

Climate change, land-water transfer, and in-stream fate of nitrogen in an agricultural setting

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5. Moon-Neilson, L., T.T., Brown, C.J.Kelley, R.A. Martin, S.Waldo, 2013, Budget Approach Provides Insights into Transformations and Fate of Nitrogen Applied to an Eastern Washington Agroecosystem, NSF IGERT Poster and Video Competition.
6. Martin, R. A., and J. A. Harrison, 2012, Hydrologic variability controls the quantity and quality of dissolved organic carbon and nitrogen exported from agricultural soils, poster, WSU Vancouver Research Showcase, Vancouver WA.
7. Martin RA and JA Harrison, 2012,Flow paths control the delivery of dissolved organic carbon and nitrogen from agricultural soils to surface water, poster, Washington State University Academic Showcase, Pullman, WA.

RESEARCH PROBLEM AND OBJECTIVES

In recent decades, climate change has significantly altered Washington precipitation and streamflow (Fu et al., 2010), and climate effects on hydrologic fluxes are predicted to intensify in coming decades (Mote and Salathé, 2010). Two robust historic and projected trends in eastern Washington precipitation include increasing fall precipitation and increases in heavy precipitation events (Meehl et al., 2005, Salathe et al., 2010). In the Palouse, these changes, along with higher predicted winter temperatures (leading to more rain, less snow) are likely to intensify pulsed hydrologic events (rainstorms and associated runoff) that carry solutes and sediments into streams. The effects of such changes on water fluxes and quality are certain to be important but are currently poorly understood.

Soluble, reactive nutrients such as nitrate (NO_3^-) and dissolved organic matter (DOM) are of particular concern. NO_3^- at high concentrations is a contaminant regulated by the Clean Water Act and is a common pollutant in agricultural watersheds. While DOM can be an important source of carbon and nitrogen to stream ecosystems at low levels, both excess NO_3^- and DOM can lead to poor water quality, contributing to low dissolved oxygen conditions (Jassby and Van Nieuwenhuysse, 2005), harmful algal blooms (Glibert et al., 2010), and drinking water contamination (Chow et al., 2007; Thouin et al., 2009). NO_3^- and DOM-related water quality problems are widespread in Washington, with over 700 water bodies currently listed as impaired with respect to dissolved oxygen (WA DoE), and more than 40 sites listed as impaired by high total phosphorus or total nitrogen levels (Washington DoE 2008).

Agricultural land is an important source of NO_3^- , dissolved organic carbon (DOC) and nitrogen (DON), to surface water (Royer et al., 2006, Warner et al., 2009, Pellerin et al., 2006). Agriculturally-derived DOC and DON can be more bioavailable than DOC and DON derived from natural systems (Wiegner and Seitzinger, 2004, Warner et al., 2009), and bioavailable DOC may stimulate in-stream nitrogen removal via denitrification in these NO_3^- -rich streams (Jansson et al., 1994). Hence, Washington's more than 2.4 million hectares of cultivated land (USDA/NRCS) are likely to have a strong influence on the state's surface water quality, and understanding the controls over NO_3^- , DOC and DON loss from these systems is critical for understanding and anticipating the effects of climate change on water quality. Current understanding of dissolved nitrogen dynamics is based almost entirely on warm-season studies of transport and in-stream processing. However, in the semi-arid region of eastern Washington, higher precipitation and discharge occur in the cooler winter months, causing the majority of NO_3^- and DOM mass flux from watersheds to occur during periods that have thus far been

largely unstudied.

Both NO_3^- and DOM are likely to be quite sensitive to changes in the frequency and intensity of hydrologic pulse events. Such events not only increase water discharged from soils to streams, but are also often associated with increased in-stream NO_3^- and DOM concentrations (e.g. Harrison and Matson, 2003; Martin and Harrison, 2011), such that high flow events of relatively short duration (days to weeks) can account for the majority of annual DOM and NO_3^- flux through streams (e.g. Inamdar et al., 2006, Bernal et al., 2005). Increased DOM and NO_3^- concentrations during

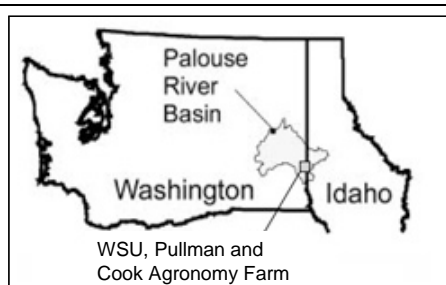


Figure 1. Location of Cook Agronomy Farm at Washington State University, after Keller et al. 2008.

hydrologic pulses can result from many factors, including: 1) changing hydrologic flow paths that mobilize novel sources of NO_3^- (e.g. Tiemeyer et al., 2008) and DOM (e.g. Hagedorn et al., 2000, Martin and Harrison 2011), and 2) the bypassing of environments (e.g. wetlands or riparian zones) or processes (e.g. plant or microbial uptake) which would otherwise retain these compounds. Much remains to be learned about interactions between hydrologic pulse events and land-to-water transport of DOM. Similarly, the relationship between such pulse events and in-stream DOM and NO_3^- processing is poorly characterized, especially in agricultural settings, and interactions between delivery of DOM and in-stream NO_3^- and DOM processing are almost completely uncharacterized despite the fact that they are likely to be important, and of increasing importance as the frequency and intensity of pulse events increases with climate change.

It is in this context that we proposed work that would enhance the ability of scientists, policy makers and stakeholders to understand, predict, respond to and/or mitigate climate change impacts on water quality. Specifically, we proposed to use WSU's Cook Agronomy Farm (Figure 1) as a study system to: 1) understand how hydrologic variability affects a) nitrate and DOM transport from agricultural fields to surface water and b) in-stream fate of nitrogen, and 2) use this information to develop, apply, test, and iteratively refine a model that utilizes a dynamic representation of hydrologic flow paths and organic matter source pools to predict terrestrial-to-aquatic NO_3^- and DOM transport, under current and anticipated future climate.

Specifically, we addressed the following questions:

- Q1.) How do hydrologic conditions affect nitrate and DOM transport from agricultural land to surface drainage waters?**
- Q2.) How do hydrologic conditions affect in-stream nitrogen retention?**

METHODOLOGY

Soil water and tile drain sampling

Nests of shallow and deep wells and lysimeters were installed along a transect that approximated the buried tile drain route and upslope of the tile line. Shallow and deep lysimeters were placed at 0.5 and 1 m depths, respectively, and shallow and deep wells were screened at depths above and below an argyllic layer, respectively, that is approximately 1 meter below the surface and intermittent across the basin.

Soil water, tile drain, and surface water samples were collected weekly to bi-weekly during the 2012 water year, and tile drain samples were collected more frequently during high flow events to capture temporal variability associated with rapid changes in discharge. During the dry season, most shallow lysimeters and wells did not yield water samples. Specific electrical conductivity was measured in the field using an Orion Model 115 with Conductivity Cell 014016 probe. Samples for DOM, nutrients, and major cations were filtered in the field, transported on ice to the laboratory, and frozen until analysis. Samples to be analyzed for absorbance and fluorescence spectra were stored at 4°C and analyzed within 5 days. Discharge from the tile drain was monitored every 15 minutes in a receiving flume equipped with a pressure transducer, and electrical conductivity was measured simultaneously with a Campbell Scientific Temperature and Conductivity probe (CS547A-L).

Surface Water Monitoring

Surface water has been sampled at 3 locations along Missouri Flat Creek since 2000 to capture nitrate export dynamics at a range of catchment sizes [660 (site 1), 3800 (site 2), 6300 (site 3) ha; the smaller catchments are nested within the larger catchments]. Samples were taken approximately twice a month all three sites, with more frequent sampling during the winter/spring runoff period. Stream discharge has been monitored at sites 1 (2000-present) and 2 (2000-2010) using digital pressure transducers in combination with rating curves. Discharge at site 3 was measured with digital pressure transducer prior to 2004 and modeled for years 2005-2010 based on discharge data at site 2 during that period and the relationship between discharge at sites 2 and 3 prior to 2004.

Laboratory Nutrient and Dissolved Organic Matter Analyses

Samples were analyzed for nitrate and ammonium according to the standard EPA methods (353.2 and 350.1, respectively) using a continuous flow analyzer (Model RFA300, Alpkem/OI Analytical) or discrete nutrient analyzer (WestCo Smartchem) and for dissolved organic carbon (DOC) and total dissolved nitrogen (TDN) on a Lachat TOC-TN analyzer (IL 550 TON-TN) equipped with electrochemical (ECD) NO and non-dispersive infrared absorption (NDIR) detectors. Dissolved organic nitrogen (DON) was calculated as the difference between TDN and total inorganic nitrogen ($\text{NO}_3^- + \text{NH}_4^+$). Samples were analyzed for major cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+}) using a Shimadzu atomic absorption emission spectrophotometer (model AA-6601F). Absorbance spectra of bulk DOM samples were analyzed using a J&M TIDAS spectrophotometer (World Precision Instruments) from 200-700 nm. Fluorescence excitation-emission matrices (ex 240-450 nm, em 300-600 nm) were generated with a Horiba Jobin Yvon Fluoromax-4 spectrofluorometer. Corrections for the instrument, water matrix (background), internal absorbance, and Raman signal were applied to fluorescence data to permit comparisons across sampling dates.

Estimating DOM and nitrate export via tile drainage

Annual DOC, DON, and nitrate loads were estimated by assuming that concentrations were constant from halfway between the previous measurement to halfway to the subsequent measurement. Concentrations were multiplied by water discharge, and summed for the year. The hydrograph was then characterized as either baseflow or event-flow, and export during each of these conditions was calculated. An event were defined as the time from the start of rapid discharge increase to the time when steady base flow returned.

DOM Export Modeling Approach

The spatial and temporal variability of the soil water data was too great to constrain shallow and deep soil water end members and meaningfully estimate the contribution of each flow path to tile drain discharge over time, with spatio-temporal variability of individual end members exceeding temporal variability of tile drain chemistry. Despite this, a mixing model was developed to qualitatively assess whether our hypotheses were broadly supported, whether various soil water compartments may be contributing preferentially to tile drain discharge over time (e.g. due to varying hillslope hydrologic connectivity), or other processes might be important for controlling DOM export via the tile drain (e.g. DOM removal via adsorption or

decomposition). Further analyses, including sensitivity of DOM export to changing climate, were not conducted because we could not obtain statistically meaningful results.

Shallow and deep soil water end members were characterized by both specific electrical conductivity (EC) and magnesium concentrations ([Mg]), which were significantly correlated with [DOC] in tile drain discharge ($p < 0.001$ for both relationships). Median EC and [Mg] values for each sampling instrument during dry and wet seasons were calculated; medians were then averaged to define the end members. During the wet season (Jan through May), EC was significantly greater in deep than shallow soil water (T-test, one-tailed, $df = 9$, $p < 0.05$), and [Mg] was borderline significantly greater in deep than shallow soil water (T-test, one-tailed, $df = 9$, $p = 0.05$) (Table 1). One shallow sampler yielded soil water during the dry season, and the median EC and [Mg] values of this sampler were within the range defined by the mean ± 2 sd of the deep samplers during the dry period. Additionally, EC and [Mg] values for tile drain samples were outside the end-member range during the dry season, so further assessment of dry-season dynamics was not conducted.

For tile drain sampling time points during the wet season, the fractions of discharge derived from shallow and deep soil water were calculated using a linear mixing model with shallow and deep end members defined by EC and [Mg], yielding two estimates for shallow and deep fractions per sampling time, which were then averaged. Average end member fractions were used to predict [DOC] and [DON] for each sampling time, and Nash-Sutcliffe Efficiency and model error in relation to tile drain discharge were examined to assess model performance.

Nitrate fluxes in nested catchments

Nitrate mass discharge (nitrate-N mass/time) and fluxes (nitrate-N mass/area/time) were calculated for the period 2000-2010 by dividing nitrate-N concentrations by water discharge (nitrate-N mass discharge) and dividing the nitrate-N mass discharge by watershed area (nitrate-N flux). At least one nitrate measurement per month and eight months per year were required to generate annual nitrate loads and fluxes for each location. Since the majority of nitrate is exported during high flows, low estimates for nitrate loads and fluxes during some years may be due to sampling bias during low flow periods, when high flow periods were not captured in the sampling regime.

PRINCIPLE FINDINGS AND SIGNIFICANCE

Annual and event dissolved N export via tile drainage

Dissolved N leaching is a concern in many agricultural regions, but particularly in the Pacific Northwest, where the Mediterranean climate results in the most hydrologic transport during cold periods when vegetation and microbial activity are limited. We found that the annual nitrate flux from the tile drain during the 2012 water year was $13.2 \text{ kg N ha}^{-1} \text{ y}^{-1}$, with 84% occurring during winter/spring runoff events. The annual DON flux was $0.7 \text{ kg N ha}^{-1} \text{ y}^{-1}$, with 71% occurring during high flow events. Combined, total dissolved N losses account for $\sim 10\%$ of the average N fertilizer applied to the tile-drained area (T. Brown, personal communication). Tile drain discharge and $[\text{NO}_3^-]$ were significantly, positively correlated ($p < 0.001$; discharge \ln -transformed for normality), and the time-averaged nitrate concentration was 9.02 mg N L^{-1} – near the EPA limit for drinking water standards and ten times higher than reference conditions described by EPA for this ecoregion. These results highlight the importance of high flow events

for delivery of dissolved N to streams in quantities that can cause environmental degradation and human harm.

Annual and event DOC export via tile drainage

DOC export from agricultural systems to surface water is poorly understood, particularly outside of the Midwest where most of the limited research has been conducted to date (e.g. Royer and David 2005, Dalzell et al. 2007, Warrner et al. 2009, Dalzell et al. 2011). Potential sources of DOM to the tile drain were characterized throughout the year, including soil water in top-soil (“shallow”) and sub-soil (“deep”). DOC concentrations exhibited high spatial-temporal variability over space and time in the basin; however, patterns were discernable, with DOC concentration generally decreasing with depth (Figure 2). Additionally, the DOM absorbance and fluorescence data indicate that DOM quality varies consistently with soil depth (Figure 2).

In this study, we found that annual DOC flux from the tile drain was 3.5 kg C ha⁻¹ y⁻¹, with 87% occurring during high flow events; DOC concentration increased with tile drain discharge, consistent with the hypothesis that shallow soil water is contributing more to tile drain discharge during high flow, although discharge did not explain a large amount of variation in DOC concentration (p < 0.001, r² = 0.17). Additionally, the quality of DOM in tile drain discharge was significantly correlated with flow rate (Figure 3), further supporting the hypothesis that DOM sources to the tile drain shift with hydrologic conditions. Similar to nitrate and DON export, these data emphasize that high flow events dominate export, and increases in precipitation due to climate change may result in a non-linear increase in delivery of DOC to surface waters.

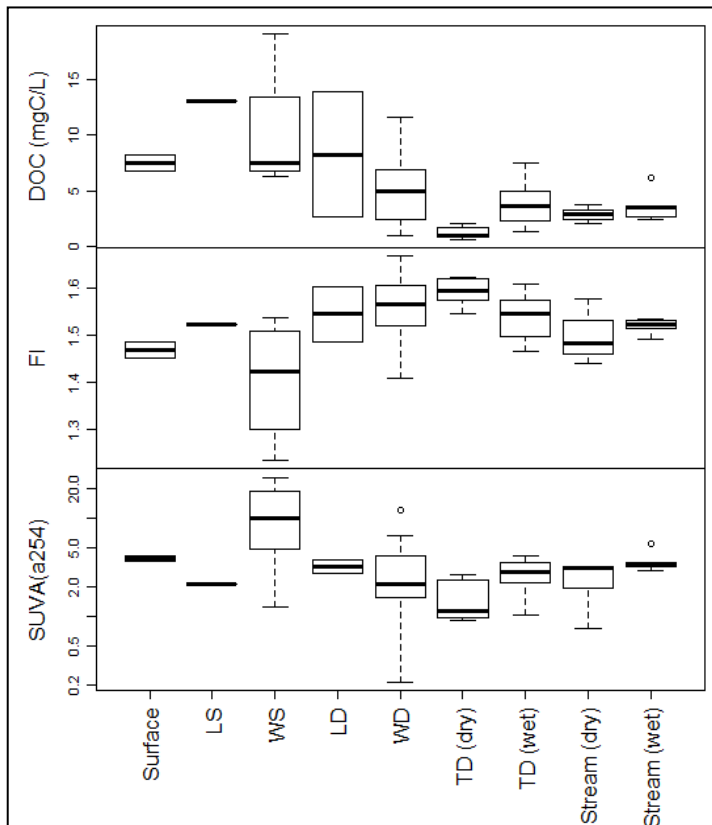


Figure 2. [DOC] and DOM quality indices across a vertical hydrologic gradient (organized from high elevation (left) to low elevation (right) on the x-axis). X-axis labels represent: (Surface) is surface runoff, (LS) shallow lysimeter, (WS) shallow well, (LD) deep lysimeter, (WD) deep well, (TD) tile drain, and (Stream) receiving stream (Missouri Flat Creek) samples. Tile drain and stream samples were separated by wet and dry seasons. (FI) Fluorescence index (FI) is traditionally used to distinguish plant-derived from microbially-derived DOM, with higher values representing microbially-derived DOM. Specific UV absorbance at 254 nm (SUVA₂₅₄) is a proxy for aromaticity of DOM, with higher values indication more aromatic

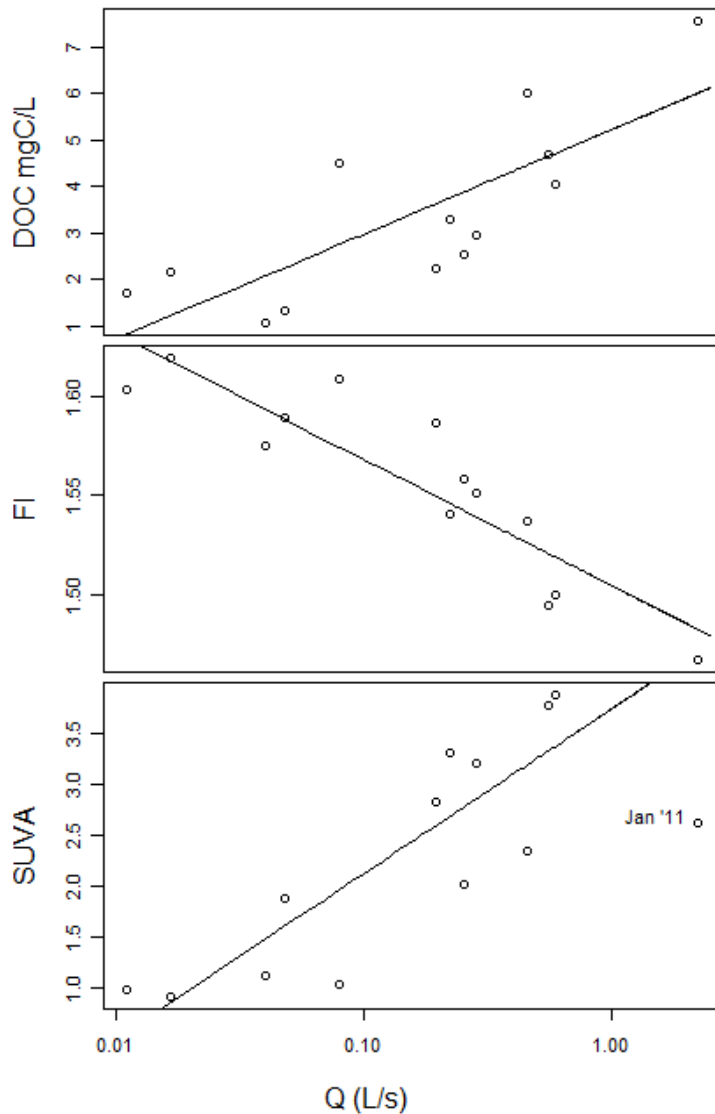


Figure 3. Relationships between DOC concentration (top), Fluorescence Index (FI) (middle), and $SUVA_{a254}$ (bottom) and tile drain discharge (Q) were all significant ($r^2 = 0.61$, $p = .002$ for [DOC]; $r^2 = 0.80$, $p < 0.001$ for FI; $r^2 = 0.74$, $p < 0.001$ for SUVA, with the point from Jan 2011 removed) Discharge was ln-transformed for all regression to meet normality assumption.

The range of DOC concentrations observed in this study is similar to those found in other Midwestern tile drains (e.g. Ruark et al. 2004; Warner et al. 2009; Dalzell et al. 2011). FI and SUVA values are also similar, although we report a wider range than the others (Warner et al. 2009; Dalzell et al. 2009). For context, the range in SUVA observed in this one site encompasses the lower 50% of values observed across 12 stream LTER stations, while FI values are more constrained relative to variability observed across LTER stations (Jaffe et al. 2008).

DOM Model Results

The mixing model predicts greater shallow contribution to tile drain flow during high discharge periods (Figure 4), but predicts [DOC] and [DON] in discharge poorly. The Nash-Sutcliffe Efficiency (NSE) for the DOC model is 0.05, indicating that the model is only marginally better at predicting [DOC] than the mean value; NSE for the DON model is -0.03; indicating that the model is worse at predicting [DON] than a simple mean. Additionally, the model systematically over-predicts [DOC] under low flow conditions (Figure 5).

Table 1. Average solute and electrical conductivity of wet season shallow and deep soil water end members. Values in bold are significantly different between deep and shallow sources (one-tailed t-test, $p < 0.05$)

	Shallow	Deep
Mg (mg/L)	17.3	27.6
EC (uS/cm)	240	409
DOC (mg C/L)	6.2	4.0
DON (mgN/L)	0.9	0.4

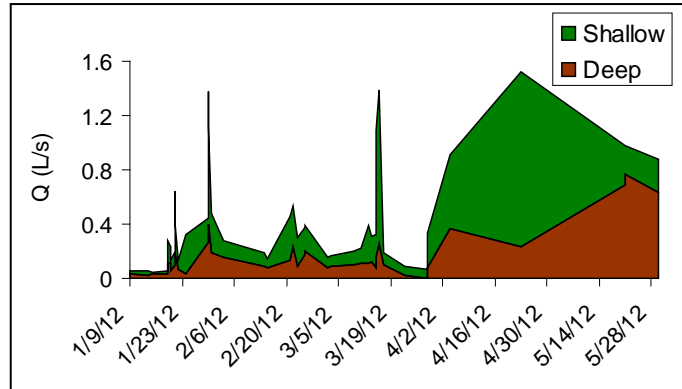


Figure 4. 2012 runoff season hydrograph partitioned into the contributions of deep and shallow soil water to tile drain discharge.

These results point to potential problems with the model and provide a basis for future investigations. First, the model may be missing important end members, as the [DOC] and [DON] values of model end members do not encompass the range of values seen in the tile drain discharge. For instance, an end member with lower “tracer” concentrations and higher [DOC] may be transiently available following precipitation or melt events when event water mobilizes and transports previously disconnected DOM pools to the tile drain rapidly through macropore flow. Second, the model accounts for vertical, but not lateral, expansion and contraction of hydrologic connectivity in the basin. High (lateral) spatial variability observed in soil water

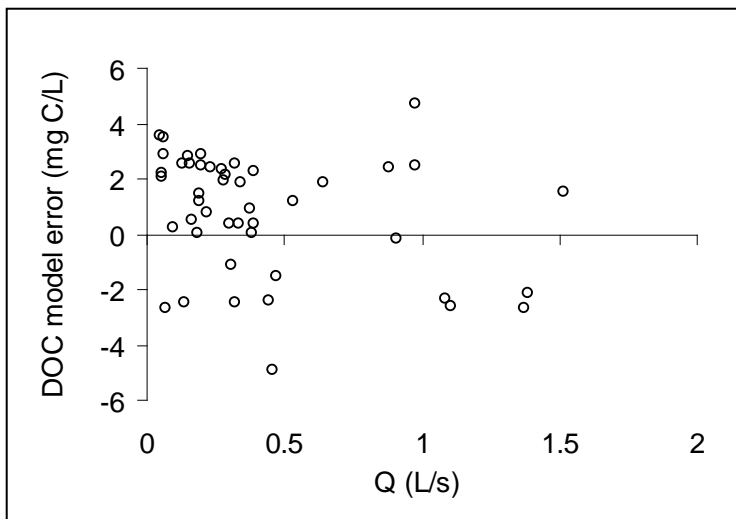


Figure 5. DOC model error relative to tile drain discharge. DOC is systematically over predicted under low discharge conditions.

chemistry is problematic for interpreting model results; however, soil water chemistry in low-elevation samplers do to correlate better than upslope samplers with tile drain discharge chemistry. Finally, the model assumes no removal of DOM along flow-paths en route to the tile drain outlet. The systematic over-prediction of [DOC] at low discharge suggests that removal processes may be important for controlling export under these conditions. Understanding adsorption and decomposition rates along various flow paths may be critical to effectively predict DOM export to surface water.

Controls over nitrate export from and attenuation within the watershed

Annual export of nitrate from the watershed is largely controlled by infrequent (2-7 per year) winter storm events. These large winter storms, which occur with very large instantaneous

fluxes (Figure 6), transport 75-99% of the total annual export of nitrate; single large flood events can account for the export of up to 5-10% of the annually applied N fertilizer. Precipitation during the study period is an underrepresentation of long-term precipitation averages in the region, and, given that eastern Washington is projected to see increases in heavy precipitation and fall precipitation (Meehl et al., 2005, Salathe et al., 2010), estimates of nitrate export from the study period may be an underrepresentation of future export from the region. The disproportionate importance of high flow events requires targeted sampling during high winter flows to better constrain the overall nitrogen budget.

Little research has focused on understanding how retention or removal of nitrate from agricultural streams may be affected by DOM availability, for instance, by fueling in-stream denitrification. During summer flows, nitrate concentrations and mass discharges decrease in the downstream direction (Figure 7). Given that there is less mass of nitrate at downstream locations (removal of nitrate mass from the system), this decrease in concentration is likely due to

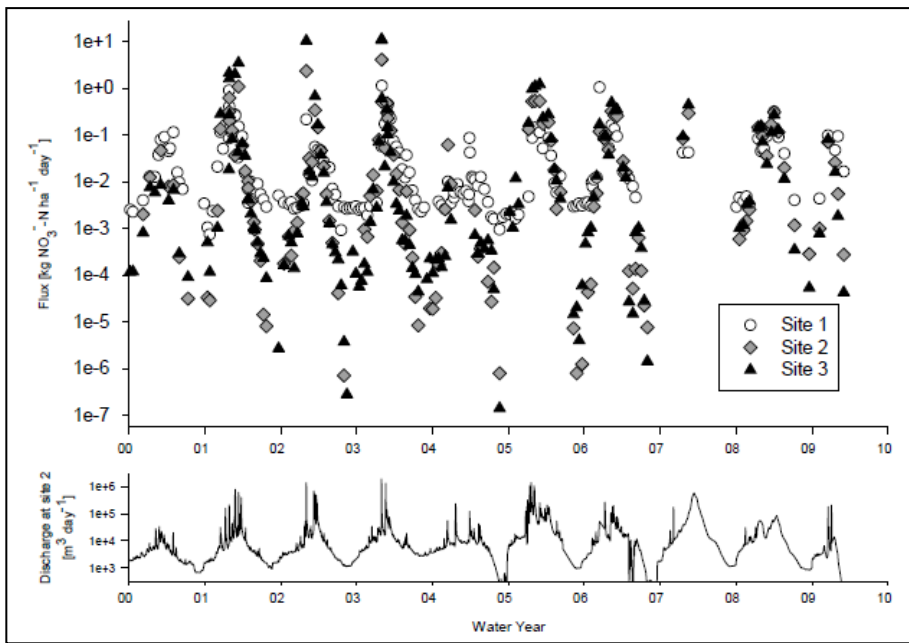


Figure 6. Flux of NO_3^- -N at three sampling sites. Fluxes of NO_3^- -N can be as high as 12 kg NO_3^- -N $\text{ha}^{-1} \text{ d}^{-1}$ during large discharges. In general, site 3 fluxes are larger than sites 1 and 2 during the winter months, and smaller than site 1 during summer months. Out of the three sites, site 1 generally has the largest flux during the summer months, though the overall difference is quite small.

dormant winter season (C.J. Kelley thesis, 2011). Given that nitrate removal in soils and in the stream channel is likely to be small during the winter, the most feasible means to control N in streams is by managing on-field application timing and rates. Without adjusting management strategies, increases in the quantity and intensity of fall and winter precipitation will lead to larger N export from the system.

biological uptake. Low flow may be a prime time to further investigate a biological signal. In contrast, we found little evidence of nitrate removal along the stream during winter conditions, suggesting that DOM availability likely has little role in any biological controls of in-stream nitrate dynamics during this period, when low temperatures and fast moving water can mean limited substrate interaction and sub-optimal conditions for biologic activity. Further, evidence from another study at Cook Farm indicates that removal of nitrate by a buffer strip along the stream is minimal during the

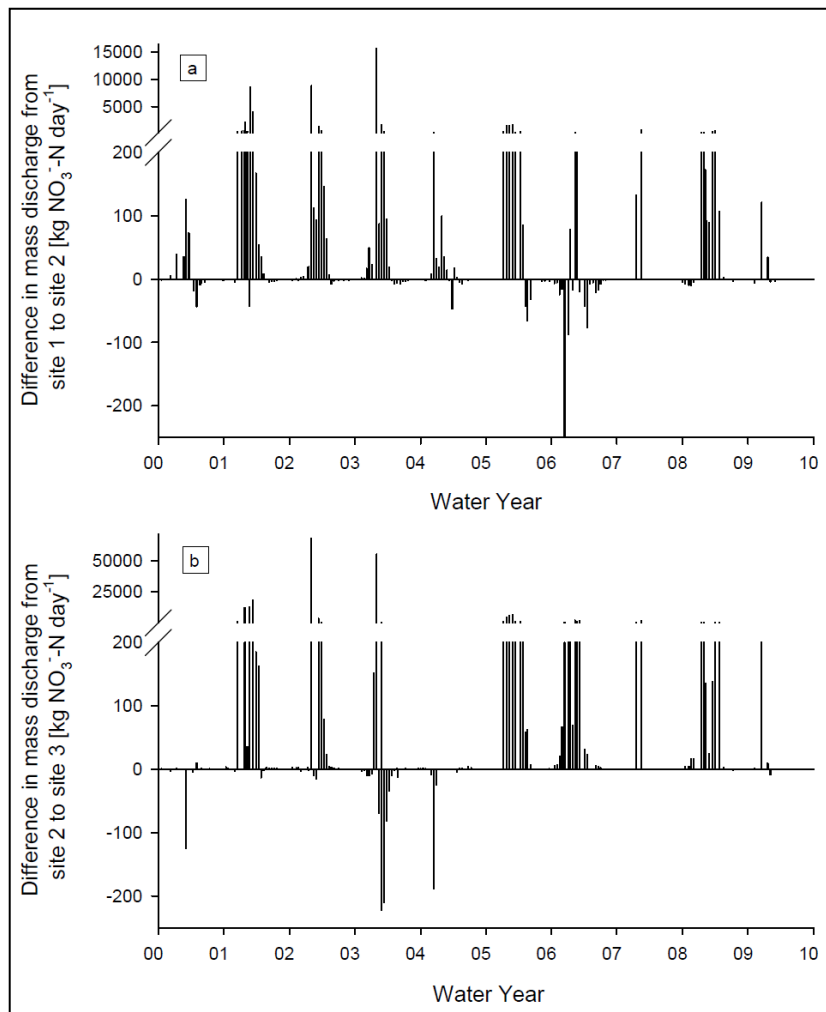


Figure 7. Changes in mass discharge of NO_3^- -N in the downstream direction. The x-axis is in water years, which start on October 1 of the year shown. (a) Mass discharge increases from site 1 to site 2 during most of the year, but differences are negligible or decrease during low flow summer months. (b) Mass discharge shows increases from site 2 to site 3 during large flows, and some losses during summer months, but the relationship between gains and losses and small and large flows is less clear.

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