inputs were coupled with streamflow and chemistry to calculate fractional N export for 22 WRB sub-watersheds in the mid-2000s. For the entire WRB, 78% of the N inputs came from agricultural activities, mainly as synthetic fertilizer (69%); the next largest inputs were deposition (10%), alder fixation (5%) and point sources (5%). Crop-specific estimates of fertilizer agreed with county fertilizer sales rates at the high end of extension recommendations. Fractional riverine N export (annual riverine N export / net watershed N input) averaged 38% of net inputs in WRB tributaries, greater than other regions of North America. Fall and winter together accounted for 60–90% of the riverine N export across all watersheds. Summer export was small but was greatest in the watersheds that receive seasonal snowmelt. Large wet season losses, when biotic sinks are less active, result in a relatively high proportion of N inputs exported in this region with a Mediterranean climate and high runoff.

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AUTHOR CONTRIBUTIONS

JEC conceived the study and drafted the manuscript. KEG and DJS were involved in the conception and design of the study, creation of the spatial input layers, interpretation of results and writing the manuscript. KEG conducted the stream and river load modeling. JL conducted the final input calculations and contributed to interpretation and writing the manuscript.

Keywords

nitrogen inputs; fertilizer; N fixation; watersheds; runoff; red alder; stream N export; watershed nitrogen dynamics

INTRODUCTION

Human activities have increased overall N inputs to the entire United States by approximately three-fold since pre-industrial times (Sobota and others 2013). This increase is spatially heterogeneous with much larger rates in some areas (David and Gentry 2000; Sobota and others 2013; Swaney and others 2018). At the watershed scale, N inputs are transported out of basins through crop harvest, hydrologic export and gaseous losses to the atmosphere, or are stored in soils, wetlands or groundwater within the basin (Howarth and others 1996; Seitzinger and others 2006). The release of N to the environment beyond its intended economic and societal uses has important effects on air and water quality, drinking water supplies, and the integrity, function and productivity of terrestrial, freshwater and coastal ecosystems (Schlesinger 2009; Davidson and others 2011; Sobota and others and others 2015). Improvements in the quality of information about nutrient release to the environment can be used to inform management of non-point source pollution, which continues to be a challenging problem at local to global scales (Dowd and others 2008; Patterson and others 2013).

Watershed N input-export budgets provide insight into the relative influence of anthropogenic N inputs on N processing and retention potential within watersheds. Here we define retention as all N storage and removal mechanisms including denitrification, and is a function of soil, plant, microbial, groundwater and riparian zone processes that remove N from flow paths via biotic uptake, incorporation into organic matter, storage, and denitrification. Variations in N supply often drive the spatial patterns in riverine N export from watersheds (Caraco and Cole 1999; Howarth and others 2011). However, interactions of N retention mechanisms and hydrology over space and time can also influence crop N use efficiencies and rates of leaching and export (Schaefer and Alber 2007; Sinha and Michalak 2016).

The concept of N saturation was developed for forests responding to increasing N deposition, with a projected set of changes in ecosystem processes that could lead to increased gaseous and hydrologic losses of N (Aber and others 1989). Human-modified landscapes dominated by urban and agricultural land use can often have much greater N inputs than deposition and may be at later stages of N saturation (David and Gentry 2000; Groffman and others 2004). The efficiency of N retention is expected to decline as N inputs increase (Aber and others 1989; Niu and others 2016), but retention can vary substantially even among watersheds close in space due to local hydrologic properties, soils, vegetation and land-use history that influence N retention (Campbell and others 2004; Sobota and others 2009). As N retention declines, fractional export of N inputs may increase.

Interactions of N supply, retention, and export in watersheds can be particularly complex in Mediterranean climates with warm, dry summers and warm, wet winters, such as the

western US (Sobota and others 2009; Mueller-Warrant and others 2012) and Italy (De Girolamo and others 2017). Examining these regions provides an opportunity to understand how climate regimes and input factors interact to shape retention and corresponding riverine N export over space and time. In western Oregon of the US, watershed processes can generate high relative and absolute riverine N export (Schaefer and others 2009), even though N inputs are low to moderate for the region as compared to other parts of the US (Sobota and others 2013). Possible explanations for these high export rates include spatial and/or temporal disconnects between N inputs, retention mechanisms and loss vectors.

In Oregon's Willamette River basin (WRB), forests of the mid-elevation Coast Range to the west and south and the high-elevation Cascade Mountains to the east surround a productive agricultural valley. Fine-textured soils and flat ground produce a diverse and valuable set of agricultural products including grass seed, wheat, vegetables, and many specialty nursery crops (Berger and Bolte 2004). Most of these products require significant inputs of synthetic N fertilizers ($100\text{--}250\text{ kg N ha}^{-1}\text{yr}^{-1}$); a substantial fraction of these N inputs leach from the rooting zones beyond the agricultural systems (Brandi-Dohrn and others 1997; Feaga and others 2010). In the upland forested landscapes bordering the agricultural valley, biological N fixation by pure stands of red alder (*Alnus rubra*) trees can reach $100\text{--}200\text{ kg N ha}^{-1}\text{yr}^{-1}$ (Binkley and others 1994) and leaching of N to surface waters can be $35\text{ kg N ha}^{-1}\text{yr}^{-1}$ in pure stands (Compton and others 2003). Other important sources of N to the WRB include atmospheric deposition, land application of manure from animal operations, biological N fixation by agricultural crops, lawn fertilizers, municipal wastewater and industrial discharges (Wise and Johnson 2011). Imports of food and feed are also important watershed inputs (Howarth and others 2012), entering the environment mainly as wastewater and application of animal manures to farmland.

The rates and spatial distribution of N inputs and retention in the WRB have important consequences for water quality, where nearly 20% of stream length is deemed in fair or poor condition by the state of Oregon due to high total N concentrations (ODEQ 2009). Nitrate contamination of groundwater is a concern in the WRB, particularly for private wells (Hoppe and others 2014). Water scarcity may become more prevalent in the region in the future (Jaeger and others 2017), and thus understanding of the interactions between N inputs, retention and export is needed for management of surface and drinking water quality in the region.

Seasonality of precipitation and runoff, characterized by wet winters and dry summers, may be a prominent factor behind the relatively high rates of fractional N export observed in western Oregon (Schaefer and others 2009). Fractional N export refers to the proportion of N inputs that are exported in streamflow and is expected to increase with precipitation (Howarth and others 2006). Prior biogeochemical research in Pacific Northwest (PNW) watersheds has focused on annual N fluxes (Wise and Johnson 2011, Steinberg and others 2011) or patterns of concentration (Poor and others 2008; Greathouse and others 2014), with few studies examining the seasonal distribution of fluxes (Vanderbilt and others 2003; Sigleo and Frick 2007). Timing and amount of rainfall can influence the ecosystem controls on N loss when uptake and removal are segregated in time and space (Perakis 2002; Ohte and others 2010). For example, during seasons when hydrologic connectivity is low, N transport

from terrestrial to aquatic systems may be low irrespective of biotic uptake (Fratterigo and Downing 2008). Considering the strong seasonality of rainfall and streamflow in the PNW, riverine export is also expected to be strongly seasonal.

In this paper we explore the proposal that fractional N export is high in systems with Mediterranean climates because of a disconnect among the timing of N inputs, retention mechanisms, and hydrology. Agricultural fertilizer inputs occur early in the growing season to coincide with higher plant demand, but riverine exports are driven by high fall and winter runoff when plant uptake is low and soil flow paths transfer water (Lin and others 2019). To examine this timing disconnect, we studied spatial patterns of N inputs and stream or river N concentrations and export in 22 WRB sub-watersheds. Recent work quantified point source, fertilizer and manure N sources for the Pacific Northwest using the USGS-SPARROW model (Wise and Johnson 2013); other recent studies provide ground-truthed, crop- and species-specific land cover layers for WRB farmland (Mueller-Warrant and others 2011) and forests (Ohmann and others 2011). Collectively, these efforts allow the generation of locally derived and spatially detailed information about N inputs to the landscape from human-driven and natural sources. For these watersheds, we use available data on N concentrations and streamflow to estimate seasonal and annual N export for the years 2002–2006 from the entire WRB and WRB sub-watersheds. The disconnect in timing between retention mechanisms and runoff may result in high absolute and fractional rates of riverine N export, creating a substantial challenge to nutrient management in the WRB.

METHODS

Spatially explicit N inputs

Inputs of land-based sources of N to the landscape were determined for 22 WRB sub-watersheds. We considered sources of N to be agricultural fertilizer, atmospheric N deposition, cultivated biological N fixation (BNF), red alder BNF, point sources, non-farm fertilizer applied to developed lands and manure from animal feeding operations (AFO) applied to farmland as fertilizer. Net food and feed imports to the WRB were small ($0.78 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in 2002; Howarth and others 2012); instead we quantify manure application and sewage inputs to rivers since these fluxes represent a direct release of food and feed derived N to the environment. Much of the input data are from 2002 or 2008 while the river and stream chemistry values are from 2002–2006. Gathering data from multiple sources for the same year is very challenging with the lack of coordination across sectors, thus the inputs and export values generally represent the mid-2000s. Land cover and livestock numbers did not change more than a few percentage ($<5\%$) between 2001–2002 vs. 2006–2007, so these representations are supported. To compare our findings to those of other watersheds, we also calculated Net Anthropogenic N Input (NANI) as total anthropogenic inputs minus crop uptake (e.g., Boyer and others 2002). For the WRB, we estimated crop N removal using the very strong linear relationship with total N inputs in the Calapooia sub-watershed of the WRB (slope = 0.40, $r^2=0.98$; Lin and others 2019). We also quantified wastewater treatment outflows and land application of AFO manure to cropland, which represent the net release of food and feed input to the environment. We did not estimate septic inputs.

Fertilizer applied to WRB agricultural crops (2008): Agricultural fertilizer inputs were calculated using a new approach based on local crop-specific fertilizer recommendations and a layer from the USDA-ARS based on remote sensing classification supported by a ground-truthed census of agricultural production practices for 2008 (G. Mueller-Warrant pers. comm., based on Mueller-Warrant and others 2011; Figure 1). The USDA-ARS maps focus on grass seed cropping systems at a 30×30 m scale and have more detailed information than the USDA Crop Data Layer by including a range of grass seed crops that are important in the southern Willamette Valley (Mueller-Warrant and others 2011) and have different fertilizer input rates (Appendix 1). The 2008 layer has 56 classes of agricultural land categories; for visual purposes, in Figure 1 we combined the agricultural crops into fewer categories. Fertilizer application rates for many of these crops were derived from Oregon State University (OSU) Extension fertilizer guides and are shown in Appendix 1. These crop-specific fertilizer rates were applied to the crop types. The OSU extension guides generally give a lower and an upper limit for the fertilizer application rates and we use these different estimates to obtain a “low” and “high” rate of fertilizer loading for the landscape. We used extension guides to calculate “low” and “high” fertilizer application rates, which we compared with county fertilizer sales (sum of twelve HUC-8 sub-watershed values in the WRB for 2008 published by the International Plant Nutrition Institute; www.ipni.net). Our analysis focuses on the “high” rates because they were more consistent with county-level fertilizer sales data than the “low” estimates at the scale of the entire WRB.

Non-farm fertilizer (2002): We used the 2002 estimates of non-farm fertilizer N applied on developed lands from Wise and Johnson (2013). Briefly, county-level estimates of 2002 non-farm fertilizer use were compiled by the USGS based on fertilizer sales and county-level expenditures, then scaled to incorporate statewide sales data assembled by the state of Oregon. These Oregon-specific, county-level estimates of non-farm fertilizer N application were disaggregated equally to developed land in each county (NLCD categories 21, 22, 23 and 24, minus areas representing roads).

N deposition (2008): To estimate total N deposition within the WRB, we used output from the Community Multiscale Air Quality model for 2008 at a 4×4 km grid resolution. The model simulation yielded total N deposition, including deposition rates for wet deposition based on the National Acid Deposition Program’s National Trends Network, plus dry deposition modeled and adjusted for bias based on measured data from monitoring networks (Schwede and Lear 2014).

Crop biological N fixation (2008): We used a crop-specific method similar to that for fertilizer inputs but applying literature values for average N fixation rates per crop (SAB 2011). N fixing crops included in the 2008 ARS land cover are: alfalfa ($224 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), dry beans ($90 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), clover ($117 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), and grassland/pasture ($1 \text{ kg N ha}^{-1} \text{ yr}^{-1}$). All other agricultural crops were assumed to have $0 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ fixation rate.

Red Alder N fixation (2006): Biological N fixation by red alder (Alder BNF) was estimated using a new approach combining layers of red alder basal area and literature estimates of N fixation. The Gradient Nearest Neighbor 30 × 30 m resolution map produced by the Landscape Ecology, Modeling, Mapping and Analysis group was used to determine the fraction of stand basal area (BA) occupied by red alder in 2006 (Ohmann and others 2011). We then multiplied this metric of red alder stand dominance by a literature-derived N fixation rate for pure alder stands (Binkley and others 1994):

$$\text{Alder BNF} = (\text{red alder BA/total tree BA}) * 100 \text{ kg N ha}^{-1} \text{ yr}^{-1} \quad [\text{Eq. 1}]$$

Most rates ranged between 100–200 kg N ha⁻¹ yr⁻¹ (Binkley and others 1994), thus our estimate of 100 kg N ha⁻¹ yr⁻¹ for pure red alder stands is likely a conservative estimate. We also do not include other PNW symbiotic N fixers like *Ceanothus* spp. and *Lupinus* spp.

Point sources (2002): We estimated point source input by clipping the existing SPAtially Referenced Regression On Watershed attributes (SPARROW) model data to the watershed boundary of the WRB (Wise and Johnson 2013). The SPARROW model used discharge of TN for 2002 from permitted wastewater treatment facilities to estimate point source contribution. It also included TN discharge from fish farms and hatcheries for 2002 by using annual fish production and feed.

Land application of confined animal manure (2002): SPARROW model input shapefile data was applied to calculate N contribution of livestock manure in the WRB. The SPARROW estimation was based on the animal populations at registered dairies and feedlots, county-level livestock populations, and animal-specific N generation factors obtained from USGS for 2002 (Wise and Johnson 2013). All manure generated by confined cattle was assumed to be applied to agricultural land within the catchment containing that dairy or feedlot. We did not quantify manure from unconfined animals and consider that manure deposited by unconfined livestock on grazing lands represents a local recycling of N either present in pasture crops or applied as fertilizer to fodder crops grown locally. A comparison between 2002 and 2007 agricultural census data indicates that there was little change (<5%) in livestock numbers in the WRB between these years (USDA-NASS 2013).

Stream data and calculation of watershed N export

Daily streamflow and periodic grab sample stream total nitrogen (TN) concentration data were retrieved from state and federal agency online databases (Appendix 2). The minimum data requirement for inclusion in this study was at least quarterly measurements from 2002–2006, with many sites having more frequent data. Where TN concentration data were not provided, total Kjeldahl nitrogen (TKN) and nitrate + nitrite concentrations were summed to give TN. Where available, streamflow data were obtained from gauges present at the location where the chemistry measurements were collected or from a nearby USGS or Oregon Water Resources Division gauge (Appendix 2). Ungauged streams, or streams where the stream gauge was not near the chemistry sampling point, were modeled using nearby basins and watershed area corrections (Appendix 3). A set of 22 sampling stations had flow and chemistry data to meet our criteria (Table 1; Figure 2).

Annual and seasonal TN loads (kg N yr^{-1}) were calculated with the USGS program LOADEST (Runkel and others 2004), using at least quarterly TN concentration measurements and daily streamflow for each station for the entire period of record. The best LOADEST model for each station was selected as the one having the lowest Akaike Information Criterion (AIC) value. Annual yields ($\text{kg N ha}^{-1} \text{ yr}^{-1}$) were calculated by dividing the load by basin area. Seasonal flow-weighted concentrations were calculated by summing the daily flux estimates and dividing by the sum of daily runoff for each season. Modeled and measured concentrations and loads for the Willamette mainstem at Portland are shown in Supplemental Figure S1. We calculate the seasonal export both as the absolute rates of export in metric tons (MT N), the areal rates of export ($\text{kg N ha}^{-1} \text{ yr}^{-1}$), and as fractional export of total N inputs and fractional export of NANI to allow comparison with other studies (e.g., Boyer and others 2002). Regression analyses using Excel compared inputs vs. outputs for the tributary sites and mainstem sites to determine whether they had different net retention of total N inputs.

RESULTS

N sources and inputs across the WRB

Agricultural sources dominated N input to the entire WRB (Figure 4), accounting for 78% of total annual N input to the entire basin. Farm fertilizer was by far the most important component of agricultural inputs and comprised 69% of total input to the WRB. The second largest source of N input to the WRB was atmospheric N deposition (10%). Manure from AFOs applied to cropland as fertilizer represented 7% of input. Alder BNF and point sources each represented 5% of input. Non-farm fertilizer and crop-related BNF were each 2% of total N inputs.

Total N input to the entire WRB was $114,000 \text{ MT N yr}^{-1}$ ($1 \text{ MT} = 10^3 \text{ kg N}$) or $30.9 \text{ kg N ha}^{-1} \text{ yr}^{-1}$. N input rates varied by 32-fold across the sub-watersheds studied, from 4.5 to $145 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ (Figure 4a). The greatest input rates were to the Zollner Creek sub-watershed, where watershed area is 90% agriculture (Table 1). Lowest input rates were in largely forested watersheds in the Cascade Mountains. Intermediate rates were seen in many Coast Range and southern Willamette Valley streams (Figure 4a). Synthetic fertilizer was the largest individual N source in 18 of the 22 watersheds and ranged from $6.8 - 92.1 \text{ kg ha}^{-1} \text{ yr}^{-1}$ among sub-watersheds (Figure 4). Manure applied to the landscape as fertilizer was generally $<5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, except for the Zollner Creek watershed where manure application was $41.8 \text{ kg N ha}^{-1} \text{ yr}^{-1}$.

Other sources such as deposition, point sources and alder BNF were important in watersheds located in the mountains or in urban areas. Little Abiqua Creek watershed, which serves as the drinking water supply for the city of Silverton (population 9,222 in 2010), had relatively low input rates, dominated by alder BNF and atmospheric deposition. Atmospheric deposition was the largest source of N input in the Middle Fork Willamette and McKenzie basins as well. Contribution of point sources to total sub-watershed input was generally very low, $<5\%$ in all watersheds except in the Tualatin River basin (9–12%).

Annual and seasonal patterns of N concentration and export from WRB sub-watersheds

Total N concentrations were very low in streams and rivers draining the Cascade Mountains ($<0.5 \text{ mg N L}^{-1}$; Figure 5). Coast Range streams had slightly greater concentrations, valley and urban streams were intermediate, and then valley streams with large proportions of agriculture had the greatest concentrations. Concentrations exceeding 10 mg N L^{-1} in fall, winter and annual flow-weighted averages were observed in Zollner Creek, where 90% of the watershed is comprised of agricultural land. Urban sub-watersheds had relatively high concentrations, over 2 mg N L^{-1} , and concentrations were similar among seasons. In contrast, differences among seasons were more pronounced in all other sub-watersheds, where concentrations were much greater in fall and winter than in the summer.

As with inputs, riverine N export varied greatly across the sub-watersheds studied, ranging over 50-fold from 1.2 to $66.3 \text{ kg ha}^{-1} \text{ yr}^{-1}$ (Figure 6a). Stream N export was very low in Cascade-draining streams; slightly greater in Coast Range-draining streams; intermediate in most valley and urban watersheds; and very high in the agriculturally-dominated catchment ($\sim 90\%$ agriculture). Winter season dominated N exports (30–60%) for nearly all watersheds, with fall and winter together accounting for 60–90% of total riverine N export (Figure 6b). Spring rates of export were third largest, and summer had the smallest export, generally less than 10% of TN export. Cascade Range-based streams had the largest proportion of N export occurring in the summer (up to 20%), with Valley and Coast Range streams having low summer export ($<5\%$).

Watershed N inputs strongly explained the variation in riverine N exports ($r^2=0.97$; Figure 7a). Since the three mainstem sites along the Willamette River at Corvallis, Albany and Portland were strong drivers of this relationship, we removed these stations from the regression. When we removed the mainstem sites, the nature of relationship changed but was still very strong ($r^2=0.89$; Figure 7b). The linear regression slope changed substantially between the tributaries vs. mainstem, as fractional export for the entire WRB was 20% but the average fractional export in the tributaries alone was 27% (Figure 7a). This decrease in fractional export may reflect additional removal in the mainstem systems.

Fractional riverine export (riverine export divided by total N inputs) for most sub-watersheds was between 8–30% (Supplemental Figure 2), with greater values observed for Zollner and Little Abiqua Creeks. The greatest fractional export (66%) was seen in Little Abiqua Creek, which had the largest contribution of N inputs from red alder BNF.

DISCUSSION

Farm fertilizer was the largest input source across much of the basin

Recent improvements in the availability of high quality, spatially explicit land cover and other important environmental data allow for better quantification of nutrient inputs to the landscape (Grohnberg and Spahr 2012; De Giralamo and others 2017; Swaney and others 2018). The areal rate of N inputs to the WRB was $38.7 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in the mid-2000s, similar to estimates for the entire US and the state of California ($\sim 45 \text{ kg N ha}^{-1} \text{ yr}^{-1}$), but much lower than inputs of $>100 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ in the Netherlands, China and Korea (Liptzin and Dahlgren 2016). Agricultural N was the largest source of N to 65% of all

HUC-8 watersheds across the conterminous US, with fertilizer, crop BNF and manure being the largest source in 41, 22 and 2% of all watersheds, respectively (Sobota and others 2013).

Our method of estimating fertilizer N inputs represents an improvement in capturing the crop-specific rates at finer spatial scales across the landscape. Previous best estimates of fertilizer inputs in the PNW used county-based sales data distributed to all NLCD crop land cover (Wise and Johnson 2013). This approach may be adequate for large areas such as states or counties (Swaney and others 2018) but assigning the same rate of input to all cropland in a county will not allow for the development of accurate input-output balances at the sub-watershed scale. One key crop group grown in the southern Willamette Valley is grass for seed, which has different species, cycles (annual vs. perennial) and N input rates. The USDA Crop Data Layer (CDL) does not distinguish between these different grass crop types that can have substantially different fertilizer inputs (see Appendix 1). Our approach allows for incorporation of crop-specific fertilization rates, which is very important in the diverse agricultural setting of the WRB where 56 crops are included in the USDA-ARS spatial data layer (Mueller-Warrant and others 2011).

The estimated input of fertilizer based on county fertilizer sales data records was strikingly similar to the high end of extension fertilizer recommendations in 2008. Total fertilizer inputs to the entire WRB based on 2008 sales data was 80,900 MT N (areal rate of 27.6 kg N ha⁻¹ yr⁻¹ to the watershed), based on the sum of twelve HUC-8 subwatershed values in the WRB for 2008 published by the International Plant Nutrition Institute (www.ipni.net). The high estimate from our method was 80,600 MT N yr⁻¹ (27.5 kg N ha⁻¹ yr⁻¹). The similarity suggests that farmers in 2008 were applying on the higher end of recommendations - but not more than extension fertilizer recommendations - in contrast to findings for other parts of the US where application far exceeds extension recommendations (Legg and others 1989; Ribaud and others 2011). It is important to note that fertilizer consumption dropped for the US in 2008, possibly in response to the 2008 financial crisis (Cao and others 2018). Thus, the agreement we observed with extension rates should be examined in other years, since the increase in price may have led to more efficient use of N fertilizer during the study period.

Other N sources played important roles in some sub-watersheds

Atmospheric N deposition often dominates total N inputs in non-agricultural areas of the US (Sobota and others 2013). Similarly, atmospheric deposition was the second largest N source to the entire WRB and represents 4–60% of total inputs among our study basins.

Atmospheric deposition was the largest N source in most of the forest sub-watersheds and was also important in urban-dominated sub-watersheds. The average rate of atmospheric N deposition to the entire basin was 3.5 kg ha⁻¹ yr⁻¹, and the highest rate was in Johnson Creek basin, 7.9 kg N ha⁻¹ yr⁻¹. Johnson Creek is the most urban watershed in this study, with 65% of the watershed categorized as developed land. Atmospheric N deposition was also dominant in the two watersheds with <1% agricultural cover, McKenzie River at Hendricks and Little Abiqua Creek. Total atmospheric N deposition in the WRB was generally lower than inputs in other areas of the US, but these rates still may affect sensitive biota in the region (Horn and others 2018). The critical load of N is the rate of N deposition

below which no harmful ecological effects occur over the long-term based on current knowledge (Pardo and others 2011). In the Northwest forested mountains ecoregion that includes the WRB, critical loads for lichens ranged from 1.2–7.1 kg N ha⁻¹ yr⁻¹, and critical loads for alpine lakes and alpine grassland were 4–10 kg N ha⁻¹ yr⁻¹ (Pardo and others 2011). Thus rates of deposition in the WRB, ranging from 2 in the forested Cascades to nearly 12 kg N ha⁻¹ yr⁻¹ near the city of Portland (Figure 3), can exceed these critical loads.

Contribution of point sources to total sub-watershed input was generally very low (<5%), except the Tualatin River basin near Portland where point source inputs were 9–12% of total N inputs. The Tualatin River experienced algal blooms in the 1980s–1990s when a Total Maximum Daily Load (TMDL) for ammonia and phosphorus was established. Work to support the TMDL also determined that wastewater effluent was an important source of N inputs to the river (Kelly and others 1999).

The third largest N source in the WRB was alder BNF, behind agricultural fertilizer and deposition. This study is one of the first to calculate N fixation by red alder at the watershed scale (along with Lin and others 2019). Alder BNF accounted for up to 49% of the N input to some WRB watersheds and supporting other work recognizing the importance of this source in western Oregon (Wise and Johnson 2011; Greathouse and others 2014). Alder BNF in the forested landscape was more important than crop BNF across most of the watersheds. In Little Abiqua Creek, red alder BNF was the dominant N source, supplying an average of 6.2 kg N ha⁻¹ yr⁻¹ across the entire watershed, the highest rate observed for alder inputs in the sub-watersheds studied here. Estimates of background BNF in the western US tend to be greater than in the northeast (Sobota and others 2009; Boyer and others 2002; Schaefer and Alber 2007). Tree-based symbiotic BNF also could play an important role in watershed N inputs for other parts of the globe where N-fixers are prominent (Cleveland and others 1999).

Crop-related BNF is the third-largest source in the US. Most crop BNF in the US is associated with soybeans and alfalfa (Sobota and others 2013), yet these crops are uncommon in the WRB. Crop BNF was <5% of inputs in the WRB watersheds studied, and generally confined to bean crops or clover in pastures.

Fertilizer applied to developed lands, usually associated with lawns, was an important flux in some areas. Non-farm fertilizer was only 2% of input to the entire WRB but was important in urban areas of the Tualatin River and was the largest N source in Portland's Johnson Creek (49% of N inputs), as observed in other metropolitan areas (Hobbie and others 2017). Forest fertilization does occur across the US, but is generally thought to have small and transient impacts on stream water quality (Binkley and others 1999). There are exceptions in areas with intensively managed plantations (e.g., Beltran and others 2010). In the WRB, much of the forest lands are in public ownership and not managed intensively, and so forest fertilization is expected to be a small contribution to watershed N budgets. However, forest fertilization could be important for watersheds dominated by private forests and is worthy of further study.

Inputs, retention and hydrology shape riverine exports

The concept of N saturation is often applied to systems with low N inputs and is generally described where N inputs in deposition can alter the processing and retention of N in terrestrial, forest, and aquatic ecosystems (Aber and others 1989; Earl and others 2006). Agricultural ecosystems become saturated with N through fertilizer inputs designed to maximize crop production (David and others 1997). In the WRB, watershed N export was strongly related to N input rates across a wide of watershed areas and land uses (Figure 7a). This strong relationship between total N inputs and riverine TN fluxes also has been observed in many other areas of the US and northern Europe (Boyer and others 2002; Howarth and others 2012; Schaefer and others 2009; Figure 8a). Export of N was greatest in the agricultural valley and urban watersheds. Theory predicts that nitrate leaching will increase exponentially after N saturation (Niu and others 2016). The proportion of N exported increased as N inputs exceeded approximately $50 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ (Figure 7b), an indication of N saturation or reduced capacity for N retention at the watershed scale above that input rate.

Seasonally, the highest N export occurred during winter and fall (60–90% of annual N export), which are periods of high hydrologic connectivity, high discharge, and low biotic uptake. This pattern is seen in Mediterranean type climates with strong seasonal precipitation (De Giralamo and others 2017); much of the N export occurs during winter months when N uptake is low but streamflow is greatest, as expected based on the hydrology of these systems (Brooks and others 2012). For many crops in the WRB, fertilizer is applied in the late winter through the mid-spring, after the winter rains have removed any remaining soil N from the previous growing season (Lin and others 2019). Thus the timing of plant uptake in spring and early summer, when runoff is low, represents the greatest disconnect between N demand and supply, resulting in uncontrolled losses of N (Perakis 2002) during the fall and winter when runoff increases. Because losses do not occur during the same period as inputs, this disconnect makes it challenging to directly connect management to changes in N export. Agricultural practices that reduce the amount of N left behind in the soil at the end of the growing season could be a way to direct management toward reducing fall and winter export (Maaz and Pan 2017), but more work is needed to connect these metrics.

Future projections for the PNW indicate warmer annual temperatures, and small changes in total precipitation with a tendency toward wetter falls and winters, more intense storms, and drier summer conditions (Mote and Salathe 2010). For urban-dominated watersheds, drier summers likely mean that wastewater may have an even greater influence on summer N concentrations, possibly creating challenges for discharge permitting during those times.

Important controls on N balances and fractional N export

The fractional rate of export in the WRB is relatively large compared to many other parts of North America (Figure 8), suggesting that greater precipitation and runoff may lead to the high fractional export seen in the WRB. Average fractional N export of total N inputs was approximately 20% of N input (slope of Figure 7a), although for just the tributaries this

number increases to nearly 29% (Figure 7b), providing support to the idea that large rivers play important roles in N removal within a river network (Wollheim and others 2006).

Our analysis of 89 watersheds across North America (Figure 8) illustrates that inputs and runoff are important and interacting drivers of N export. Stream N export increased rapidly and non-linearly in response to net N input in the WRB (Figure 8a) when compared to the northeast U.S. and Canada (Boyer and others 2002; Goyette and others 2019) and much more rapidly overall than in California and southeastern U.S. (Schaefer and Alber 2007; Sobota and others 2009). For the WRB, fractional export of NANI averaged 38%. Across all sites, fractional export of NANI increased with annual runoff until about 500 mm yr⁻¹ (Figure 8b). Above that runoff value, fractional export remained high, but there was a great deal of variation among and within regions. It is important to note that fractional N export is statistically confounded with runoff since runoff is used to calculate N export; however, the relationship between runoff and river N export per unit area was poor ($r^2 = 0.014$ across all sites) and thus we expect this confounding to have a small effect on the pattern of increasing fractional export with increasing runoff. Greater export of net N inputs in the WRB may be related to the strong seasonal patterns of N export, with most runoff-mediated N transfer occurring in the winter when there is less capacity for N uptake or removal (Lin and others 2019).

Native, non-crop N-fixers were important for N supply in some sub-watersheds, accounting for up to 49% of total N inputs. Alder cover has been shown to be an important driver of riverine N concentrations and fluxes in western Oregon (Compton and others 2003; Greathouse and others 2014) and across the PNW (Wise and Johnson 2011). Little Abiqua Creek had the highest proportion of N inputs coming from red alder biological N fixation, and had by far the highest fractional export (66%). The high fractional export may result from an underestimation of the alder N fixation input rates using our conservative approach, so we addressed this by calculating an upper bound of alder N fixation, using the rate of 200 kg N ha⁻¹ yr⁻¹ (double the conservative rate, but still within the measured range of Binkley and others [1994]). Using this upper bound for all watersheds, the fractional export of total N inputs declined to 7–46%, with Little Abiqua and Zollner Creek having fractional export rates of 45 and 46%, respectively. Little Abiqua had the highest fractional export at 110% of net N inputs (Figure 8b), likely because we are underestimating alder BNF. The high fractional N export also could be explained by the streamside location of much of the alder in Little Abiqua Creek watershed, providing inputs directly to the stream with little opportunity for terrestrial removal. Further work is needed to estimate landscape controls on alder BNF inputs in this region.

CONCLUSIONS AND IMPLICATIONS

Across the WRB, agricultural inputs dominate N inputs in most watersheds, and the fractional export of net N inputs was among the highest observed in 89 watersheds in North America. These high rates of export can contribute to local water quality issues. For example, in Zollner Creek, where 90% of the watershed is comprised of agricultural land, Oregon

Department of Environmental Quality identified Zollner Creek as exceeding state water quality standards for nitrate according to Section 303(d) of the Clear Water Act leading to a nitrate TMDL that applies to that sub-watershed year-round (Goffin and others 2010).

Our crop-specific approach for estimating agricultural N inputs represents an improvement over county and sales-based approaches for estimating N inputs that distribute fertilizer evenly to cropland across the county using NLCD (e.g., Wise and Johnson 2011; Swaney and others 2018), by incorporating spatial variability in crop types that can influence N balances for smaller watersheds. The USDA's farm resource region of the Fruitful Rim has a high diversity of crops including fruits, vegetables and grass seed, and was omitted from a large-scale assessment of agricultural N use that focused on grains, legumes and forage (Swaney et al. 2018). Similar locally derived, crop-specific approaches could be useful throughout the Fruitful Rim region, that also includes parts of California, Washington, Idaho, Texas and Florida.

N deposition and alder BNF played important roles in forested watersheds, and point sources were substantial in some urban-dominated areas. Non-farm fertilizer applied to urban lawns was important in urban watersheds near Portland. Red alder BNF had high rates and proportions of N inputs, future studies of how that translates to watershed N export is needed to better constrain our estimates of the effects of this source.

Fractional export of net N inputs generally increased with runoff in sites across North America, indicating that hydrologic forcing decreases N retention within the watershed. Projected wetter falls and winters and more extreme storm events could continue to increase fractional N export in the WRB. Analysis of the trends in N inputs and export using locally-derived, seasonal data can inform nutrient management and our understanding of the timing and rate of N delivery to aquatic ecosystems.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

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Appendix

Appendix 1.

Willamette Basin land use categories, area and fertilization rates.

Land use	Total area (ha)	Low rate (kg N ha ⁻¹ yr ⁻¹)	High rate (kg N ha ⁻¹ yr ⁻¹)
NLCD 43 evergreen forest	1,018,852	0	0
Reforestation	770,977	0	0
Pasture	174,785	100	120
Established trees other than poplars	165,459	0	0
NLCD 53 scrub/shrub	117,205	0	0
Bare ground in fall (not otherwise classed)	87,766	20	40
NLCD 23 developed medium intensity	66,272	0	0
Established tall fescue	57,575	120	180
NLCD 22 developed low intensity	54,552	0	0
Fall plant Italian ryegrass	45,726	120	160
Wheat or other cereals	38,298	100	230
NLCD 44 mixed forest	36,431	0	0
Established perennial ryegrass	36,154	150	200
Urban development	26,946	0	0
Nursery crops	24,974	0	0
Fallow	23,723	0	0
Haycrop	21,124	40	90
Christmas trees	20,728	0	150
NLCD 41 deciduous forest	20,692	0	0
NLCD 21 developed open space	17,089	0	0
NLCD ² 11 open water	15,582	0	0
NLCD 90 woody wetlands	14,199	0	0
NLCD 24 developed high intensity	13,376	0	0
Full straw Italian ryegrass (annual ryegrass)	11,415	120	160
Spring planting new grass seed stand	11,365	20	40
Established fine fescue	11,077	45	90
Fall plant perennial ryegrass	10,554	150	200
Filberts	10,386	99	115
Caneberry	8,143	50	70
Oak trees	7,004	0	0
Vineyard	6,777	0	6
Established orchardgrass	5,273	110	140
Fall plant clover	3,776	0	0
Established clover	3,475	0	0
Orchard crops (apple, cherry)	1,567	15	50
Established bentgrass	1,397	100	130

Land use	Total area (ha)	Low rate (kg N ha ⁻¹ yr ⁻¹)	High rate (kg N ha ⁻¹ yr ⁻¹)
NLCD 95 herbaceous wetlands	1160	0	0
Shrubs, wildlife refuge	1,071	0	0
Fall plant tall fescue	817	120	180
Beans - summer annual cropping	734	50	80
Meadowfoam	649	40	60
Established hops	546	100	150
Spring planting unidentified grass seed, peas, mint	393	0	0
Flowers, assorted	217	0	0
Alfalfa new planting	182	0	0
Wetlands restoration	149	0	0
Established blueberries	148	100	165
Poplars	141	0	0
Wildrice	140	0	0
Established mint	108	200	250
NT fall plant OG and no-till TF, HR, CL ¹	106	20	20
Radish, brassicas	98	50	75
Established alfalfa	80	0	0
New planting poplars, blueberries, hops	74	0	0
Volunteer Italian ryegrass grazed as pasture	70	0	0
Corn	62	40	215
Strawberries	29	25	50

¹ NLCD: National Land Cover Dataset, numbers refer to category class in NLCD.

² NT: no till; OG: orchardgrass; TF: tall fescue; HR: hybrid ryegrass; CL: clover.

Appendix 2.

List of sites and source of chemistry and streamflow data.

Watershed	Chemistry source and station number	Streamflow station	Record period
Coast Fork Willamette River	ODEQ: 11275	USGS: 14157500	1974–2009
Middle Fork Willamette River	ODEQ: 10386	USGS: 14152000	1974–2006
Mohawk River	STORET: 402340 ODEQ: 10663	USGS: 14165000	1995–2006
McKenzie River at Hendricks	STORET: 402339 ODEQ: 10662	USGS: 14163900	1995–2006
McKenzie R. at Coburg	STORET: 402044 ODEQ: 10376	Modeled: 14164900; 1416500; 14162500	1975–2009
Long Tom River	STORET: 402820 ODEQ: 11140	USGS: 14170000	1974–2009
South Yamhill River	STORET: 402625 ODEQ: 10948	OWRD: 1419400 (observed 80–94); 14194150 (area corrected 95–09)	1980–2009

Watershed	Chemistry source and station number	Streamflow station	Record period
South Santiam River	STORET: 402034 ODEQ: 10366	USGS: 14187500 +modeled Hamilton Creek + area correction	1978–2009
North Santiam River	STORET: 402469 ODEQ: 10792	USGS:14184100 modeled from 14183000	1978–2006
Clackamas River	STORET: 402913 ODEQ: 11233	USGS: 14210000 (modeled)	1974–2009
Little Abiqua Creek	USGS: 14200400	USGS: 14200400	1994–2004
Zollner Creek	USGS: 14201300	USGS: 14201300	1994–2008
Pudding River at Woodburn	STORET: 402317 ODEQ: 10640	USGS: 14202000 (modeled 93–97); 14201340 (observed 98–09)	1993–2009
Pudding River at Aurora	STORET: 402594 ODEQ: 10917	USGS: 14202000 (observed 93–97; 02–09); 14201340 (used to model 9402)	1993–2009
Molalla River	STORET: 402314 ODEQ: 10637	USGS:14210000 (modeled 91–00); 14200000 (observed 00–09)	1991–2009
Johnson Creek	ODEQ: 11321	USGS:14211550	1974–2009
Tualatin River near Elsner Rd	ODEQ:10458	USGS: 14207500	1974–2006
Tualatin R. at Boones Ferry Rd	CWS: 3701080	USGS: 14207500	1979–2006
Tualatin R. at Weiss Br	CWS: 3701002	USGS: 14207500	1988–2005
Willamette River at Corvallis	STORET:402020 ODEQ:10352	USGS:14174000	1975–2009
Willamette R. at Albany	STORET:402018 ODEQ:10350	USGS:14174000	1974–2009
Willamette R. at Portland	USGS: 14211720	USGS: 14211720	1974–2006

Appendix 3.

Approach for modeling streamflow for sites without streamflow using nearby stations.

Site (y)	Model	b	c	d	R ²
South Santiam River	$Q_y = \left(\frac{A_y}{A_b + A_c} \right) \left(Q_b + Q_d \left(\frac{A_c}{A_d} \right) \right)$	14187500	Hamilton Creek (modeled)	1416500 (Mohawk R.)	
North Santiam River	$Q_y = 1.09 * Q_b - 275$	14183000			0.98
Willamette River at Corvallis	$Q_y = \left(\frac{A_y}{A_b} \right) Q_b$	14174000			
South Yamhill River	$Q_y = \left(\frac{A_y}{A_b} \right) Q_b$	14194150			
McKenzie River at Coburg Rd	$Q_y = Q_c + Q_d$ $Q_d = (1.45 * Q_b - 1446)$	14162500	14165000	14164900	0.93
Pudding River at Woodburn	$Q_y = 0.68 * Q_b$	14202000			0.99

Site (y)	Model	b	c	d	R ²
Pudding River at Aurora	$Q_y = 1.46 * Q_b$	14201340			0.99
Clackamas River	$Q_y = 1.40 * Q_b - 474$	14210000			0.96
Tualatin River at Elsner Rd	Area correction for chemistry sampling point between two Q gauges	14207500	14206500		0.99

Q: discharge, A: area

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MANUSCRIPT HIGHLIGHTS:

- 78% of N inputs to the WRB came from agriculture, mostly as synthetic fertilizer.
- Crop-specific estimates of fertilizer can improve small watershed N budgets.
- Most of riverine export (60–90%) occurred during the fall and winter dormant seasons.

Figure 1.

Land cover across the WRB. ARS data is 2008 data adapted from the National Agricultural Statistics Service Crop Data Layer as described by Mueller-Warrant and others (2011).

NCLD is the National Land Cover data base forested land cover categories. See Appendix 1 for a complete list of crops used in the analysis.

Figure 2.
Locations of WRB stream monitoring stations used in this study (Oregon, USA). Sites are color coded by physiographic region and indicate the pour point of the 22 sub-watersheds studied.

Figure 3.

Major N inputs to the WRB from multiple sources. Sources include synthetic fertilizer applied to farmland, deposition, point sources (shown at location of input), and alder and crop biological N fixation (BNF).

Figure 4.

Total N inputs (a) and apportionment of inputs (b) to each WRB sub-watershed. Lines below figure indicate physiographic category.

Figure 5. Annual and seasonal flow-weighted TN concentrations in rivers and streams. Lines below figure indicate physiographic category.

Figure 6.
Stream N export (a) and seasonal proportion of export (b) for each WRB sub-watershed.

Figure 7.
 Relationship between a) total and b) areal N inputs and export for the WRB sub-watersheds.

Figure 8.
Riverine export across North America a) net inputs vs. export and b) runoff vs. fractional export of net inputs.

Table 1.

List of WRB study stations and associated watershed land cover data.

Chemistry Station ID	Name of river or creek at station	Category	Area km ²	Runoff mm yr ⁻¹	% Dev	% Ag	% Hdwd/mixed	% Conifer	% Other
10386	Middle Fork Willamette	Cascades	3481	958	1	1	8	87	3
402339	McKenzie at Hendricks	Cascades	2830	716	1	0	5	89	5
402044	McKenzie at Coburg	Cascades	3451	943	1	2	8	84	4
402469	N. Santiam	Cascades	1894	1170	1	7	6	83	3
402034	S. Santiam	Cascades	1852	974	1	6	11	79	3
402913	Clackamas	Cascades	2436	1543	3	7	11	78	2
11275	Coast Fork Willamette	Coast	1698	1520	3	7	14	73	2
402625	S. Yamhill	Coast	1289	571	4	30	18	47	2
402820	Long Tom	Coast	1074	1009	12	29	20	33	6
402340	Mohawk	Valley	460	1138	1	6	19	73	1
402314	Molalla	Valley	894	1059	3	20	2	62	14
14200400	Little Abiqua	Valley	25	884	0	0	23	76	2
402317	Pudding at Woodburn	Valley	817	829	8	53	11	26	2
402594	Pudding at Aurora	Valley	1247	1368	6	51	14	27	2
14201300	Zollner	Valley	37	467	5	90	2	2	1
10458	Tualatin at Elsner Rd	Urban	1641	418	16	28	19	36	2
3701080	Tualatin at Boones F Rd	Urban	1789	709	19	27	18	33	2
3701002	Tualatin at Weiss Br	Urban	1834	702	20	27	18	33	2
11321	Johnson	Urban	143	685	65	16	16	3	1
402020	Willamette at Corvallis	Mainstem	11428	879	4	12	11	70	4
402018	Willamette at Albany	Mainstem	12561	879	4	15	11	67	4
14211720	Willamette at Portland	Mainstem	28914	965	6	21	12	58	3