

Key components and contrasts in the nitrogen budget across a US-Canadian transboundary watershed

Jiajia Lin¹, Jana Compton², Chris Clark³, Shabtai Bittman⁴, Donna Schwede⁵, Peter S. Homann⁶, Peter M. Kiffney⁷, David Hooper⁸, Gary Bahr⁹, and Jill Baron¹⁰

¹The Oak Ridge Institute for Science and Education

²US EPA Corvallis

³Whatcom Conservation District

⁴Agriculture and Agri-Food Canada

⁵U.S. Environmental Protection Agency

⁶Western Washington Univ, Huxley Coll Environm, Bellingham, WA 98225 USA

⁷NOAA Fisheries Service, Northwest Fisheries Science Center

⁸Western Washington University

⁹Washington State Department of Agriculture

¹⁰United States Geological Survey

November 23, 2022

Abstract

Watershed nitrogen (N) budgets provide insights into drivers and solutions for groundwater and surface water N contamination. We constructed a comprehensive N budget for the transboundary Nooksack River Watershed (BC, Canada and WA, US) using locally-derived data, national statistics and standard parameters. Feed imports for dairy (mainly in the US) and poultry (mainly in Canada) accounted for 30 and 29% of the total N input to the watershed, respectively. Synthetic fertilizer was the next largest source contributing 21% of inputs. Food imports for humans and pets together accounted for 9% of total inputs, slightly lower than atmospheric deposition (10%). Returning salmon represented <0.06% of total N input but was an important ecological flux by importing marine-derived nutrients. Quantified N export was 80% of total N input, driven by ammonia emission (32% of exports). Animal product export was the second largest output of N (31%) as milk and cattle in the US and poultry products in Canada. Riverine export of N was estimated 28% of total N export. The commonly used crop nitrogen use efficiency (crop NUE) alone did not provide sufficient information on farming activities and should be combined with other criteria such as farm-gate NUE to understand management efficiency. Agriculture was the primary driver of N inputs to the environment despite widespread adoption of conservation practices, illustrating a need to optimize management to minimize hydrologic and volatilization losses. The N budget provides key information for stakeholders across sectors and borders to create environmentally and economically viable and effective solutions.

Hosted file

jgr-nrw-supporting information-jlin.docx available at <https://authorea.com/users/545797/articles/602154-key-components-and-contrasts-in-the-nitrogen-budget-across-a-us-canadian-transboundary-watershed>

Key components and contrasts in the nitrogen budget across a US-Canadian transboundary watershed

Jiajia Lin^{1*,2}, Jana E. Compton², Chris Clark³, Shabtai Bittman⁴, Donna Schwede⁵, Peter S. Homann⁶, Peter Kiffney⁷, David Hooper⁸, Gary Bahr⁹, Jill S. Baron¹⁰

¹The Oak Ridge Institute for Science and Education (ORISE). 200 SW 35th St., Corvallis, OR 97333. ORCID: 0000-0002-1493-2832

²U.S. Environmental Protection Agency, Pacific Ecological Systems Division, 200 SW 35th St., Corvallis OR 97333. ORCID: 0000-0001-9833-8664

³Whatcom Conservation District, Lynden, WA.

⁴Agriculture and Agri-Food Canada, Agassiz, BC, Canada. shabtai.bittman@canada.ca

⁵U.S. Environmental Protection Agency, Center for Environmental Measurement & Modeling, Research Triangle, NC. ORCID: 0000-0002-7880-0548

⁶Dept. of Environmental Sciences, Western Washington University, Bellingham, WA.

⁷National Oceanic and Atmospheric Administration, Northwest Fisheries Science Center, Seattle, WA.

⁸Dept. of Biology, Western Washington University, Bellingham, WA.

⁹Natural Resources Assessment, Washington State Department of Agriculture, Olympia, WA.

¹⁰U.S. Geological Survey, Fort Collins Science Center, Fort Collins, CO. ORCID:0000-0002-5902-6251

Corresponding author: Jiajia Lin (jlin42@outlook.com)

Key Points:

- Nearly 81% of nitrogen inputs to the Nooksack River Watershed were used to support agricultural production, most of which was animal feed
- The largest export was in the form of ammonia from the agriculture sector (32%)
- Different policy frameworks between US and Canada had impacts on components on nutrient management in different portions of the watershed

Keywords:

Nitrogen budget, transboundary watershed, ammonia emission, nitrogen use efficiency, agriculture, land use

1 **Abstract**

2 Watershed nitrogen (N) budgets provide insights into drivers and solutions for groundwater and
3 surface water N contamination. We constructed a comprehensive N budget for the
4 transboundary Nooksack River Watershed (BC, Canada and WA, US) using locally-derived data,
5 national statistics and standard parameters. Feed imports for dairy (mainly in the US) and
6 poultry (mainly in Canada) accounted for 30 and 29% of the total N input to the watershed,
7 respectively. Synthetic fertilizer was the next largest source contributing 21% of inputs. Food
8 imports for humans and pets together accounted for 9% of total inputs, slightly lower than
9 atmospheric deposition (10%). Returning salmon represented <0.06% of total N input but was
10 an important ecological flux by importing marine-derived nutrients. Quantified N export was
11 80% of total N input, driven by ammonia emission (32% of exports). Animal product export was
12 the second largest output of N (31%) as milk and cattle in the US and poultry products in
13 Canada. Riverine export of N was estimated 28% of total N export. The commonly used crop
14 nitrogen use efficiency (crop NUE) alone did not provide sufficient information on farming
15 activities and should be combined with other criteria such as farm-gate NUE to understand
16 management efficiency. Agriculture was the primary driver of N inputs to the environment
17 despite widespread adoption of conservation practices, illustrating a need to optimize
18 management to minimize hydrologic and volatilization losses. The N budget provides key
19 information for stakeholders across sectors and borders to create environmentally and
20 economically viable and effective solutions.

21 **1 Introduction**

22 The production and consumption of food and energy are increasing the cycling of
23 reactive nitrogen in the environment (Davidson et al., 2011; Galloway et al., 2004; van Meter et
24 al., 2016). While the usage of N to produce food and energy sustains human health and well-
25 being, intentional and unintentional release of excess N has led to significant ecological
26 consequences, such as eutrophication of fresh and coastal waters, hypoxia of aquatic systems,
27 contamination of drinking water, degradation of air quality, deposition-induced acidification, and
28 loss of biodiversity (Baron et al., 2011; Greaver et al., 2012; Pennino et al., 2017). Developing
29 the best available information on N sources and transport is needed at different scales to promote
30 effective management activities, yet this is a challenging task because of the wide variety of
31 sources, forms, processing and loss vectors along the N cascade (Alexander et al., 2009; Erisman
32 et al., 2003; Galloway et al., 2003).

33 One useful approach to bridge the gap between N flows and nutrient reduction goals can
34 be found by assembling integrated, multi-source, multi-sectoral N budgets for specific areas of
35 concern. The creation of a N budget is an essential step towards an integrated approach to
36 solving problems associated with N release. Input-output budgets can help decision-makers
37 better understand and manage N release by providing quantification of N fluxes at scales
38 appropriate for making management decisions. Many types of accounting approaches have
39 emerged to provide decision-makers information about N sources and loadings (such as NANI,
40 SPARROW, WSAM) (Hong et al., 2011; Sprague et al., 2000; Swaney et al., 2018). These
41 efforts have provided information at county, state and country scales, but N in the environment
42 does not follow geopolitical boundaries. Through long-range transport in the atmosphere and
43 waters, the environmental impacts of N can extend from local to regional to continental to global

44 scales, depending on the form and fate (Erismann et al., 2003; Galloway, 2003). Partnerships
45 between countries and institutions may assist in development and implementation of effective N
46 management, especially where N crosses international boundaries. Successful partnership
47 examples on other environmental issues include the Great Lakes Water Quality Agreement
48 between the United States (US) and Canada that works to develop new nutrient reduction targets
49 and explore pathways to reach the common goal (Team, 2015), and the Baltic Sea Action Plan, a
50 multinational collaboration that has made great progress in reducing nutrient inputs to the Baltic
51 (McCrackin et al., 2018).

52 Straddling the border of Washington State, US, and British Columbia, Canada, the
53 Nooksack River Watershed (NRW) supports agriculture, fisheries, wildlife, and urban
54 communities from the North Cascades to Bellingham Bay in Puget Sound, and from the Fraser
55 River towards Vancouver, BC. Agricultural land in the watershed is dominated by forage crop
56 production supporting confined animal operations (dairy and poultry) as well as berries. Land
57 application of livestock manure is a common agricultural practice as a source of nutrients for
58 crop production (Bittman et al., 2019; Cox et al., 2018). Excess N in both air and water have
59 elevated both environmental and human health risks in the watershed. Caused by enhanced N
60 emission to the atmosphere and subsequent deposition, exceedances of N critical loads were
61 observed or expected in urban and agricultural corridors in this region, which can potentially
62 lead to significant harmful effects on local species and a cascade of effects on the entire
63 ecosystem (Baron et al., 2011; Geiser et al., 2010; Greaver et al., 2012; Sheibley et al., 2014).
64 Elevated N emission can impair air quality by lowering visibility and contributing to particulate
65 matter and ozone precursors that are harmful to human health.

66 For decades, groundwater nitrate concentrations have exceeded the maximum
67 contaminant level (MCL) for drinking water (10 mg L^{-1}) in the transboundary Sumas-Blaine
68 Aquifer (SBA) (Zebarth et al., 2015). The SBA, which partially overlaps the NRW, is the
69 primary source of drinking water for the transboundary area (Carey et al., 2017) (Figure 1).
70 About 29% of private wells sampled on the US side of the SBA exceeded the MCL (Carey &
71 Cummings 2013). Recent studies have shown decreasing trends both in nitrate concentrations in
72 some wells and in the total number of monitoring wells exceeding the MCL (Carey et al., 2017),
73 but high nitrate concentrations in drinking water wells remain a concern in the area (Cox et al.,
74 2018).

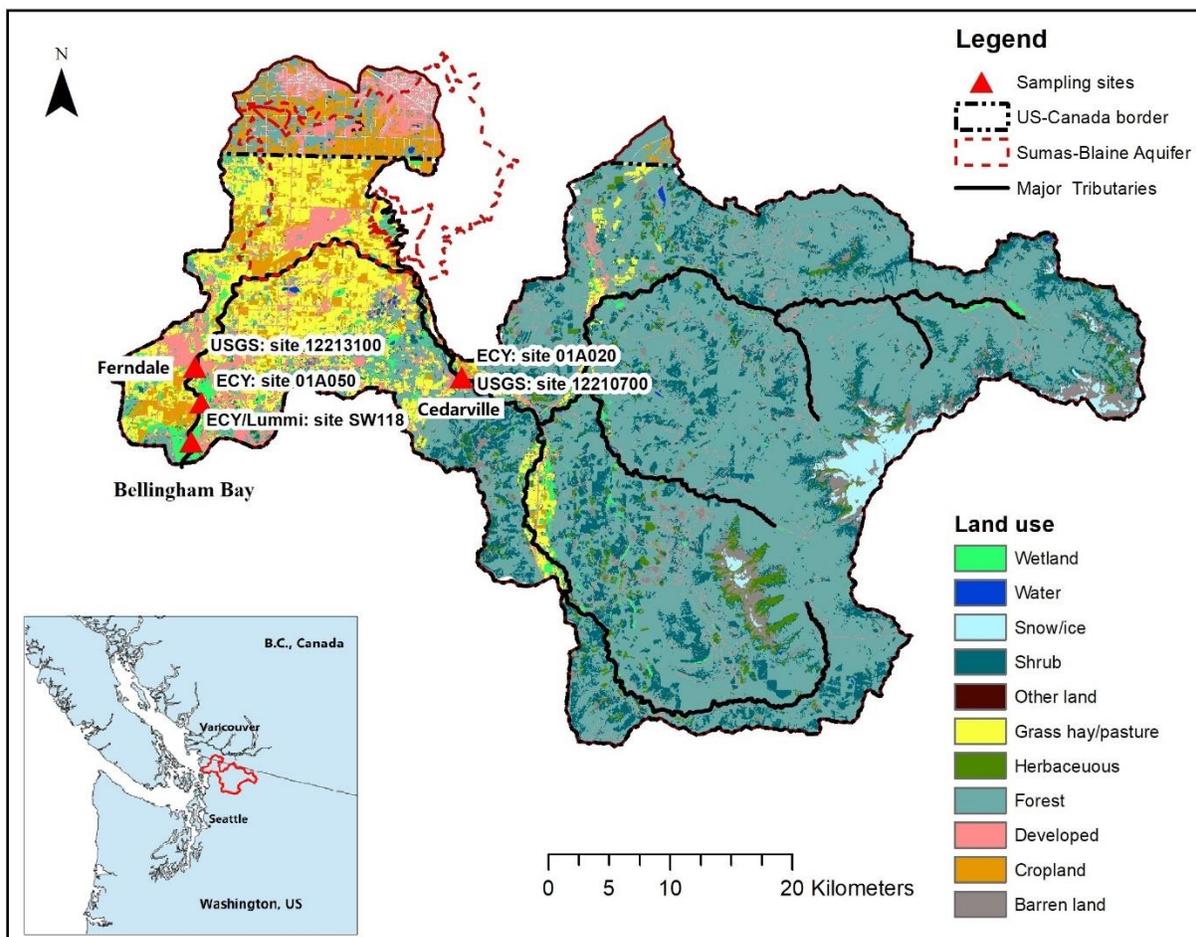
75 The transboundary nature of the watershed has complicated efforts to trace N pollutant
76 sources in air and waters and to develop effective nutrient management plans. Construction of a
77 transboundary total N budget allows us to integrate information from all sectors, compare
78 different management practices across the border, and link these activities to the environmental
79 outcomes. An informal partnership formed in 2016 between scientists and stakeholders in the
80 US and Canada to study N budgets and sustainability for the transboundary watershed. The
81 NRW N budget project is the North American demonstration for the International Nitrogen
82 Management System (INMS), which aspires to bring together scientists and communities to
83 improve nitrogen management across the globe (http://www.inms.international/about_INMS).

84 The objectives of the NRW budget study were: 1) to construct the first comprehensive N
85 -budget of the NRW using local data on N sources and exports; and 2) to combine the
86 information on cross-boundary N inputs and outputs to gain a better understanding of local N
87 retention and transport mechanisms and N use efficiencies. We hope to use the binational N
88 budget findings to facilitate future studies on how differences in management and policies affect

89 N fates in the environment, which could help create environmentally effective and economically
90 viable solutions to improve air and water qualities in the region.

91 **2 Study area**

92 The headwaters of the Nooksack River are in the western North Cascade Mountains (Mt.
93 Baker and Mt. Shuksan), and the river flows west through lowlands before discharging to
94 Bellingham Bay north of the city of Bellingham. The Nooksack River drains an approximately
95 2130 km² area of northwest Washington State in the US and southwestern British Columbia in
96 Canada. Most of the watershed area is in the US (94%; Figure 1). Mean annual discharge
97 ranges from 80 to 110 cms (Dickerson-Langer & Mitchell, 2014). The watershed climate is a
98 mixture of temperate maritime and Mediterranean-type according to the Köppen climate
99 classification (Kottek et al., 2006). About 70% of annual rainfall occurs from October to March
100 (Cox et al., 2018), and summers (July-October) are generally dry (Pelto 2015). About 80% of
101 the watershed area lies in mountainous forests dominated by coniferous trees. Urban and
102 residential land together is about 10% of the watershed area, with a total population of over
103 110,000 people. The agricultural land area in 2014 was about 174 km² on the US side and 42
104 km² on the Canadian side, comprising 10% of the land area of the watershed. In 2014,
105 cultivation of forage crops (grass and corn) together accounted for about 63% of agricultural land
106 on the US side (WSDA, 2015), while berry crops dominated on the Canadian side, accounting
107 for 80% of cropland. Much of the crop production on the US side supports dairy operations,
108 which remain an important economic component in the state despite recent declines in the state's
109 animal populations (Cox et al., 2018; USDA, 2017). In 2014, there were over 30,000 dairy cows
110 on the US side of the watershed. On the Canadian side, poultry farms were the major animal
111 production with a 2014 accumulated chicken population in the watershed of over 127 million.

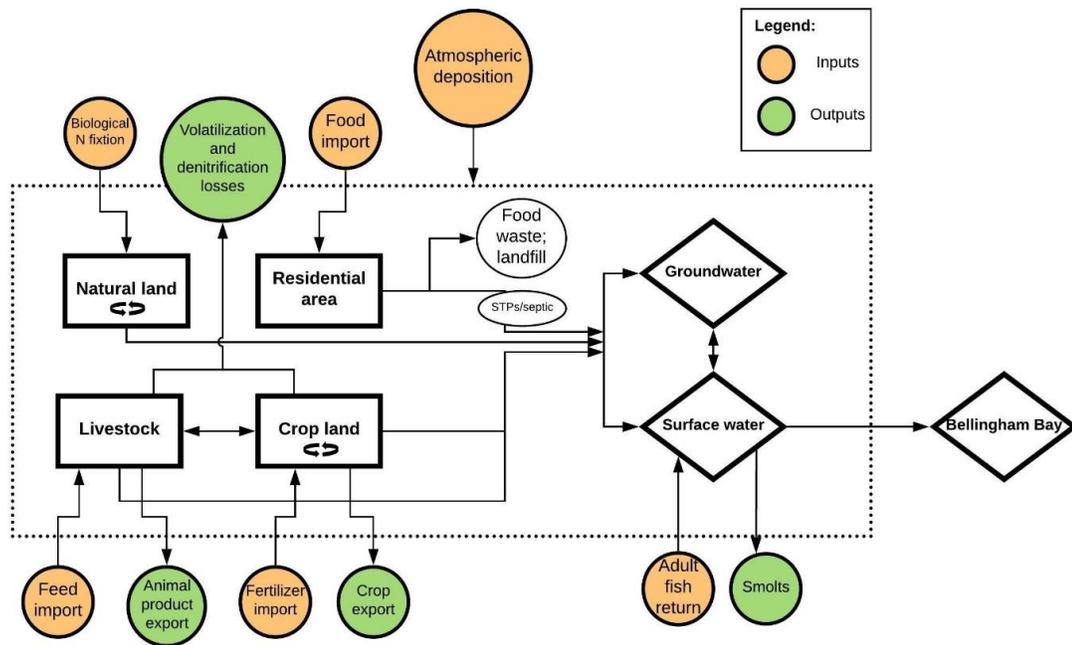


112
 113 Figure 1: The Nooksack River Watershed (NRW): Land use and its major tributaries. The Sumas-Blaine
 114 Aquifer (SBA) underlays part of the agricultural land of the NRW in both the US and Canada. Gaging
 115 station measurements and years of data are as follows: USGS Site 12213100: Daily discharge (1977-
 116 2018) and TKN concentration (1995-1998); ECY Site 01A050: Nitrate concentration (1977-2016);
 117 ECY/Lummi Site SW118: TKN concentration (2001-2018); USGS Site 12210700: Daily discharge
 118 (2004-2018); ECY Site 01A120: Nitrate concentration (1977-2016).

119

120 3 Methods

121 For this study, the US portion of the watershed will be referred to as US-NRW, and
 122 Canadian portion as Canada-NRW. Due to disparity in accessibility and forms of data between
 123 the two countries, and because of differences in their agricultural practices (e.g., animal and crop
 124 types, and regulations), several major N fluxes in the US and Canadian portions of the watershed
 125 were calculated separately using different approaches. Most of the budget results for Canada-
 126 NRW were extracted from an existing nutrient budget model and N assessment for the Lower
 127 Fraser Valley in BC that included the Canada-NRW (Bittman et al., 2019), except for the
 128 following: atmospheric deposition, food import, human waste, and food waste. For these fluxes,
 129 the same approaches and assumptions were applied to calculate these components for both the
 130 US and Canadian portions of the watershed.



131 Figure 2: Budget components and N fluxes estimated in the Nooksack River Watershed. Dotted line:
 132 Watershed boundary; Circles: N inputs (orange) and exports (green) of the watershed; size of the circles is
 133 not indicative of the flux magnitude. Squares: Watershed land components; Diamonds: Hydrological
 134 components; Internal cycles in the natural and agricultural soils involving mineralization, nitrification,
 135 immobilization, and uptake.
 136

137
 138 Table 1: Budget Components and Data Sources (for all US Components and Some of the Canadian
 139 Components) for Nooksack River Watershed.
 140

141 We integrated data from federal and state agencies and local agriculture experts with
 142 modeling results and literature values to quantify N fluxes in the NRW (Table 1). Fluxes were
 143 divided into three categories associated with input, export, and internal processes, respectively
 144 (Figure 2). More details of these methods can be found in the Supporting Information (SI). We
 145 also calculated watershed N retention and use efficiencies. We used 2014 as our target year
 146 because it was the year with the most available monitoring and survey data. When data were not
 147 available for 2014, we used data from the closest year available (Table 1).

148 3.1 N inputs

149 External N inputs to the watershed include: atmospheric deposition, food import for
 150 human and pets, feed import for farm animals, fertilizer import, and biological nitrogen fixation.
 151 Adult anadromous fish returning from the Pacific Ocean to the NRW were also calculated as an
 152 N input from outside the watershed (Figure 2).

153 **Atmospheric deposition**

154 Atmospheric deposition of total N and different forms of N in the whole watershed was
 155 extracted from simulation results of the Community Multiscale Air Quality Modeling System
 156 (CMAQ v5.2.1; <https://zenodo.org/record/1212601>) (Appel et al., 2017) at 4x4 km grid

157 resolution. Meteorology was generated using the Weather Research and Forecasting model
 158 (WRF) (Skamarock et al, 2008). The Environmental Policy Integrated Climate (EPIC) model
 159 was used to provide land use and management data to CMAQ. The CMAQ and EPIC model
 160 simulations were conducted for our study region at the National Exposure Research Laboratory
 161 at EPA using specialized emissions inputs generated by Washington State University and
 162 emissions for CAFOs from Environment Canada (Bittman et al., 2019). More details of the air
 163 quality modeling can be found in Table S1.

164 **Food import for humans and pets**

165 Food consumption by humans was calculated based on census data in both countries
 166 (Supporting Information, SI) and per capita consumption of N in food (4.7 kg N yr⁻¹). The
 167 average per capita estimate of the county was made using nutritional data by human age classes
 168 for protein (USDA & HHS, 2016). We assumed that all food was imported into the watershed as
 169 suggested by local agricultural experts. Canadian population and household census data for BC
 170 subdivisions was downloaded and clipped to Canada-NRW boundary in ArcMap 10.7 (ESRI,
 171 2011). Human food import was then calculated assuming 60% of available food N was
 172 consumed and 40% was not as a result of spoilage and wastage (Hall et al., 2009). Food import
 173 for pets was also calculated based on population and nutritional needs of dogs and cats. US Pet
 174 Ownership Statistics (AVMA, 2012) showed 37% of US households (census data) own dogs and
 175 30% cats. These pet ownership values were assigned to Canada-NRW as well. Pet N
 176 consumption was calculated by converting average body weights to energy needs then further to
 177 nutrition intakes (Table 1). We assumed the average body weights to be 20 kg for dogs and 3.6
 178 kg for cats (Baker et al., 2001).

179 **Feed import for farm animals**

180 For US-NRW, feed import was calculated as the difference between total N required by
 181 farm animals and local feed production, with the former calculated as the product of animal
 182 numbers and their nutritional needs. Dairy animal populations were estimated based on data from
 183 the Washington State Department of Agriculture (WSDA) dairy inspection program for 2014
 184 (WSDA, 2018). We downscaled USDA census data at the Whatcom county level to estimate
 185 population data for other animals. Data were downscaled based on the proportional agricultural
 186 land of the county falling within the NRW boundary. Information on the daily N intake by
 187 lactating cows was provided by local experts. Nutritional needs for other animals in US-NRW
 188 were retrieved from various primary sources (Table 1). Feed import to Canada-NRW was
 189 downscaled from the existing Lower Fraser Valley grid model quantifying nutrient flows
 190 (Bittman et al., 2019), where data on animal population and local feed acreages were derived
 191 from Census of Agriculture and the BC Ministry of Agriculture (Bittman et al., 2019).

192 **Fertilizer import**

193 For US-NRW, we calculated imported synthetic fertilizer as the difference between N
 194 ‘requirement’ of each crop and available local manure. The N ‘requirement’ term described total
 195 crop uptake of fertilizer N (both synthetic fertilizer and manure) after various losses, and was
 196 calculated as:

$$N_{crop,rqr} = \sum_{i=1}^i A_i \times F_i / f \quad Eq. 1$$

197 where $N_{crop,rqr}$ is the total crop N need (kg N yr⁻¹) in the watershed; A_i and F_i are respectively
 198 the planting area (ha) (WSDA, 2015) and recommended uptake N (kg N ha⁻¹ yr⁻¹) of crop i ,
 199 based on suggestion from local expert and extension documents (Table S2); f is a fertilizer
 200 coefficient that converts crop uptake N to total required fertilizer N (both synthetic fertilizer and
 201 manure) by factoring various losses under local conditions (Table 1; Supporting Information).

202 In Canada-NRW, fertilizer N import was extracted from the Lower Fraser Valley model,
 203 where fertilizer application was summarized from weekly application data collected from
 204 industry experts and farm surveys (Bittman et al., 2019).

205 **Biological N-fixation (BNF)**

206 For US-NRW, alder N fixation could be a substantial natural N source in the Northwest
 207 region (Compton et al. 2003; Wise & Johnson, 2011) and was calculated using the approach
 208 developed by Lin et al. (2019). A conservative annual fixation rate (100 kg ha⁻¹ yr⁻¹) (Binkley,
 209 1994) was multiplied by total alder basal area (ha), extracted from the Gradient Nearest
 210 Neighbor Structure map (Ohmann et al., 2011). Alder N fixation was not calculated for the
 211 Canada-NRW because tree species data were not available. Agricultural N fixation was not
 212 calculated because the area lacked major N fixing crops such as alfalfa, soybeans or leguminous
 213 cover crops.

214 **Anadromous fish return**

215 Return of adult anadromous salmonids from the ocean to their natal rivers and streams to
 216 spawn and die has historically been a source of marine-derived nitrogen to freshwater and
 217 riparian habitats in the Pacific Northwest (Compton et al., 2006; Gresh et al., 2000; Janetski et
 218 al., 2009). Current salmonid populations in Salish Sea watersheds are far below historical levels
 219 (Gresh et al., 2000). While some stocks are healthy, others are listed as threatened by the US
 220 Fish and Wildlife Service, and others are supported mainly by hatchery operations (Puget Sound
 221 Partnership 2017, <https://www.psp.wa.gov/salmon-recovery-watersheds.php>). We calculated the
 222 2014 N input to the NRW from returning salmon and steelhead as a function of fish population,
 223 body mass, and the N content of the fish. Average body weights and N contents of fish were the
 224 mean values from regional literature values (Table 1). Fish populations were derived from
 225 spawning ground escapement estimates provided by the Nooksack Stock Assessment.

226 **3.2 N outputs (exports)**

227 N outputs included riverine export, ammonia (NH₃) volatilization, denitrification loss,
 228 and animal and crop product export. In this study, we also included N export from smolt
 229 migration out of the watershed (Figure 2).

230 **Riverine export**

231 The US Geological Survey (USGS) Load Estimator model (LOADEST) (Runkel et al.,
 232 2004; USGS, 2013) was used to simulate riverine transport of nitrate N at two locations (Figure
 233 1): The upstream location (Cedarville) represented the upland watershed, which was
 234 predominantly forest (> 95%); model input data were daily discharge measured by USGS (Site
 235 12210700, 2004-2018; Figure 1) and monthly nitrate concentration measured by Washington
 236 State Department of Ecology (ECY; Site 01A120, 1977-2016; Figure 1). The downstream
 237 location (Ferndale) near the mouth of the River represented export from the whole watershed;
 238 daily discharge was measured by USGS at Ferndale (Site 12213100, 1977-2018; Figure 1) and

239 nitrate by ECY at nearby Brennan (ECY; Site 01A050, 1977-2016; Figure 1). Nitrate flux
 240 contributed by the lowland watershed was calculated as the difference between the whole
 241 watershed nitrate flux and upland nitrate flux.

242 Total Kjeldahl N (TKN, total organic N + total ammonia N) flux was estimated for the
 243 Ferndale location by LOADEST simulation using daily discharge data measured by USGS at
 244 Ferndale and concentration data measured by both USGS (Site 12213100, 1995-1998; Figure 1)
 245 and a collaboration between ECY and the Lummi Nation (Site SW118, 2001-2018; Figure 1).

246 **Volatilization and denitrification losses**

247 We calculated manure NH₃ volatilization in US-NRW based on National Resources
 248 Conservation Service (NRCS) estimates for Western Washington and information from local
 249 agricultural experts (Table 1 & SI): We assumed 35% pre-application volatilization loss during
 250 manure storage and housing; of what was applied in field, we assumed an average of 15%
 251 volatilization loss for both manure and synthetic fertilizer (Carey & Harrison, 2014; USDA-
 252 NRCS, 1998). Volatilization in Canada-NRW was extracted from the Lower Fraser Valley
 253 model results based on proportional agricultural land area. Denitrification loss was estimated to
 254 be 10% of applied manure and synthetic fertilizer in the entire NRW (USDA-NRCS, 1998).
 255 Denitrification in natural lands was not calculated and assumed to be part of N retention.

256 **Crop product exports**

257 In US-NRW, crop removal of N was calculated based on crop removal rate (extension
 258 documents, local expert, survey, and scientific literature, see Table 1), crop N content, and crop
 259 area (WSDA), as shown in Eq. 2:

$$N_{crop,rmv} = \sum_{i=1}^i A_i \times Y_i \times (1 - m_i) \times n_i \quad Eq. 2$$

260 where $N_{crop,rmv}$ is the total crop removal of N (kg N yr⁻¹) of the watershed; A_i and Y_i are
 261 respectively the planting area (ha) and yield (kg crop mass ha⁻¹ yr⁻¹) of crop i ; m_i is the moisture
 262 content (%) of crop i , and n_i is the N content (%) of crop i on a dry weight basis. N export in
 263 crop product for Canada-NRW was derived from the Lower Fraser Valley model, where crop
 264 export was computed as harvest removal in berries (raspberries and blueberries), the dominant
 265 export cash crop in this part of Canada (Bittman et al., 2019). Export of forest product was not
 266 calculated.

267 **Animal product exports**

268 Because most of the milk, eggs and other products produced in the US-NRW were not
 269 consumed locally, animal product export was calculated as the sum of N in milk and eggs and N
 270 export of live animals. Data on production rates, animal populations, and N contents were from
 271 USDA and WSDA (Table 1). Animal product export of N from Canada-NRW (mainly N in meat
 272 and milk products from poultry, dairy, and pigs) was estimated from downscaled data from the
 273 Lower Fraser Valley model results on milk and meat N exports.

274 **Smolt export**

275 Smolts are juvenile salmon migrating from rivers to the ocean. We included smolts from
 276 both natural and hatchery origins. Smolt mass was estimated based on fork length (the length of
 277 a fish from its nose to the middle caudal fin rays) data and length-to-weight equations. Smolt

278 mass was then coupled with N content and population data to calculate total smolt N export.
279 Data and equations were provided by the Skagit River System Cooperative, the Lummi Nation,
280 and literature review (Table 1).

281 3.3 N internal processes

282 **Sewage treatment plants and septic export**

283 We treated most of the N fluxes in sewage treatment plants and septic systems as internal
284 transfers under the assumption that releases from these sources either went into soil and/or
285 groundwater retention, or surface water fluxes (Figure 2). Total nitrogen (TN) load from sewage
286 treatment plants draining to the Nooksack River was calculated as the product of observed and
287 extrapolated effluent discharge and TN concentration (SI). When a measurement was missing at
288 certain sewage treatment plants, TN load was extrapolated based on the population size served.
289 There was no sewage treatment plant outlet within the Canada-NRW boundary, therefore,
290 sewage effluent in Canada-NRW was counted as N export that left the watershed. To estimate
291 septic inputs to the whole watershed, the population not on sewage was multiplied by an average
292 per capita waste rate (4 kg N yr^{-1}) (USEPA, 2002) and 91% septic leaching rate (USEPA, 2002).
293 In US-NRW, the ratio of population on sewage and population on septic system was about 2:3.
294 The same ratio, which was also applied to Canada-NRW.

295 **Food waste**

296 Food waste was estimated to be 40% of the available food supply based on Hall et al.
297 (2009). We assumed that all food waste was part of N retention and went to landfills, which in
298 the long term can be subject to volatilization and/or other losses that we were unable to quantify
299 in this project.

300 Crop application of dairy manure Annual manure application was calculated based on
301 animal populations, excretion rates, and pre-application emission losses. In US-NRW, the total
302 crop N 'requirement' and proportional application of manure vs. synthetic fertilizer were
303 provided by local farmers for each crop type. Pre-application volatilization loss was taken into
304 account to calculate total manure required. This value was then compared with dairy manure
305 excreted to decide if there was a net import or export of manure fertilizer. Manure application in
306 Canada-NRW was extracted from the Lower Fraser Valley model, where excretion rates were
307 computed as the difference between N fed based on industry data and N in animal products
308 (Bittman et al., 2019).

309 **Crop to animal feed**

310 We assumed all the feed crops were retained in the watershed and used as local animal
311 feed. Local production of silage corn and grass hay provided about 50% of the dry matter
312 required by lactating cows, with the other 50% of their feed was imported as soybean and alfalfa
313 required for milk production. The remaining US-NRW feed crops were used to feed other
314 livestock. In Canada-NRW, all local feed was consumed by dairy cows based on the Lower
315 Fraser Valley model (Bittman et al., 2019), and thus we calculated local feed as the difference
316 between total feed required and the imported feed for cows (Bittman et al., 2019). Total feed
317 required was estimated based on surveyed cow populations and their nutritional needs for N, and
318 the proportion of feed from import was acquired from a previous survey (Bittman et al., 2019;
319 Sheppard et al., 2010).

320 3.4 N retention and use efficiency

321 N retention was defined as the amount of annual N inputs remaining in the watershed
322 after accounting for removal via known pathways such as riverine, gaseous, and agricultural
323 exports. Fates of N retention include storage in plant and animal tissues, soil and groundwater,
324 and landfill, but may also include unaccounted losses.

325 We calculated crop N use efficiency (NUE) as the ratio of crop N harvest removal and
326 the sum of manure and synthetic fertilizer N applied. We also calculated NUE for production in
327 the whole watershed using two methods: 1) the farm-gate method calculated NUE as the ratio of
328 N removed off-farm in products vs. total N inputs to the entire watershed (Ovens et al, 2008),
329 and 2) 'commercial' whole-farm NUE was the ratio of N in crop and animal products over the
330 import of feed and fertilizer N only (Bittman et al., 2016). The crop NUE helps interpret
331 efficiency of cropping systems and potential losses, though losses to other pools (e.g., ground
332 and surface water) are not explicitly separated from N storage in soils and plant parts not
333 removed in harvest (residues, root tissue, etc.). Farm-gate NUE provides critical information on
334 both agronomic efficiency and environmental risks for the whole watershed, and has been used
335 as a policy instrument and the basis of regulation of farm nutrient levels and losses (Ovens et al,
336 2008; van der Meer, 2001). The 'commercial' whole-farm NUE method excludes 'free' N inputs
337 and mitigates the need to account for inputs beyond the farmers control such as deposition and
338 fixation (Bittman et al., 2016; Buckley et al., 2016). It also helps with the assessment of
339 economic consequences.

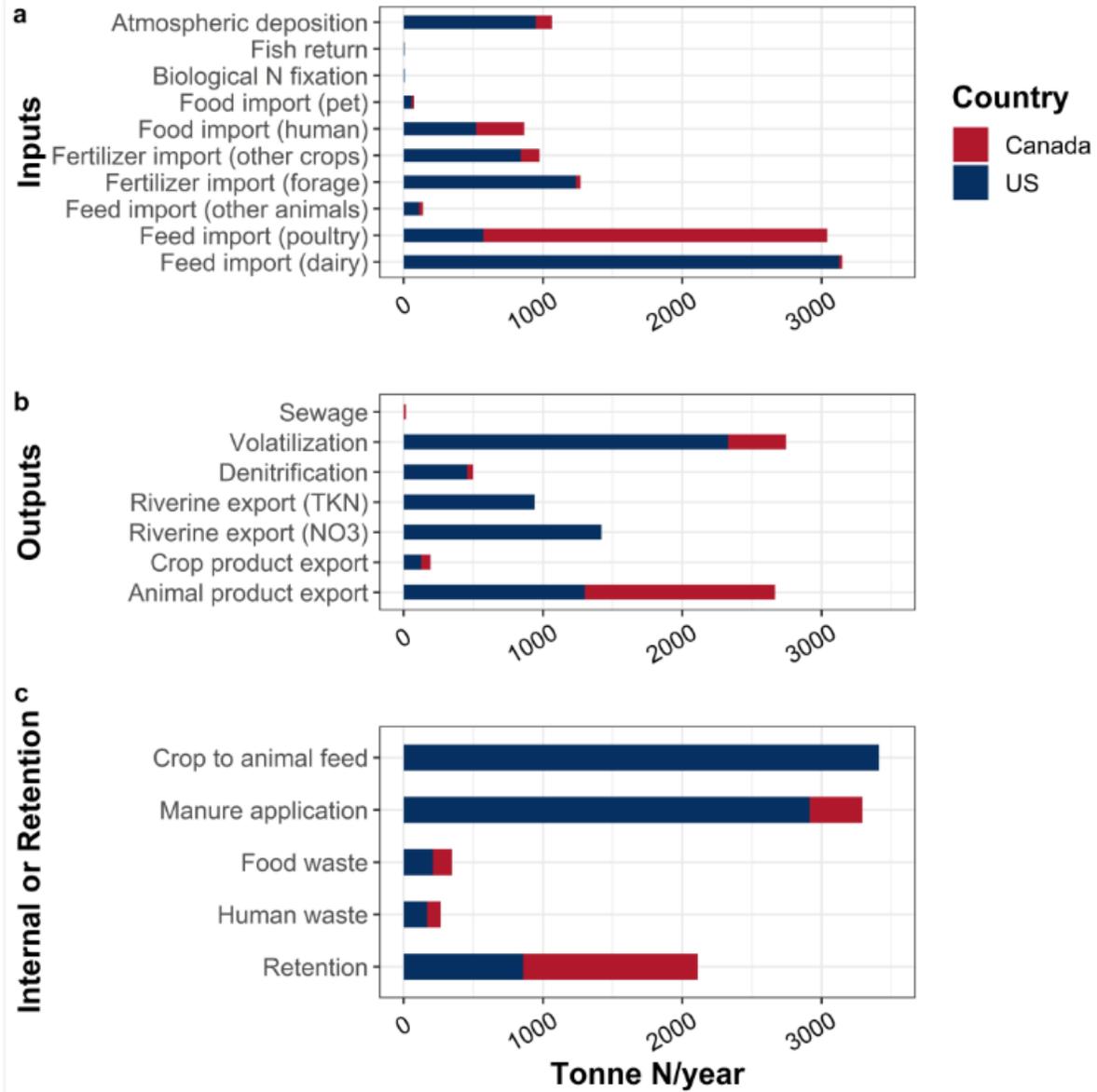
340 4 Results

341 4.1 N inputs

342 N imported as animal feed and synthetic fertilizer was about 8,600 tonnes N yr⁻¹ in total
343 and contributed 81% of N influx to the watershed (Figure 4; Table S3). The largest influx of N
344 was animal feed accounting for about 58% of N inputs to the watershed. Feed imports
345 supporting dairy and poultry production were nearly equal for the entire watershed, accounting
346 for 30% and 29% of total NRW input, respectively, with most of the dairy production on the US
347 side, and much of the poultry production on the Canadian side (Table S3). In the US-NRW,
348 imported feed for dairy cows was more than 3,100 tonnes (metric ton) N yr⁻¹, making up 42% of
349 US-NRW N input. In the Canada-NRW, annual dairy feed import was about 21 tonnes N yr⁻¹
350 representing < 0.7% of Canada-NRW N input, while imported feed for poultry was over 2,400
351 tonnes yr⁻¹ representing 78% of Canada-NRW N input. On the watershed level, annual import of
352 over 2,200 tonnes synthetic fertilizer was the second largest N source representing 21% of total
353 input. About 57% of imported fertilizer was applied to feed crops (grass hay and corn silage)
354 and the rest was applied to other crops.

355 Other sources of N contributed approximately 19% of N inputs to the watershed (Figure
356 3&4). Atmospheric deposition contributed 10% to the total N input: about 4% was deposited on
357 urban and agricultural lands and 6% was deposited on upland forest. Food imports for humans
358 and pets contributed about 8% and 1%, respectively. Alder fixation and marine-derived return of
359 adult anadromous fish both represented about <0.07% of N inputs each. Smaller amounts of
360 these inputs in Canada-NRW than US-NRW arose because of smaller proportions of land area
361 and total population in the former than the latter (Figure 3).

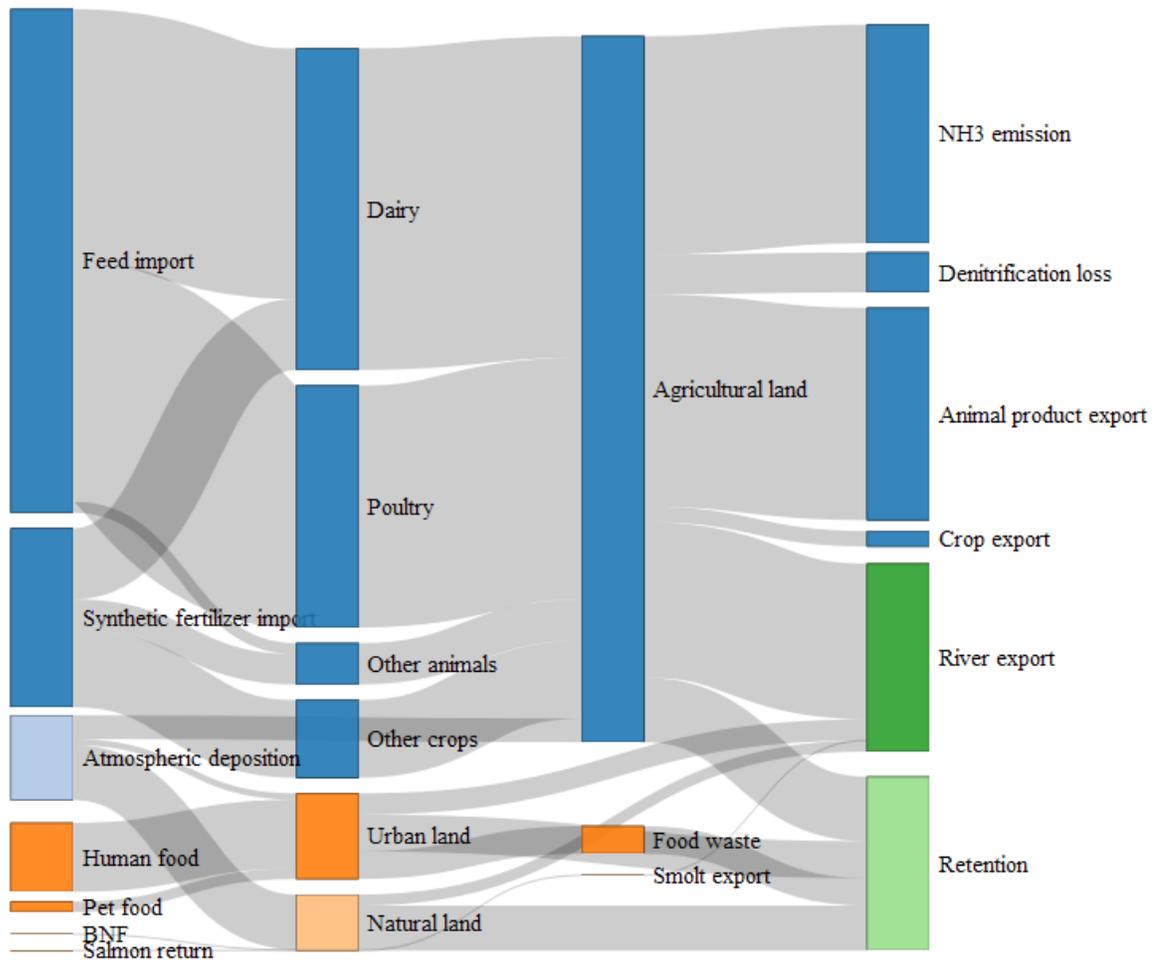
362



363
364

365 Figure 3: Annual N fluxes of the Nooksack River Watershed (NRW). a. Inputs; b. Outputs. ‘Sewage’
366 refers to N in the effluents from sewage treatment plants in Canada-NRW that drained out of the
367 watershed; c. Internal fluxes or N retention. Retention includes storage in groundwater, soil, biomass and
368 unaccounted N losses. ‘Human waste’ refers to N in sewage effluents in US-NRW and septic fluxes in
369 both Canada-NRW and US-NRW. Forage is defined as crops for animal feed.

370



371
 372 Figure 4: N fluxes in the Nooksack River Watershed. Grey bars represent N fluxes flowing from external
 373 inputs on the left side to internal cycling in the middle and then export/storage on the right side. Different
 374 colors represent different sectors – dark blue is agriculture, light blue is deposition, orange is residential
 375 fluxes and green represents river export and retention within the basin on an annual timestep. Bar height
 376 is proportional to the magnitude of the N flux.

377 4.2 N exports

378 The largest N export was NH₃ volatilization loss, with an estimated 2,745 tonnes N yr⁻¹
 379 and 32% of total export (Figure 4; Table S3). Nearly three quarters of the NH₃ volatilization was
 380 associated with dairy manure in US-NRW, and most volatilization (78%) occurred during
 381 manure storage and housing processes whereas 22% occurred after field application.
 382 Volatilization loss associated with poultry manure application was only 10% of total
 383 volatilization loss. Export of N in animal products was the second largest flux from the
 384 watershed, contributing 2,666 tonnes N yr⁻¹ or 31%. Milk was the primary product in US-NRW
 385 and poultry products (meat and eggs) dominated Canada-NRW export (Table S3).
 386 Denitrification (as N₂ and N₂O) associated with the application of manure and synthetic
 387 fertilizers accounted for 6% of total N export. In comparison, crop export was relatively small
 388 accounting for only about 2% of N export, with 126 tonnes N from exporting various horticulture
 389 crops in US-NRW and 64 tonnes N from berry production in Canada-NRW annually.

390 Hydrological export was another major pathway for N leaving the watershed, responsible
 391 for 28% of all N loss. In 2014, the Nooksack River transported 1,420 tonnes of NO₃-N and 940
 392 tonnes of TKN into Bellingham Bay (Figure 3). Nitrate was thus approximately 60% of the
 393 riverine N export. The upland watershed (Cedarville) contributed about 750 tonnes NO₃-N, 53%
 394 of the total riverine nitrate export. Export of N via smolt migration out of the watershed was less
 395 than 0.001% of the total N export.

396
 397

398 Table 2: Nitrogen use efficiencies (NUEs) in the Nooksack River Watershed

399

400 4.3 N retention, internal fluxes, and nutrient use efficiency (NUE)

401 The watershed N balance or N retention, calculated as the difference between inputs and
 402 exports, was about 2,130 tonnes N or about 20% of total N inputs (Figure 3; Table S3). This
 403 may include potential losses to groundwater, which we did not quantify. The 20% N retention
 404 may include accumulation in biomass of perennial crops (e.g., berries) and natural vegetation
 405 (e.g., forests), in biomass of animals (humans, pets, and stock), and in soils. It also may include
 406 other fluxes such as NO_x emission and natural denitrification that we could not quantify at this
 407 scale.

408 The largest internal N fluxes were associated with dairy production. Locally-grown feed
 409 provided about 1,767 tonnes N to dairy farms in US-NRW to support forage production for
 410 nearly 35,000 cows. About 2,554 tonnes N from dairy cow manure was applied on the US side.
 411 N fluxes associated with dairy cows on the Canadian side were smaller than their US
 412 counterparts and much lower than poultry manure application. In Canada-NRW, local crop feed
 413 provided 100 tonnes N for 1,323 dairy cows, and about 96 tonnes N from dairy manure was
 414 applied to crops; in comparison, 269 tonnes N in poultry manure was applied to crops. Internal
 415 N flux associated with human waste was only 2.4% of inputs or 265 tonnes in the NRW.
 416 Unexpectedly, estimated N flux in food waste was 346 tonnes for the entire watershed, slightly
 417 higher than N flux in human waste.

418 Crop NUE for the entire watershed was 51% regarding total manure and fertilizer (pre-
 419 volatilization loss). However, crop NUE for the watershed was at 67% for applied manure and
 420 fertilizer, and was greater in the US-NRW (71%) than in Canada-NRW (31%). Using the farm-

421 gate method (Ovens et al., 2008), we estimated that about 27% of total N input to the entire
422 watershed was transferred into final crop and animal products, all of which was exported from
423 the watershed (2,860/10,594 tonnes). Because all animals were transported elsewhere for
424 slaughter, we assumed no N retention due to slaughtering and rendering processes. Using the
425 ‘commercial’ whole-farm method (Bittman et al., 2016), we found the crop and animal product
426 export equaled 33% of feed and total fertilizer inputs for the entire watershed, 24% for US-NRW
427 (1427/5889 tonnes), and 53% for Canada-NRW (1433/2685 tonnes). In addition, animal
428 products equaled about 36% of total feed and fertilizer (for feed crops) inputs for the watershed,
429 with poultry products accounting for 43% of poultry feed import and milk export accounting for
430 29% of feed and fertilizer inputs to the dairy system.

431 **5 Discussion**

432 **5.1 Inputs and internal cycling**

433 N inputs were high on both the Canadian and US portions of the watershed. Input rates
434 averaged about 50 kg N ha⁻¹ yr⁻¹ across the entire NRW, comparable to the state of California
435 and the entire US (45 kg N ha⁻¹ yr⁻¹), but smaller than the Netherlands or China (100 kg N ha⁻¹ yr⁻¹)
436 (Liptzin and Dahlgren 2016). Most of these inputs were concentrated in the lower valley of
437 the NRW (23% of the area). N imports were largely related to agriculture, primarily animal
438 production – either directly as feed for poultry (Canada) or dairy cows (US) or as fertilizer for
439 cow forage. Much of the manure produced by animals was applied to crops in both the US and
440 Canadian portions of the watershed. This application reduced the need for synthetic fertilizer
441 and provided an important way to recycle feed N within the watershed. However, given the
442 quantities of N imported for animal feed, application of substantial quantities of manure to the
443 relatively small land base provided opportunity for inadvertent N losses. Relative to other
444 sources of externally derived N, background sources (i.e., N fixation in natural lands and salmon
445 returns) were each <1% of inputs at the NRW scale.

446 The high inputs were used with relatively low efficiency (Table 2). The 67% crop NUE
447 for the watershed regarding applied manure and fertilizer was lower than US national average
448 crop NUE of 70% (Zhang et al., 2015). Crop NUE was higher in US-NRW than in Canada-
449 NRW (Table 2). This could be caused by the relative amounts of manure applied. US farmers
450 have higher numbers of regulations and rules under local and state efforts to reduce the
451 agricultural loading of nitrate to the environment (Cox et al., 2005), therefore they were more
452 likely to follow extension recommendations on fertilization rates. Another major reason for
453 different overall crop NUEs was based on crop types. Berries were the dominant crop types in
454 Canada-NRW and had low N content of about 0.1% in the exported fruits, while forage grass and
455 corn, harvested 4-6 times per year as dairy feed, had N content as high as 3%. Crop NUE alone
456 does not provide comprehensive information on farming activities, and should be combined with
457 other criteria such as farm-gate NUE to understand management efficiency.

458 In contrast to crop NUE, both the farm-gate and commercial whole-farm NUEs were
459 higher in Canada-NRW than in US-NRW (Table 2). This could be attributed to a higher feed to
460 animal product ratio of the US-NRW dairy system (2.7:1) compared to that of the poultry
461 production in Canada-NRW (2:1). A recent Lower Fraser Valley study showed that using
462 rendering products as poultry feed was a very effective reuse of local N and could improve NUE
463 in British Columbia (Bittman et al., 2019), but using rendering products is prohibited in dairy
464 production due to health concerns. In addition to animal types, stocking rate can also have

465 important consequences for NUEs (Powell and Rotz, 2015): the dairy stocking rate in Canada-
466 NRW was about 1 cow acre⁻¹, whereas in US-NRW it averaged 1.3 cow acre⁻¹ for all forage crop
467 land and 1.8 cow acre⁻¹ for some crop land with high management intensity where most dairy
468 cattle were kept. The farms with lower stocking rate required less feed import since local feed
469 production was sufficient, which resulted in higher whole farm NUEs (Bittman et al., 2019).

470 5.2 Release of N to the environment

471 Loss of over 50% of N inputs to the environment, primarily as volatilized ammonia and
472 hydrological N exports to surface water and groundwater, has a strong potential to adversely
473 affect human health and the environment (Townsend et al., 2003). Ammonia, predominantly
474 from losses related to housing and storage of manure, can contribute to regional smog and odor
475 problems (Barthelmie & Pryor, 1998; Kotchenruther & Taylor, 2014), and can harm human
476 respiratory health (Paulot & Jacob 2014) Enhanced N deposition resulting from elevated N
477 emissions can cause significant damage to terrestrial and aquatic ecosystems, including cation
478 leaching, altered nutrient stoichiometry in streams and lakes, and changing biodiversity (Clark et
479 al., 2018; McMurray et al., 2013).

480 Annual riverine N export for the NRW was 28% of total N input, which may contribute
481 to current and future eutrophication and hypoxia in Bellingham Bay (Khangaonkar et al., 2019;
482 Mohamedali et al., 2011). TKN accounted for 40% of NRW riverine N export, indicating
483 substantial surface input from organic N and ammonia, potentially originating from soils rich in
484 organic matter and anthropogenic N (Bronk et al., 2007; Kroeger et al., 2006). Hydrologic
485 export that primarily occurs during the cool, wet seasons when there is low biotic removal
486 potential poses a substantial challenge to nutrient management (Compton et al., 2019; De
487 Girolamo et al., 2017; Welter & Fisher, 2016). Wet season precipitation and rising groundwater
488 levels were also linked to high seasonal soil nitrate concentration, which could lead to elevated N
489 loading to ground waters and high nitrate levels in the aquifer (Carey, 2017; Cox et al., 2018).

490 Both the forested upland and the agriculturally influenced lowland make substantial
491 contributions to the riverine N export. The lowland comprised 24% of the entire watershed, was
492 66% agricultural land, and contributed 47% of the riverine NO₃-N export. The upland watershed
493 comprises 76% of the whole watershed, was >95% forest, and contributed 53% of the riverine
494 NO₃-N export. Forest edges, which have been increasing as a result of forest fragmentation, may
495 function as nutrient traps and concentrators (Weathers et al., 2001), particularly for ammonia
496 emissions. This phenomenon may influence the forest riverine N export. Our results indicated
497 the importance of forest management to downstream water quality and nutrient balance.

498 We did not directly quantify N flux to groundwater due to its complexity and instead
499 included it as part of watershed N retention, but we acknowledge that some portion of the N
500 applied leaches into groundwater. For example, rates of nitrate leaching from the soil were
501 substantial below raspberry fields in the area (80-240 kg N ha⁻¹ yr⁻¹) (Loo et al., 2019).
502 Combining crop area data with published soil nitrate data in this area or in watersheds with
503 similar land use and weather, we did a back-of-envelope estimation of the range of N flux
504 leaching under different land uses. Based on nitrate leaching rates under raspberry field (Loo et
505 al., 2019) and post-harvest soil survey of different crops in South Abbotsford and West Sumas
506 (Sullivan & Poon, 2016), townships that are located in northern NRW, we estimated about 260 -
507 430 tonnes N entered the groundwater annually in Canada-NRW, assuming about 80% of post-
508 harvest soil nitrate-N was lost to leaching (Carey, 2002). Previous studies showed that nitrate

509 leaching following dairy manure application on forage crop land ranged between 32 and 153 kg
510 N ha⁻¹ depending on fertilization rate (Demurtas et al., 2016; Paul & Zebarth, 1997; Tarkalson et
511 al., 2006). Hence, potentially there was about 930-1,100 tonne N leaching under forage crop
512 land in US-NRW, given that 70% of the forage land there was managed with high intensity.
513 This represents about 9-10% of all N inputs. These results cannot be viewed as a complete
514 quantification of groundwater N flux in the watershed, yet they provide insights about the
515 potential N contamination of groundwater. We estimated that N loss to groundwater could
516 represent about 56-72% of N retention in the NRW.

517 Much of the applied N could be incorporated in soil organic matter and remain in the soil
518 for many years to contribute to future risk of contamination of water resources (Sebilo et al.,
519 2013). Studies have shown that legacy nutrients can become a dominant and long-term (>10 yr)
520 source of excess nutrients in many intensively managed watersheds (Chen et al., 2018; van Meter
521 et al., 2016). Groundwater N might eventually contribute to surface water export over time,
522 directly through irrigation using groundwater or indirectly as the groundwater flowpaths emerge
523 in streams.

524 Even though the study area contained a small portion of urban land, management of food
525 and food waste could represent an opportunity to reduce N loss based on our budget results. The
526 food waste portion was slightly greater than the sewage treatment plant contribution in the
527 watershed (Figure 3). Some of the negative impacts of excess N due to food production could be
528 partially addressed by reducing food waste and dietary N footprints in urban areas (Shibata et al.,
529 2017), which must include community collaboration. For example, systems thinking can support
530 an integrated agricultural and food system to optimize food utilization, and technologies can help
531 improve the efficiency of using food waste for biogas and compost (Halloran et al., 2014).
532 These efforts need to be promoted through partnerships among the government, society groups,
533 and industry

534 5.3 Implications for effective N management

535 Enhancing both dairy and cropping efficiencies are vital to achieving effective nutrient
536 management (Harrison, 2007). On the US side of the watershed, there have been many
537 conservation efforts by local and state agencies aiming at improving N management efficiencies
538 and reducing agricultural loading of nutrients to the environment. For example, Whatcom
539 County adopted a Manure Control Ordinance that restricted field manure application timing for
540 forage production to April through September to reduce leaching during wet seasons (Cox et al.,
541 2005). Whatcom Conservation District and USEPA developed a Progressive Manure
542 Application Risk Management (ARM) System, a decision-making tool using real-time field and
543 weather information, to help guide manure applications and reduce manure losses (Embertson,
544 2016). Washington State also mandated the development of Nutrient Management Plans for all
545 dairy farming operations that handled more than 700 dairy cattle. These initiatives may lead to
546 important reductions in N release to the environment for the NRW in the future.

547 As the major N loss pathway in the NRW, ammonia emission is controlled by multiple
548 factors such as livestock and manure management systems (Sanchis et al., 2019). Previous
549 research found that it was necessary to shift from single-stage emission abatement options
550 towards a whole-chain perspective (Sajeev et al., 2019). In the NRW, livestock housing and
551 storage was a major source of ammonia emission. Reducing volatilization loss during this stage
552 can be achieved by quantitatively understanding of the effect of temperature, wind speed,

553 relative humidity and ventilation rate on ammonia release rates from dairy cattle housing
554 (Sanchis et al., 2019). Moreover, multiple mitigation strategies can be combined at different
555 stages (housing, storage, and application) to reduce overall whole-farm emission, for example,
556 frequent removal of manure, anaerobic digestion, and manure acidification were all found
557 effective in reducing emissions (Sajeev et al., 2019). Adjusting cattle diet such as lowering
558 dietary crude protein were also associated with decreases in ammonia emissions rates and
559 emission as a percentage of N intake (Liu et al., 2017). Subsurface application of dairy slurry
560 can also decrease ammonia volatilization compared to surface application (Saunders et al.,
561 2012).

562 The potential contribution of nitrate leaching under agricultural land in the watershed is
563 substantial. Increasing manure application rates were associated with higher leaching in the
564 dairy system in the region (Hill, 2013; Paul & Zebarth, 1997) To improve N management on
565 agricultural lands in this area, efforts should not be limited to forage crops that were most
566 commonly associated with dairy farms, because high leaching rates were also measured under
567 berries and vegetable crop lands (Loo et al., 2019). Nitrate leaching under the same land use can
568 also vary widely in response to variations in climate factors, management practices and soil
569 properties (Loo et al., 2019). Different N treatments can be imposed on cropping systems to
570 reduce nitrate leaching. For example, the use of nitrification inhibitor dicyandiamide and/or
571 biochar was found successful in reducing nitrate leaching (Di & Cameron, 2002; Lehmann &
572 Joseph, 2009). Switching fertilization types (such as using compost) can also help reduce
573 leaching (Basso & Ritchie, 2005). There were also seasonal variations: nitrate leaching during
574 the growing season may be minimal compared to leaching losses that occur between the harvest
575 of one crop and the planting of the next (Basso & Ritchie, 2005). Cover and relay crops could
576 help minimize N leaching during the winter depending on conditions (van Vliet et al., 2002).
577 Any nutrient reduction strategies developed should account for the strongly seasonal hydrology
578 of this area.

579 Integrated nutrient management should also focus on reducing imports and seeking
580 export opportunities for excess nutrients. Harrison et al. (2012) suggested that the most effective
581 approach should include accounting of managed nutrient imports and exports from the farm, and
582 the estimation of on-farm excess (or deficits) of nutrients. Decreasing stocking rate (animal per
583 unit of land) can help reduce imports of both fertilizer and animal feed. Higher animal stocking
584 rates placed more challenges on nutrient management, since high animal densities resulted in
585 higher expenses for feed import and also higher excretion rates and ammonia loss rates (Powell
586 & Rotz, 2015). Lower stocking rates can also represent more land area being converted to
587 agriculture, representing an extensification (van Grinsven et al., 2015). Planting N-fixing cover
588 crops can also help reduce the usage and import of fertilizer. Transporting excess manure offsite
589 to be used as fertilizer elsewhere can help with the overapplication issue and reduce emission
590 and leaching losses.

591 Harrison et al. (2012) suggested that strategies and technologies to achieve N reduction
592 vary in their degree of economic feasibility and environmental impact. Site-specific and cost-
593 effective Best Management Practices (BMPs) can only be developed with the collaborations of
594 farmers, agencies, and scientists. Continuous soil and groundwater monitoring programs can
595 help establish quantifiable solutions. Temporary lack of water quality improvements cannot be
596 interpreted as a failure of the BMPs without knowing the residence time of groundwater and
597 associated soil conditions, because accumulated organic matter mineralizes gradually over time

598 and can cause lags in soil and groundwater quality improvements (Carey, 2002; Sebiló et al.,
599 2013; van Meter et al., 2016; Wassana et al., 2006).

600 5.4 N budget uncertainties

601 The integrative NRW-N budget helps us understand N cycling in the watershed and can
602 be used as an environmental performance indicator to guide future nutrient management; Still,
603 major uncertainties in our assessment could arise from several issues:

- 604 1) There was limited information about specific farm practices such as total manure
605 application rates and methods on each farm, which was regarded as confidential business
606 information. It may have resulted in inaccurate representations of the agroecosystems
607 and nutrient flows into and out of the watershed (Oenema et al., 2003).
- 608 2) Even though we attempted to capture most key sectors in the NRW, we did not estimate
609 N fluxes from some other components in the N cycle. For example, forest fertilizers on
610 private land, seed inputs or N-containing deicer used at the Abbotsford airport in the
611 Canada-NRW (personal communication: Environment and Climate Change Canada), or
612 N influx from migrating birds. Where studied, these fluxes have generally been a small
613 proportion of N input budgets (McBroom et al., 2008; Olson et al., 2005). We also may
614 underestimate denitrification and volatilization losses by not accounting for emission
615 sources other than fertilizer and manure.
- 616 3) Generalization about certain processes could result in further computational errors. For
617 instance, we used average denitrification (10%) and volatilization (35% pre-application
618 and 15% post-application) loss rates for manure and fertilizers for the entire US-NRW,
619 even though they probably varied among fields in real practice due to variabilities in
620 application method, timing, weather, soil, and other factors. Denitrification in manured
621 soils in the Pacific Northwest can range between 5 to 30% (Paul & Zebarth, 1997;
622 USDA-NRCS, 1998), and a 17% of annual denitrification loss was measured in BC dairy
623 farms (Paul & Zebarth, 1997). Based on these assumptions, annual agricultural
624 denitrification was estimated ranging between 220 and 1400 tonnes, with our current
625 result being on the lower end. Similarly, volatilization loss in western Washington can
626 range from 10 to 50% during storage and housing and from 5 to 30% after application
627 (USDA-NRCS, 1998), representing a potential error ranging from -68 to 44% in our
628 volatilization estimation.
- 629 4) Non-continuous water sampling and potential errors during sampling and flux simulation
630 (LOADEST) could lead to deviation from the actual riverine N loads.
- 631 5) There were uncertainties associated with CMAQ and EPIC simulations. For example,
632 meteorology in the region is challenging to model; CMAQ could underestimate
633 deposition from fog in complex terrain such as the forested upland; fertilization rates for
634 many local crops could be underestimated or overestimated in EPIC; also, EPIC did not
635 account for manure that was generated and applied locally—ammonia emission from
636 animal manure was simulated separately in CMAQ.
- 637 6) Lastly, as a bi-national study, resolving issues caused by differences in data collection
638 and resolution between the two countries and the limit of our understanding of the
639 transboundary ecosystem could contribute to uncertainties in our budget. Downscaling N

640 budget results from the Canadian Lower Fraser Valley model could have induced certain
641 systematic bias and errors because of applying different boundaries.

642 Despite these limitations, we consider this budget to be a current best estimate of N
643 inputs, exports and internal cycling using local data and knowledge—this type of budget is still
644 rare for watersheds in the Pacific Northwest area (Swaney et al., 2018). The NRW N budget can
645 provide a potential roadmap for prioritization of pathways to reduce N release to the
646 environment.

647 **6 Summary**

648 Our nitrogen budget of the transboundary watershed helped to identify several key issues
649 related to better N management. Nearly 81% of the N inputs to the basin were used to support
650 agricultural production, most of which was animal feed import. Watershed N retention was
651 about 20% of the total input. The largest export from the NRW was in the form of ammonia
652 from the agriculture sector (32%), which could have air quality implications for local residents
653 and surrounding areas. Riverine export of nitrogen in to Bellingham Bay was a substantial
654 portion of the export (28%). While the climate and physiography are similar between the US
655 and Canada in the NRW, the different sides of the border provide contrasts in N management
656 and use efficiency: Crop NUE was higher on the US side of the watershed, but both the farm-
657 gate and commercial whole-farm NUEs were higher in Canada-NRW. These differences were
658 driven by the types of animals raised, manure management regulations and reporting, and farm
659 economics. As might be expected, different policy frameworks had a large impact on key
660 components of nutrient management in different portions of the watershed. We had several N
661 fluxes that were difficult to quantify with the available information. Improved information will
662 help close our knowledge gap in the future. Similarly, better quantification of N fluxes from the
663 US to Canada (in airflow) and from Canada to the US (in surface and groundwater flow), will
664 help provide better identifications of N imbalances, and thereby enhance strategic policy-making
665 to address those challenges.

666 **Acknowledgments, Samples, and Data**

667 We are grateful to Barbara Carey (Washington State Dept. of Ecology) for extensive knowledge
668 and research on groundwater conditions in the study area. We thank Martin Suchy (Environment
669 and Climate Change Canada), and Eugene Freeman and Eric Daiber (Washington State Dept. of
670 Ecology) for providing guidance on transboundary groundwater conditions. Joe Vaughn, Brian
671 Lamb, and their colleagues at Washington State University provided regional information for
672 CMAQ. We thank David Poon (BC Ministry of Agriculture) and Cecelia Wong (Environment
673 and Climate Change Canada) for providing information support. Michael Isensee (WSDA)
674 provided key dairy information. We also appreciate Julie Klacan (Washington State Dept. of
675 Fish and Wildlife), Sandra O'Neil (Washington State Dept. of Fish and Wildlife), and Eric
676 Beamer (Skagit River System Cooperative) for their support with the salmon information. We
677 also thank Hanna Winter (Lummi Natural Resources Department) for providing support on
678 riverine flux simulation. Many thanks to the Nooksack Tribe, Lummi Nation, and BC Ministry
679 of Agriculture for providing support on this project. We appreciate the many partners and
680 stakeholders who have participated in the overall project. Repository of datasets generated in
681 this study to calculate nitrogen fluxes is underway and will be publicly available at EPA
682 repository via the Environmental Dataset Gateway (EDG) prior to publication. They are now

683 available in Table S3 for review purposes. The views expressed in this article are those of the
684 author(s) and do not necessarily represent the views or policies of the government agencies.

685

686 **References**

- 687 Alexander, R. B., Böhlke, J. K., Boyer, E. W., David, M. B., Harvey, J. W., Mulholland, P. J., ...
688 & Wollheim, W. M. (2009). Dynamic modeling of nitrogen losses in river networks
689 unravels the coupled effects of hydrological and biogeochemical processes.
690 *Biogeochemistry*, 93(1-2), 91-116. <https://doi.org/10.1007/s10533-008-9274-8>
- 691 Appel, W., Napelenok, S., Hogrefe, C., Pouliot, G., Foley, K., Roselle, S., ... & Mathur, R.
692 (2017). Overview and Evaluation of the Community Multiscale Air Quality (CMAQ)
693 Modeling System Version 5.2. Chapter 11, *Air Pollution Modeling and its Application*
694 XXV. Springer International Publishing AG, Cham (ZG), Switzerland, 69-73, (2017),
695 https://doi.org/10.1007/978-3-319-57645-9_11
- 696 AVMA (American Veterinary Medical Association) (2012). *Pet Ownership & Demographics*
697 *Sourcebook*. 2012. Schaumburg, IL. Retrieved from:
698 <https://www.avma.org/KB/Resources/Statistics/Pages/default.aspx>
- 699 Baker, L. A., Hope, D., Xu, Y., Edmonds, J., & Lauver, L. (2001). Nitrogen balance for the
700 Central Arizona–Phoenix (CAP) ecosystem. *Ecosystems*, 4(6), 582-602.
701 <https://doi.org/10.1007/s10021-001-0031-2>
- 702 Basso, B., & Ritchie, J. T. (2005). Impact of compost, manure and inorganic fertilizer on nitrate
703 leaching and yield for a 6-year maize–alfalfa rotation in Michigan. *Agriculture,*
704 *ecosystems & environment*, 108(4), 329-341. <https://doi.org/10.1016/j.agee.2005.01.011>
- 705 Barthelmie, R. J., & Pryor, S. C. (1998). Implications of ammonia emissions for fine aerosol
706 formation and visibility impairment: A case study from the Lower Fraser Valley, British
707 Columbia. *Atmospheric Environment*, 32(3), 345-352. [https://doi.org/10.1016/S1352-](https://doi.org/10.1016/S1352-2310(97)83466-8)
708 [2310\(97\)83466-8](https://doi.org/10.1016/S1352-2310(97)83466-8)
- 709 Baron, J. S., Driscoll, C. T., Stoddard, J. L., & Richer, E. E. (2011). Empirical critical loads of
710 atmospheric nitrogen deposition for nutrient enrichment and acidification of sensitive US
711 lakes. *BioScience*, 61(8), 602-613. <https://doi.org/10.1525/bio.2011.61.8.6>
- 712 Binkley, D. (1994). Nitrogen fixation by red alder: biology, rates, and controls. *The biology and*
713 *management of red alder*, 57-72.
- 714 Bittman, S., Sheppard, S., Amiro, B., & Ominski, K. (2016). Nitrogen budget estimated for beef
715 operations across Canada. Paper presented at conference on Efficient use of different
716 sources of nitrogen in agriculture—from theory to practice Skara, Sweden 27 June–29
717 June 2016, 107.
- 718 Bittman, S., Sheppard, S. C., Poon, D., & Hunt, D. E. (2019). How efficient is modern peri-urban
719 nitrogen cycling: A case study. *Journal of environmental management*, 244, 462-471.
720 <https://doi.org/10.1016/j.jenvman.2019.05.054>

- 721 Boyer, E. W., Goodale, C. L., Jaworski, N. A., & Howarth, R. W. (2002). Anthropogenic
722 nitrogen sources and relationships to riverine nitrogen export in the northeastern USA.
723 *Biogeochemistry*, 57(1), 137-169. <https://doi.org/10.1023/A:1015709302073>
- 724 Bronk, D. A., See, J. H., Bradley, P., & Killberg, L. (2007). DON as a source of bioavailable
725 nitrogen for phytoplankton. *Biogeosciences*, 4(3), 283-296. [https://doi.org/10.5194/bg-4-](https://doi.org/10.5194/bg-4-283-2007)
726 283-2007
- 727 Buckley, C., Wall, D. P., Moran, B., O'Neill, S., & Murphy, P. N. (2016). Farm gate level
728 nitrogen balance and use efficiency changes post implementation of the EU Nitrates
729 Directive. *Nutrient cycling in agroecosystems*, 104(1), 1-13.
730 <https://doi.org/10.1007/s10705-015-9753-y>
- 731 Carey, B. M. (2002). Effects of Land Application of Manure on Groundwater at Two Dairies
732 Over the Sumas-Blaine Surficial Aquifer: Implications for Agronomic Rate Estimates.
733 Washington State Department of Ecology. No. 02-03-007.
- 734 Carey, B. (2017). Sumas-Blaine Aquifer Long-Term Groundwater Quality Monitoring, 2009-
735 2016. Washington State Department of Ecology, Olympia, Washington. No. 17-03-013.
- 736 Carey, B. M., & Cummings, R. (2013). Sumas-Blaine Aquifer nitrate contamination summary.
737 Washington State Department of Ecology. No. 12-03-026.
- 738 Carey, B.M., Harrison, J. H. (2014). Nitrogen dynamics at a manured grass field overlying the
739 Sumas–Blaine Aquifer in Whatcom County. Washington State Department of Ecology,
740 Olympia, Washington. Publication Number 14-03-001.
741 <https://fortress.wa.gov/ecy/publications/SummaryPages/1403001.html>
- 742 Carey, B. M., Pitz, C. F., & Harrison, J. H. (2017). Field nitrogen budgets and post-harvest soil
743 nitrate as indicators of N leaching to groundwater in a Pacific Northwest dairy grass field.
744 *Nutrient cycling in agroecosystems*, 107(1), 107-123. [https://doi.org/10.1007/s10705-](https://doi.org/10.1007/s10705-016-9819-5)
745 016-9819-5
- 746 Chen, D., Shen, H., Hu, M., Wang, J., Zhang, Y., & Dahlgren, R. A. (2018). Legacy nutrient
747 dynamics at the watershed scale: principles, modeling, and implications. In *Advances in*
748 *Agronomy* (Vol. 149, pp. 237-313). Academic Press.
749 <https://doi.org/10.1016/bs.agron.2018.01.005>
- 750 Clark, C. M., Phelan, J., Doraiswamy, P., Buckley, J., Cajka, J. C., Dennis, R. L., ... & Spero, T.
751 L. (2018). Atmospheric deposition and exceedances of critical loads from 1800– 2025 for
752 the conterminous United States. *Ecological applications*, 28(4), 978-1002.
753 <https://doi.org/10.1002/eap.1703>
- 754 Cole, D.W., H. Van Miegroet, and P.S. Homann. (1992). In D.W. Johnson and S.E. Lindberg
755 (ed.) *Atmospheric Deposition and Forest Nutrient Cycling: A Synthesis of the Integrated*
756 *Forest Study*. Springer-Verlag, New York.
- 757 Compton, J. E., Andersen, C. P., Phillips, D. L., Brooks, J. R., Johnson, M. G., Church, M. R., ...
758 & Shaff, C. D. (2006). Ecological and water quality consequences of nutrient addition for
759 salmon restoration in the Pacific Northwest. *Frontiers in Ecology and the Environment*,
760 4(1), 18-26. [https://doi.org/10.1890/1540-9295\(2006\)004\[0018:EAWQCO\]2.0.CO;2](https://doi.org/10.1890/1540-9295(2006)004[0018:EAWQCO]2.0.CO;2)

- 761 Compton, J. E., Church, M. R., Larned, S. T., & Hogsett, W. E. (2003). Nitrogen export from
762 forested watersheds in the Oregon Coast Range: the role of N₂-fixing red alder.
763 *Ecosystems*, 6(8), 773-785. <https://doi.org/10.1007/s10021-002-0207-4>
- 764 Compton, J. E., Goodwin, K. E., Sobota, D. J., & Lin, J. (2019). Seasonal Disconnect Between
765 Streamflow and Retention Shapes Riverine Nitrogen Export in the Willamette River
766 Basin, Oregon. *Ecosystems*, 1-17. <https://doi.org/10.1007/s10021-019-00383-9>
- 767 Cox, S. E., Simonds, F., Doremus, L., Huffman, R. L., & Defawe, R. M. (2005). Ground
768 water/surface water interactions and quality of discharging ground water in streams of the
769 lower Nooksack River Basin, Whatcom County, Washington (Scientific Investigations
770 Report 2005-5255). U. S. Geological Survey.
- 771 Cox, S. E., Spanjer, A. R., Huffman, R. L., Black, R. W., Barbash, J. E., & Embertson, N. M.
772 (2018). Concentrations of Nutrients at the Water Table beneath Forage Fields Receiving
773 Seasonal Applications of Manure, Whatcom County, Washington, Autumn 2011–Spring
774 2015 (Scientific Investigations Report 2018-5124). US Geological Survey.
- 775 Davidson, E. A., David, M. B., Galloway, J. N., Goodale, C. L., Haeuber, R., Harrison, J. A., ...
776 & Peel, J. L. (2011). Excess nitrogen in the US environment: trends, risks, and solutions.
777 *Issues in Ecology*, (15).
- 778 De Girolamo, A. M., Balestrini, R., D'Ambrosio, E., Pappagallo, G., Soana, E., & Porto, A. L.
779 (2017). Anthropogenic input of nitrogen and riverine export from a Mediterranean
780 catchment. The Celone, a temporary river case study. *Agricultural water management*,
781 187, 190-199. <https://doi.org/10.1016/j.agwat.2017.03.025>
- 782 Demurtas, C. E., Seddaiu, G., Ledda, L., Cappai, C., Doro, L., Carletti, A., & Roggero, P. P.
783 (2016). Replacing organic with mineral N fertilization does not reduce nitrate leaching in
784 double crop forage systems under Mediterranean conditions. *Agriculture, Ecosystems &
785 Environment*, 219, 83-92. <https://doi.org/10.1016/j.agee.2015.12.010>
- 786 Di, H. J., & Cameron, K. C. (2002). The use of a nitrification inhibitor, dicyandiamide (DCD), to
787 decrease nitrate leaching and nitrous oxide emissions in a simulated grazed and irrigated
788 grassland. *Soil use and management*, 18(4), 395-403. [https://doi.org/10.1111/j.1475-
789 2743.2002.tb00258.x](https://doi.org/10.1111/j.1475-2743.2002.tb00258.x)
- 790 Dickerson-Lange, S. E., & Mitchell, R. (2014). Modeling the effects of climate change
791 projections on streamflow in the Nooksack River basin, Northwest Washington.
792 *Hydrological Processes*, 28(20), 5236-5250. <https://doi.org/10.1002/hyp.10012>
- 793 Embertson, N., & District, W. C. (2016). Protecting Puget Sound Watersheds from Agricultural
794 Pollution Using a Progressive Manure Application Risk Management (ARM) System.
- 795 Erisman, J. W., Grennfelt, P., & Sutton, M. (2003). The European perspective on nitrogen
796 emission and deposition. *Environment International*, 29(2-3), 311-325.
797 [https://doi.org/10.1016/S0160-4120\(02\)00162-9](https://doi.org/10.1016/S0160-4120(02)00162-9)
- 798 ESRI 2011. ArcGIS Desktop: Release 10. Redlands, CA: Environmental Systems Research
799 Institute.

- 800 Galloway, J. N., Aber, J. D., Erisman, J. W., Seitzinger, S. P., Howarth, R. W., Cowling, E. B.,
801 & Cosby, B. J. (2003). The nitrogen cascade. *Bioscience*, 53(4), 341-356.
802 [https://doi.org/10.1641/0006-3568\(2003\)053\[0341:TNC\]2.0.CO;2](https://doi.org/10.1641/0006-3568(2003)053[0341:TNC]2.0.CO;2)
- 803 Galloway, J. N., Dentener, F. J., Capone, D. G., Boyer, E. W., Howarth, R. W., Seitzinger, S. P.,
804 Asner, G.P., Cleveland, C.C., Green, P.A., Holland, E.A. & Karl, D. M. (2004). Nitrogen
805 cycles: past, present, and future. *Biogeochemistry*, 70(2), 153-226.
806 <https://doi.org/10.1007/s10533-004-0370-0>
- 807 Geiser, L. H., Jovan, S. E., Glavich, D. A., & Porter, M. K. (2010). Lichen-based critical loads
808 for atmospheric nitrogen deposition in Western Oregon and Washington Forests, USA.
809 *Environmental Pollution*, 158(7), 2412-2421.
810 <https://doi.org/10.1016/j.envpol.2010.04.001>
- 811 Goyette, J. O., Bennett, E. M., Howarth, R. W., & Maranger, R. (2016). Changes in
812 anthropogenic nitrogen and phosphorus inputs to the St. Lawrence sub-basin over 110
813 years and impacts on riverine export. *Global Biogeochemical Cycles*, 30(7), 1000-1014.
814 <https://doi.org/10.1002/2016GB005384>
- 815 Greaver, T. L., Sullivan, T. J., Herrick, J. D., Barber, M. C., Baron, J. S., Cosby, B. J., ... &
816 Herlihy, A. T. (2012). Ecological effects of nitrogen and sulfur air pollution in the US:
817 what do we know? *Frontiers in Ecology and the Environment*, 10(7), 365-372.
818 <https://doi.org/10.1890/110049>
- 819 Gresh, T., Lichatowich, J., & Schoonmaker, P. (2000). An estimation of historic and current
820 levels of salmon production in the Northeast Pacific ecosystem: evidence of a nutrient
821 deficit in the freshwater systems of the Pacific Northwest. *Fisheries*, 25(1), 15-21.
822 [https://doi.org/10.1577/1548-8446\(2000\)025<0015:AEOHAC>2.0.CO;2](https://doi.org/10.1577/1548-8446(2000)025<0015:AEOHAC>2.0.CO;2)
- 823 Hall, K. D., Guo, J., Dore, M., & Chow, C. C. (2009). The progressive increase of food waste in
824 America and its environmental impact. *PloS one*, 4(11), e7940.
825 <https://doi.org/10.1371/journal.pone.0007940>
- 826 Halloran, A., Clement, J., Kornum, N., Bucatariu, C., & Magid, J. (2014). Addressing food waste
827 reduction in Denmark. *Food Policy*, 49, 294-301.
828 <https://doi.org/10.1016/j.foodpol.2014.09.005>
- 829 Harrison, J. H., Nennich, T. D., & White, R. (2007). Nutrient management and dairy cattle
830 production. *CAB Reviews: Perspectives in Agriculture, Veterinary Science, Nutrition and*
831 *Natural Resources*, 2(020).
- 832 Harrison, J., White, R., Ishler, V., Erickson, G., Sutton, A., Applegate, T., ... & Meyer, D.
833 (2012). CASE STUDY: Implementation of feed management as part of whole-farm
834 nutrient management. *The Professional Animal Scientist*, 28(3), 364-369.
835 [https://doi.org/10.15232/S1080-7446\(15\)30369-7](https://doi.org/10.15232/S1080-7446(15)30369-7)
- 836 Homer, C., Dewitz, J., Yang, L., Jin, S., Danielson, P., Xian, G., ... & Megown, K. (2015).
837 Completion of the 2011 National Land Cover Database for the conterminous United
838 States—representing a decade of land cover change information. *Photogrammetric*
839 *Engineering & Remote Sensing*, 81(5), 345-354.

- 840 Hong, B., Swaney, D. P., & Howarth, R. W. (2011). A toolbox for calculating net anthropogenic
841 nitrogen inputs (NANI). *Environmental Modelling & Software*, 26(5), 623-633.
842 <https://doi.org/10.1016/j.envsoft.2010.11.012>
- 843 Hong, B., Swaney, D. P., & Howarth, R. W. (2013). Estimating net anthropogenic nitrogen
844 inputs to US watersheds: comparison of methodologies. *Environmental science &*
845 *technology*, 47(10), 5199-5207. <https://doi.org/10.1021/es303437c>
- 846 Janetski, D. J., Chaloner, D. T., Tiegs, S. D., & Lamberti, G. A. (2009). Pacific salmon effects on
847 stream ecosystems: a quantitative synthesis. *Oecologia*, 159(3), 583-595.
848 <https://doi.org/10.1007/s00442-008-1249-x>
- 849 Khangaonkar, T., Nugraha, A., Xu, W., & Balaguru, K. (2019). Salish Sea Response to Global
850 Climate Change, Sea Level Rise, and Future Nutrient Loads. *Journal of Geophysical*
851 *Research: Oceans*. <https://doi.org/10.1029/2018JC014670>
- 852 Kotchenruther, R., & Taylor, B. The Georgia Basin-Puget Sound Airshed Characterization
853 Report 2014: chapter 10.
- 854 Kottek, M., Grieser, J., Beck, C., Rudolf, B., & Rubel, F. (2006). World map of the Köppen-
855 Geiger climate classification updated. *Meteorol. Z.*, 15(3), 259-263.
856 <https://doi.org/10.1127/0941-2948/2006/0130>
- 857 Kroeger, K. D., Cole, M. L., & Valiela, I. (2006). Groundwater-transported dissolved organic
858 nitrogen exports from coastal watersheds. *Limnology and Oceanography*, 51(5), 2248-
859 2261. <https://doi.org/10.4319/lo.2006.51.5.2248>
- 860 Lin, J., Compton, J. E., Leibowitz, S. G., Mueller-Warrant, G., Matthews, W., Schoenholtz, S.
861 H., ... & Coulombe, R. A. (2019). Seasonality of nitrogen balances in a Mediterranean
862 climate watershed, Oregon, US. *Biogeochemistry*, 142(2), 247-264.
863 <https://doi.org/10.1007/s10533-018-0532-0>
- 864 Liptzin D, Dahlgren R. (2016). A California Nitrogen Mass Balance for 2005. In Tomich TP,
865 Brodt SB, Dahlgren RA, Scow KM (Eds.), *The California Nitrogen Assessment* (pp. 79-
866 112). University of California Press.
- 867 Liu, Z., Liu, Y., Murphy, J. P., & Maghirang, R. (2017). Ammonia and methane emission factors
868 from cattle operations expressed as losses of dietary nutrients or energy. *Agriculture*,
869 7(3), 16. <https://doi.org/10.3390/agriculture7030016>
- 870 Lehmann, J., & Rondon, M. (2006). Bio-char soil management on highly weathered soils in the
871 humid tropics. *Biological approaches to sustainable soil systems*, 113(517), e530.
- 872 Loo, S. E., Zebarth, B. J., Ryan, M. C., Forge, T. A., & Cey, E. E. (2019). Quantifying Nitrate
873 Leaching under Commercial Red Raspberry Using Passive Capillary Wick Samplers.
874 *Vadose Zone Journal*, 18(1). <https://doi.org/10.2136/vzj2018.08.0152>
- 875 McBroom, M. W., Beasley, R. S., Chang, M., & Ice, G. G. (2008). Water quality effects of
876 clearcut harvesting and forest fertilization with best management practices. *Journal of*
877 *Environmental Quality*, 37(1), 114-124. <https://doi.org/10.2134/jeq2006.0552>
- 878 McCrackin, M. L., Muller-Karulis, B., Gustafsson, B. G., Howarth, R. W., Humborg, C.,
879 Svanbäck, A., & Swaney, D. P. (2018). A century of legacy phosphorus dynamics in a

- 880 large drainage basin. *Global Biogeochemical Cycles*, 32(7), 1107-1122.
881 <https://doi.org/10.1029/2018GB005914>
- 882 McMurray, J. A., Roberts, D. W., Fenn, M. E., Geiser, L. H., & Jovan, S. (2013). Using
883 epiphytic lichens to monitor nitrogen deposition near natural gas drilling operations in the
884 Wind River Range, WY, USA. *Water, Air, & Soil Pollution*, 224(3), 1487.
885 <https://doi.org/10.1007/s11270-013-1487-3>
- 886 Mohamedali, T., Roberts, M., Sackmann, B., & Kolosseus, A. (2011). Puget sound dissolved
887 oxygen model nutrient load summary for 1998-2008. Washington State Department of
888 Ecology. No. 11-03-057.
- 889 Moore, J. W., Hayes, S. A., Duffy, W., Gallagher, S., Michel, C. J., & Wright, D. (2011).
890 Nutrient fluxes and the recent collapse of coastal California salmon populations.
891 *Canadian Journal of Fisheries and Aquatic Sciences*, 68(7), 1161-1170.
892 <https://doi.org/10.1139/f2011-054>
- 893 National Research Council. (1994). *Nutrient Requirements of Poultry Ninth Revised Edition*
894 National Academy Press. Washington DC. <https://doi.org/10.17226/2114>
- 895 Nennich, T. D., Harrison, J. H., Vanwieringen, L. M., Meyer, D., Heinrichs, A. J., Weiss, W. P.,
896 ... & Block, E. (2005). Prediction of manure and nutrient excretion from dairy cattle.
897 *Journal of Dairy Science*, 88(10), 3721-3733. [https://doi.org/10.3168/jds.S0022-0302\(05\)73058-7](https://doi.org/10.3168/jds.S0022-0302(05)73058-7)
- 898
- 899 Oenema, O., Kros, H., & de Vries, W. (2003). Approaches and uncertainties in nutrient budgets:
900 implications for nutrient management and environmental policies. *European Journal of*
901 *Agronomy*, 20(1-2), 3-16. [https://doi.org/10.1016/S1161-0301\(03\)00067-4](https://doi.org/10.1016/S1161-0301(03)00067-4)
- 902 Ohmann, J. L., Gregory, M. J., Henderson, E. B., & Roberts, H. M. (2011). Mapping gradients of
903 community composition with nearest-neighbour imputation: extending plot data for
904 landscape analysis. *Journal of Vegetation Science*, 22(4), 660-676.
905 <https://doi.org/10.1111/j.1654-1103.2010.01244.x>
- 906 Olson, M. H., Hage, M. M., Binkley, M. D., & Binder, J. R. (2005). Impact of migratory snow
907 geese on nitrogen and phosphorus dynamics in a freshwater reservoir. *Freshwater*
908 *Biology*, 50(5), 882-890. <https://doi.org/10.1111/j.1365-2427.2005.01367.x>
- 909 Ovens, R., Weaver, D., Keipert, N., Neville, S., Summers, R., & Clarke, M. (2008). Farm gate
910 nutrient balances in south west Western Australia—An overview. Paper presented at 12th
911 International Conference on Integrated Diffuse Pollution Management, Khon Kaen,
912 Thailand
- 913 Paulot, F., & Jacob, D. J. (2014). Hidden cost of US agricultural exports: particulate matter from
914 ammonia emissions. *Environmental science & technology*, 48(2), 903-908.
915 <https://doi.org/10.1021/es4034793>
- 916 Paul, J. W., & Zebarth, B. J. (1997). Denitrification and nitrate leaching during the fall and
917 winter following dairy cattle slurry application. *Canadian journal of soil science*, 77(2),
918 231-240. <https://doi.org/10.4141/S96-052>

- 919 Pelto, M. (2015). Climate driven retreat of mount baker glaciers and changing water resources.
920 Switzerland: Springer International Publishing. [https://doi.org/10.1007/978-3-319-22605-](https://doi.org/10.1007/978-3-319-22605-7)
921 7
- 922 Pennino, M. J., Compton, J. E., & Leibowitz, S. G. (2017). Trends in drinking water nitrate
923 violations across the United States. *Environmental science & technology*, 51(22), 13450-
924 13460. <https://doi.org/10.1021/acs.est.7b04269>
- 925 Powell, J. M., & Rotz, C. A. (2015). Measures of nitrogen use efficiency and nitrogen loss from
926 dairy production systems. *Journal of Environmental Quality*, 44(2), 336-344.
927 <https://doi.org/10.2134/jeq2014.07.0299>
- 928 Runkel, R. L., Crawford, C. G., & Cohn, T. A. (2004). Load Estimator (LOADEST): A
929 FORTRAN program for estimating constituent loads in streams and rivers (No. 4-A5).
930 <https://doi.org/10.3133/tm4A5>
- 931 Sajeev, M., Purath, E., Winiwarer, W., & Amon, B. (2018). Greenhouse gas and ammonia
932 emissions from different stages of liquid manure management chains: abatement options
933 and emission interactions. *Journal of environmental quality*, 47(1), 30-41.
934 <https://doi.org/10.2134/jeq2017.05.0199>
- 935 Sanchis, E., Calvet, S., del Prado, A., & Estellés, F. (2019). A meta-analysis of environmental
936 factor effects on ammonia emissions from dairy cattle houses. *Biosystems engineering*,
937 178, 176-183. <https://doi.org/10.1016/j.biosystemseng.2018.11.017>
- 938 Saunders, O. E., Fortuna, A. M., Harrison, J. H., Cogger, C. G., Whitefield, E., & Green, T.
939 (2012). Gaseous nitrogen and bacterial responses to raw and digested dairy manure
940 applications in incubated soil. *Environmental science & technology*, 46(21), 11684-
941 11692. <https://doi.org/10.1021/es301754s>
- 942 Sebilo, M., Mayer, B., Nicolardot, B., Pinay, G., & Mariotti, A. (2013). Long-term fate of nitrate
943 fertilizer in agricultural soils. *Proceedings of the National Academy of Sciences*, 110(45),
944 18185-18189. <https://doi.org/10.1073/pnas.1305372110>
- 945 Sheibley, R. W., Enache, M., Swarzenski, P. W., Moran, P. W., & Foreman, J. R. (2014).
946 Nitrogen deposition effects on diatom communities in lakes from three national parks in
947 Washington State. *Water, Air, & Soil Pollution*, 225(2), 1857.
948 <https://doi.org/10.1007/s11270-013-1857-x>
- 949 Sheppard, S. C., Bittman, S., Swift, M. L., & Tait, J. (2010). Farm practices survey and
950 modelling to estimate monthly NH₃ emissions from swine production in 12 Ecoregions
951 of Canada. *Canadian journal of animal science*, 90(2), 145-158.
952 <https://doi.org/10.4141/CJAS09050>
- 953 Sheppard, S., Bittman, S., Swift, M., & Tait, J. (2011). Modelling monthly NH₃ emissions from
954 dairy in 12 Ecoregions of Canada. *Canadian Journal of Animal Science*, 91(4), 649-661.
955 <https://doi.org/10.4141/cjas2010-005>
- 956 Shibata, H., Galloway, J. N., Leach, A. M., Cattaneo, L. R., Noll, L. C., Erisman, J. W., ... &
957 Dalgaard, T. (2017). Nitrogen footprints: Regional realities and options to reduce
958 nitrogen loss to the environment. *Ambio*, 46(2), 129-142. [https://doi.org/10.1007/s13280-](https://doi.org/10.1007/s13280-016-0815-4)
959 [016-0815-4](https://doi.org/10.1007/s13280-016-0815-4)

- 960 Skamarock, W. C., & Klemp, J. B. (2008). A time-split nonhydrostatic atmospheric model for
961 weather research and forecasting applications. *Journal of computational physics*, 227(7),
962 3465-3485. <https://doi.org/10.1016/j.jcp.2007.01.037>
- 963 Sprague, L. A., Langland, M. J., Yochum, S. E., Edwards, R. E., Blomquist, J. D., Phillips, S.
964 W., ... & Preston, S. D. (2000). Factors affecting nutrient trends in major rivers of the
965 Chesapeake Bay watershed (No. 2000-4218). US Dept. of the Interior, US Geological
966 Survey; Branch of Information Services [distributor]. <https://doi.org/10.3133/wri004218>
- 967 Sullivan, C.S., & Poon, D., (2016). Fraser Valley Soil Nutrient Survey 2012. BCMA. Retrieved
968 from: [http://www2.gov.bc.ca/gov/content/industry/agriculture-seafood/agricultural-land-](http://www2.gov.bc.ca/gov/content/industry/agriculture-seafood/agricultural-land-andenvironment/soil-nutrients/nutrient-management/technical-reports/soil-nutrientstudies)
969 [andenvironment/soil-nutrients/nutrient-management/technical-reports/soil-nutrientstudies](http://www2.gov.bc.ca/gov/content/industry/agriculture-seafood/agricultural-land-andenvironment/soil-nutrients/nutrient-management/technical-reports/soil-nutrientstudies)
- 970 Swaney, D. P., Howarth, R. W., & Hong, B. (2018). Nitrogen use efficiency and crop
971 production: patterns of regional variation in the United States, 1987–2012. *Science of*
972 *The Total Environment*, 635, 498-511. <https://doi.org/10.1016/j.scitotenv.2018.04.027>
- 973 Team, T. (2015). Recommended phosphorus loading targets for Lake Erie. In Annex 4
974 Objectives and Targets Task Team Final Report to the Nutrients Annex Subcommittee.
- 975 Tarkalson, D. D., Payero, J. O., Ensley, S. M., & Shapiro, C. A. (2006). Nitrate accumulation
976 and movement under deficit irrigation in soil receiving cattle manure and commercial
977 fertilizer. *Agricultural water management*, 85(1-2), 201-210.
978 <https://doi.org/10.1016/j.agwat.2006.04.005>
- 979 Townsend, A. R., Howarth, R. W., Bazzaz, F. A., Booth, M. S., Cleveland, C. C., Collinge, S.
980 K., ... & Mallin, M. A. (2003). Human health effects of a changing global nitrogen cycle.
981 *Frontiers in Ecology and the Environment*, 1(5), 240-246. [https://doi.org/10.1890/1540-](https://doi.org/10.1890/1540-9295(2003)001[0240:HHEOAC]2.0.CO;2)
982 [9295\(2003\)001\[0240:HHEOAC\]2.0.CO;2](https://doi.org/10.1890/1540-9295(2003)001[0240:HHEOAC]2.0.CO;2)
- 983 USDA (US Department of Agriculture) (1998). *Agricultural Waste Management Field*
984 *Handbook*, 210-AWMFH Supplement, WA-2. Natural Resources Conservation Service,
985 Washington DC. Retrieved from:
986 https://www.nrcs.usda.gov/Internet/FSE_DOCUMENTS/nrcs144p2_035264.pdf
- 987 USDA (US Department of Agriculture). (2017). *Quick Stats 2.0*. U.S. Department of
988 Agriculture, National Agricultural Statistics Service, Washington DC. Retrieved from:
989 <https://quickstats.nass.usda.gov/>
- 990 USDA (US Department of Agriculture). (2018). *USDA National Nutrient Database for Standard*
991 *Reference, Legacy*. Agricultural Research Service, Nutrient Data Laboratory. Retrieved
992 from: [https://www.ars.usda.gov/northeast-area/beltsville-md-bhnrc/beltsville-human-](https://www.ars.usda.gov/northeast-area/beltsville-md-bhnrc/beltsville-human-nutrition-research-center/nutrient-data-laboratory/docs/sr28-page-reports/)
993 [nutrition-research-center/nutrient-data-laboratory/docs/sr28-page-reports/](https://www.ars.usda.gov/northeast-area/beltsville-md-bhnrc/beltsville-human-nutrition-research-center/nutrient-data-laboratory/docs/sr28-page-reports/)
- 994 USDA (US Department of Agriculture). (2019). *The PLANTS Database*. Natural Resources
995 Conservation Service, National Plant Data Team, Greensboro, NC. Retrieved from:
996 <http://plants.usda.gov>
- 997 USDA (US Department of Agriculture, Agricultural Research Service, Beltsville Human
998 Nutrition Research Center, Food Surveys Research Group) (Beltsville, MD) and HHS
999 (US Department of Health and Human Services, Centers for Disease Control and
1000 Prevention, National Center for Health Statistics) (Hyattsville, MD). (2016). *What We*
1001 *Eat in America, NHANES 2013-2014 Data: Dietary Interview - Total Nutrients Intakes --*

- 1002 First Day (DR1TOT_C). Retrieved from:
1003 https://www.ars.usda.gov/ARUserFiles/80400530/pdf/1314/Table_1_NIN_GEN_13.pdf
- 1004 USEPA (US Environmental Protection Agency). (2002). Onsite wastewater treatment systems
1005 manual. Retrieved from: [https://www.epa.gov/sites/production/files/2015-](https://www.epa.gov/sites/production/files/2015-06/documents/2004_07_07_septics_septic_2002_osdm_all.pdf)
1006 [06/documents/2004_07_07_septics_septic_2002_osdm_all.pdf](https://www.epa.gov/sites/production/files/2015-06/documents/2004_07_07_septics_septic_2002_osdm_all.pdf)
- 1007 USGS (US Geological Survey). (2016). National Water Information System data available on the
1008 World Wide Web (USGS Water Data for the Nation). Retrieved from:
1009 <https://waterdata.usgs.gov/nwis/>
- 1010 USGS (US Geological Survey). (2013). Revision to LOADEST. Online document for Load
1011 Estimator (LOADEST): A Program for Estimating Constituent Loads in Streams and
1012 Rivers. Retrieved from: https://water.usgs.gov/software/loadest/doc/loadest_update.pdf
- 1013 van der Meer, H. G. (2001). Reduction of nitrogen losses in dairy production systems: the Dutch
1014 experience. In K. Kanwar & R. Baggett (Eds), *Nutrient Management Challenges in*
1015 *Livestock and Poultry Operations: The Dutch Experience*, (pp. 82-97). Madison,
1016 Wisconsin: Babcock Institute.
- 1017 van Grinsven, H. J., Erisman, J. W., de Vries, W., & Westhoek, H. (2015). Potential of
1018 extensification of European agriculture for a more sustainable food system, focusing on
1019 nitrogen. *Environmental Research Letters*, 10(2), 025002. [https://doi.org/10.1088/1748-](https://doi.org/10.1088/1748-9326/10/2/025002)
1020 [9326/10/2/025002](https://doi.org/10.1088/1748-9326/10/2/025002)
- 1021 van Meter, K. J., Basu, N. B., Veenstra, J. J., & Burras, C. L. (2016). The nitrogen legacy:
1022 emerging evidence of nitrogen accumulation in anthropogenic landscapes. *Environmental*
1023 *Research Letters*, 11(3), 035014. <https://doi.org/10.1088/1748-9326/11/3/035014>
- 1024 van Vliet, L. J., Zebarth, B. J., & Derksen, G. (2002). Effect of fall-applied manure practices on
1025 runoff, sediment, and nutrient surface transport from silage corn in south coastal British
1026 Columbia. *Canadian journal of soil science*, 82(4), 445-456. [https://doi.org/10.4141/S01-](https://doi.org/10.4141/S01-041)
1027 [041](https://doi.org/10.4141/S01-041)
- 1028 Wassenaar, L. I., Hendry, M. J., & Harrington, N. (2006). Decadal geochemical and isotopic
1029 trends for nitrate in a transboundary aquifer and implications for agricultural beneficial
1030 management practices. *Environmental science & technology*, 40(15), 4626-4632.
1031 <https://doi.org/10.1021/es060724w>
- 1032 Weathers, K. C., Cadenasso, M. L., & Pickett, S. T. (2001). Forest edges as nutrient and
1033 pollutant concentrators: potential synergisms between fragmentation, forest canopies, and
1034 the atmosphere. *Conservation Biology*, 15(6), 1506-1514. [https://doi.org/10.1046/j.1523-](https://doi.org/10.1046/j.1523-1739.2001.01090.x)
1035 [1739.2001.01090.x](https://doi.org/10.1046/j.1523-1739.2001.01090.x)
- 1036 Welter, J. R., & Fisher, S. G. (2016). The influence of storm characteristics on hydrological
1037 connectivity in intermittent channel networks: implications for nitrogen transport and
1038 denitrification. *Freshwater biology*, 61(8), 1214-1227. <https://doi.org/10.1111/fwb.12734>
- 1039 Wise, D. R., & Johnson, H. M. (2011). Surface-Water Nutrient Conditions and Sources in the
1040 United States Pacific Northwest1. *JAWRA Journal of the American Water Resources*
1041 *Association*, 47(5), 1110-1135. <https://doi.org/10.1111/j.1752-1688.2011.00580.x>

- 1042 WSDA (Washington State Department of Agriculture) (2015). 2014 WSDA Agricultural Land
1043 Use. Olympia, WA. Washington State Department of Agriculture. Retrieved from:
1044 <https://agr.wa.gov/departments/land-and-water/natural-resources/agricultural-land-use>
- 1045 WSDA (Washington State Department of Agriculture) (2018). Dairy Nutrient Management
1046 Program, Inspection Report Data Summary, 2013-2015. Retrieved from:
1047 <http://arcg.is/KHCmq>
- 1048 Zebarth, B. J., Ryan, M. C., Graham, G., Forge, T. A., & Neilsen, D. (2015). Groundwater
1049 Monitoring to Support Development of BMPs for Groundwater Protection: The
1050 Abbotsford-Sumas Aquifer Case Study. *Groundwater Monitoring & Remediation*, 35(1),
1051 82-96. <https://doi.org/10.1111/gwmmr.12092>
- 1052 Zhang, X., Davidson, E. A., Mauzerall, D. L., Searchinger, T. D., Dumas, P., & Shen, Y. (2015).
1053 Managing nitrogen for sustainable development. *Nature*, 528(7580), 51.
1054 <https://doi.org/10.1038/nature15743>

1055 **Table 1.** Budget Components and Data Sources (for all US Components and Some of the Canadian Components) for Nooksack River
1056 Watershed.
1057

1058

	Component	Parameter	Data source
INPUTS	Atmospheric Deposition	Total N deposition	EPA-CMAQ (Appel et al., 2017)
	Food Import (Human)	Human population	USDA-NASS, 2017: 2015 census
		Nutritional consumption, per capita	USDA & HHS, 2016; Hall et al., 2009
	Food Import (Pet)	Watershed household	USDA-NASS, 2017 (2015 census)
		Population and body weights: Dogs and cats	Dogs - 37% of watershed households; Cats - 30% of watershed households. Assuming one pet per household; US Pet Ownership Statistics (AVMA, 2012); Baker et al., 2001
		Nutritional and energy needs	Veterinary online manual (link); Pet Basic Calorie Calculator (link)
	Feed Import	Animal populations (other than dairy cow, such as duck, goat, turkey, hogs, sheep, etc.)	USDA-NASS, 2017: 2012 data
		Dairy cow population	WSDA (2018)
		Nutritional needs of farm animals	Boyer et al., 2002; Hong et al., 2011, 2013; National Research Council, 1994; Veterinary online manual (link); Nennich et al., 2005; Bittman et al., 2019; Goyette et al., 2016
	Fertilizer Import	Crop land	WSDA, 2015
		Crop fertilization rates	Local agriculture experts (personal communication: WCD); Lin et al., 2019; Oregon (link) and Washington (link) Extension online documentations
	Biological N Fixation	Alder density	Ohman et al., 2011
		Alder N fixation rate	Binkley, 1994
	Adult Fish Return	Salmon population and size	Nooksack Stock Assessment (personal communication: WDFW Fish Program)
		Adult fish body weight	Gresh et al., 2000
Adult fish body N content		Moore et al., 2011	

1059

	Component	Parameter	Data source
OUTPUTS	Riverine Nitrate/TKN Export	Flow	USGS site 12213100 (USGS, 2016)
		Concentrations	Nitrate: WA Dept. of Ecology site 01A050; TKN: Lummi Nation site SW118; USGS site 12213100
		Natural land area	NLCD 2011 (Homer et al., 2015)
		Forest N leaching rate	Cole et al., 1992
	NH₃ Volatilization	Animal manure application rates	Bittman et al., 2019; Hong et al., 2011, 2013; Nennich et al., 2005; Sheppard et al., 2011; USDA-NASS, 2017 (2012 data); WSDA (2018)
		Synthetic fertilizer application rates	Local agriculture experts (personal communication: WCD); Lin et al., 2019; Oregon (link) and Washington (link) Extension online documentations; WSDA, 2015
		Fertilizer and manure volatilization rate/percentage	Carey & Harrison, 2014; USDA-NRCS (1998)
	Denitrification Loss	Fertilizer and manure denitrification rate/percentage	USDA-NRCS (1998)
	Animal Product (Milk)	Dairy cow population	WSDA (2018) (2014 data)
		Milk N production rate	USDA-ARS, 2018 ; Bittman et al., 2019 ; Goyette et al., 2016
	Animal Product (Other)	Animal populations (other than dairy cow)	USDA-NASS, 2017 (2012 data)
		Animal product N content	USDA-ARS, 2018 ; Bittman et al., 2019 ; Goyette et al., 2016
	Crop Product	Crop land	WSDA (2015)
		Crop N content	USDA-NRCS, 2019
	Smolt Export	Smolt population and size	Lummi Nation (personal communication: Julie Klacan and Sandra O'Neil, Washington State Dept. of Fish and Wildlife)
		Smolt body weight equation	Skagit River System Cooperative (personal communication: Eric Beamer, SRSC Research Department)
		Smolt body N content	Moore et al., 2011

	Component	Parameter	Data source
INTERNAL CYCLING	Human Waste	Sewage Treatment Plants (STPs) monitored N in effluents	Everson STP (link); Lynden STP (link); Ferndale STP (link)
		Septic population: total population - service population on sewage	USDA-NASS, 2017; Everson STP; Lynden STP; Ferndale STP
		Septic leaching rate, per capita	USEPA, 2002
	Food Waste	40% of total available food	Hall et al., 2009
	Manure Application	Animal populations (other than dairy cow)	USDA-NASS, 2017 (2012 data); WSDA (2018)
		Animal excretion rates	Bittman et al., 2019 ; Hong et al., 2011, 2013 ; Nennich et al., 2005; Sheppard et al., 2011
	Crop to Animal Feed	Feed crop production rate	Local agriculture experts (personal communication: WCD); USDA-NASS, 2017 (2012 data)
		Crop N content	USDA-NRCS (2019); local agriculture experts (personal communication: WCD)

1060

1061

1062

1063

1064

Note: Most of the budget results for Canada-NRW were extracted from an existing nutrient budget model that conducted a N assessment for the Lower Fraser Valley in BC (Bittman et al., 2019), except for the following: atmospheric deposition, food import, human waste, and food waste. For these fluxes, the same approaches and assumptions were applied to calculate these components for both the US and Canadian portions of the watershed.

1065 **Table 2.** Nitrogen use efficiencies (NUEs) in the Nooksack River Watershed

1066

	<u>Nitrogen use efficiency (NUE)</u>		
	US-NRW	Canada-NRW	Whole NRW
Crop NUE (Total Manure and Fertilizer)	54%	22%	51%
Crop NUE (Applied Manure and Fertilizer)	71%	31%	67%
Farm-Gate NUE	19%	45%	27%
Commercial Whole-Farm NUE	24%	53%	33%

1067