

## Seasonal dynamics of dissolved nitrogen species in two High Arctic rivers, Melville Island, Canada

Melissa Lafrenière

*Department of Geography, Queen's University, Kingston, ON, K7L 3N6 CANADA*

*\*Corresponding author, e-mail: [melissa.lafreniere@queensu.ca](mailto:melissa.lafreniere@queensu.ca)*

### ABSTRACT

This study examines the magnitude, seasonal patterns, and characteristics N export from two small watersheds on Melville Island, in the Canadian High Arctic. The dominant N species in both rivers was dissolved organic nitrogen (DON), comprising >80% of the seasonal nitrogen flux from both rivers. The total DON and dissolved inorganic nitrogen (DIN) mass fluxes from the two catchments were similar (242 and 245 kg DON, and 42-45 kg DIN), but there were differences in the patterns and concentrations of the DIN species in the two catchments. The West river had higher initial DON and DIN concentrations that decreased to stable concentrations around 0.150 ppm and 0 ppm by mid July. The East river had variable early season DON and NO<sub>3</sub><sup>-</sup> concentrations ranging from 0.138-0.415, and 0-0.125 ppm, respectively. End of season DON and DIN concentrations increased from 0.134 to 0.240, and 0 to 0.085 ppm, respectively. The DOC:DON ratio also decrease from 13 to 6, indicating a change in the composition of dissolved organic matter in this river at the end of season. The total DON fluxes from these two small arctic watersheds are similar to fluxes reported for other catchments in temperate environments.

### KEYWORDS

Nitrogen, High Arctic rivers, nutrient fluxes, DON, NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>.

### 1. INTRODUCTION

Fluvial input of dissolved organic matter (DOM) represents an important source of carbon and bioavailable nitrogen (N) to freshwater and marine ecosystems (Cole, J. J. and Caraco, N. F. 2001, Cornell, S., *et al.* 1995, Dittmar, T. 2004, Mayorga, E., *et al.* 2005, Pace, M. L., *et al.* 2004). Because N is often the limiting nutrient for phytoplankton growth, the export of nitrogen species (including DON, NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup>) from the terrestrial catchments plays an important role in aquatic elemental cycles (Dittmar, T. 2004). Two of the primary controls on the export of N from the land surface are the volume and the routing of catchment runoff through shallow soils (Cooper, R., *et al.* 2007, Hagedorn, F., *et al.* 2000, Hood, E., *et al.* 2003a, Perakis, S. 2002, Pinay, G., *et al.* 2002). Hence, there is a need to understand how the hydrologic components of the climate system influence the interactions between terrestrial nitrogen sources, and the leaching or export of N in stream runoff

Arctic catchments store a significant proportion of the world's soil organic matter (Dittmar, T. and Kattner, G. 2003) and Arctic rivers yield large amounts of terrigenous organic matter relative to other river basins (Hansell, D., *et al.* 2004). In addition, observations indicate that the central Arctic has experienced substantial warming over the 20<sup>th</sup> century and models project this warming trend will continue over the next century (ACIA 2005, IPCC 2001, Serreze, M. C., *et al.* 2000). Observations of recent changes in precipitation and snow cover suggest that precipitation in the Arctic appears to have increased, while snow cover duration and extent have decreased in most areas (ACIA 2005, IPCC 2001, Serreze, M. C., *et al.* 2000) Through their effect on runoff (timing and quantity), permafrost degradation, and slope stability, these changes in climatic conditions will exert significant influence on nutrient dynamics in terrestrial catchments. There is, therefore, a pressing need to understand the response of terrestrial nutrient dynamics to changing water, permafrost and climate regimes in the High Arctic.

This study examines the magnitude, seasonal patterns, and characteristics N export from two small watersheds on Melville Island, in the Canadian High Arctic. Specifically, the paper discusses the difference in controls on nitrogen concentrations and fluxes from these adjacent watersheds, and evaluates the significance of these fluxes with respect to other temperate environments.

## 2. METHODS

### 2.1 Site Description

The research site is Cape Bounty (74°54N, 109°35W), Melville Island, Nunavut (Figure 1). This study reports on the rivers draining the two main watersheds unofficially named West (8.0 km<sup>2</sup>) and East (11.6 km<sup>2</sup>). The two catchments are topographically similar, consisting of rolling terrain, with elevations ranging from approximately 5 m.a.s.l. to 165 m. a.s.l. Both streams are incised into the surrounding terrain and drain into similar small lakes (Figure 1). The study area is underlain by continuous permafrost that develops an active layer ca. 0.5–1 m deep during the spring and summer. The geology consists of steeply dipping Devonian sedimentary rocks that are covered with glacial and regressive early Holocene marine sediments (Hodgson, D. A., *et al.* 1984). Vegetation and soil cover is heterogeneous and varies with moisture conditions. The upland regions may be classified as polar desert and generally lack soils and vegetation, while the wetter lowland areas have thin soils and patchy dwarf prostrate shrub tundra vegetation (Walker, D. A., *et al.* 2005).

### 2.2 Field methods

Local meteorological conditions during the period of study were captured by two weather stations in the catchment. For the spring and summer of 2006 Westmet (Figure 8) measured temperature at 1.5 m above the ground (0.4°C accuracy) and precipitation (0.2 mm resolution). In addition to temperature and precipitation, Mainmet (Figure 1), monitored solar and net radiation, wind speed and direction, and relative humidity.

Hydrological measurements for both streams were obtained from gauging stations near the lake inlets (Figure 1). Stage was recorded with a logging pressure-transducer and was compensated for atmospheric pressure changes using a second pressure-temperature logger in each river. Discharge was rated at each gauging station using manual area-velocity measurements obtained throughout the seasons with a Swiffer 2100 current velocity meter. Errors on the discharge measurements are estimated to be on the order of 10-15%.

End of winter snowpack depth (snow water equivalent SWE) for 2005 and 2006 was calculated using a terrain classification model to spatially average measurements from 42 transects (McLeod, B., *et al.* 2005). Each transect was 100m in length, depth measurements were taken every 10m, and density at 0, 30, 50, 70, and 100m. Each transect was marked at each end with a fixed stakes at the time of measurement in 2005. Most of the stakes (38 of 42 transects) were in place upon return in 2006, hence 90% of the measurements were repeated at the same locations each year ( $\pm 10$ m). For 2003 and 2004, end of winter SWE were calculated based on a network of only 13 transects (which were part of the 42 transects measured in subsequent years) (Lamoureux, S. F., *et al.* 2006). Transects were 100m in length, with depth measured every 10m, and one density measurement at the centre of the transect. Mean SWE was calculated for each transect and spatially averaged for each watershed on the basis of terrain type units (channel, slopes, and plateau).

Water samples were collected in amber (HDPE) bottles that were triple rinsed with deionized (DI) water, and then triple rinsed with stream water prior to sampling. The bottles were filled completely to eliminate headspace in the bottle, and transported back to Camp for filtration and processing (Figure 1). All samples were filtered and bottled within 2 hours of collection, stored in snow packed coolers, and refrigerated upon return to the lab at Queen's University. Sample aliquots for dissolved organic carbon (DOC) and total nitrogen (TN) analysis were filtered using pre-combusted glass fiber filters (GF/F) and glass filtration apparatus. The glass filtration apparatus was acid washed and combusted before the field season, then soaked in 30% hydrogen peroxide overnight at the end of each day, and rinsed three times with DI water and sample between each sample. Filtered sample for DOC/TN was collected in 40ml amber glass EPA vials, with Teflon lined septa, and acidified with hydrochloric acid immediately upon return to the lab in Kingston (within 30 days of sampling). Aliquots for dissolved inorganic ion

analyses were filtered through 0.45  $\mu\text{m}$  nitrocellulose membrane filters, using a polysulfone filter holder. Ion samples were bottled in plastic scintillation vials.

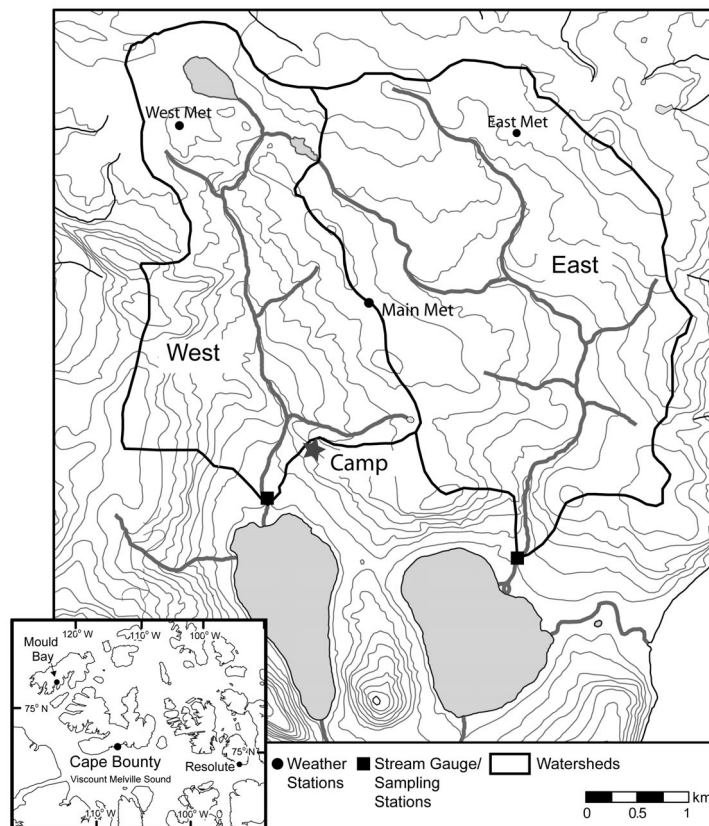


Figure 1. Topographic map of the West and East watersheds at Cape Bounty, illustrating the locations of the weather stations and stream gauging and sampling sites.

### 2.3 SAMPLE COLLECTION AND ANALYTICAL METHODS

DOC and TN were determined simultaneously by high temperature combustion and NDIR and chemiluminescent detection using a Shimadzu TOC-VPCH/TNM system equipped with high sensitivity catalyst. Error on these analyses were determined to be less than 1% ( $\pm 0.020$  ppm) for DOC and less than 2% ( $\pm 0.01$  ppm) for TN based on duplicate analyses of replicate field samples, and replicate analysis of standards.

Dissolved inorganic anions and cations (including  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Mg}^{2+}$ ,  $\text{Ca}^{2+}$ ,  $\text{Cl}^-$ ,  $\text{Br}^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{PO}_4^{3-}$ ) were measured by ion chromatography. The anions and cations were determined simultaneously on separate systems using a Dionex 3000CS system. The anions were separated by gradient elution with 23-40 mM KOH (using an EG II KOH), with a 1.0 ml/min flow rate, and AS18 analytical and guard columns, and self regenerated suppression (ASRS-ULTRA II). Cations were measured isocratically with 23 mM methanesulphonic acid eluent, flowing at 0.5 ml/min with a CS12A analytical and guard columns with self regenerated suppression (CSRS-ULTRA II). Errors on most analytes were less than 1% based on replicate analysis of samples and standards, while errors on  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ , and  $\text{NH}_4^+$  were 1.7% ( $\pm 0.002$  ppm), 20.7% ( $\pm 0.003$  ppm) and 5% ( $\pm 0.002$  ppm), respectively. DIN is reported as the sum of the nitrogen mass from  $\text{NO}_3^-$ ,  $\text{NO}_2^-$ , and  $\text{NH}_4^+$ , while DON was determined as the difference between the TN and DIN (propagated error on DON  $\pm 0.012$  ppm; approximately 5%).

## 2.4 Mass Flux Calculations

The yields of the DOC and N species were estimated as the product of the measured concentrations and the mean discharge over the period of measurement. In the case where there was only one measurement per day the flux was determined as the concentration times the total mean daily discharge (i.e. mean daily Q in  $\text{m}^3/\text{s} \times 60 \text{ s}/\text{min} \times 60 \text{ min}/\text{hr} \times 24 \text{ hr}/\text{d}$ ). Where more than one measurement was available the flux was determined as the product of the concentration and the total discharge accumulated over the time period represented by that sample 12-18hrs, depending on the time of measurement. The overall error on involved in calculating the seasonal mass flux is estimated to be within 15-18%.

## 3. RESULTS AND DISCUSSION

### 3.1 Climate and Hydrology

The snow accumulation at the end winter 2006 was the highest observed at this site over the last four years (Table 1). The June air temperatures were near the mean of the past four years, however July 2006 was quite warm relative to the previous three years. The high snowfall yielded the highest discharges recorded over the four years of observation (Table 1).

Table 1: End of winter SWE for West and East catchments and mean June and July air temperatures. Data for 2003, 2004 from Lamoureux *et al.* (2006).

Year	SWE (mm) West	SWE (mm) East	West River Discharge (mm)	June Temp °C	July Temp °C
2003	43	20		-1.0	
2004	82	41	101	-0.1	3.1
2005	70	40	86	2.0	3.1
2006	120	120	171	1.0	6.2

Seasonal discharges in the two streams were similar in 2006 (Figure 2). Both streams began to flow June 18<sup>th</sup> and peaked June 26<sup>th</sup>, however the East river had a higher maximum hourly discharge ( $3.06 \text{ m}^3/\text{s}$ ) than the West River ( $2.44 \text{ m}^3/\text{s}$ ). The total runoff from the two catchments was  $1.40 \times 10^6 \text{ m}^3$ , or on a catchment area basis 170 mm from the West, and 120 mm from the East.

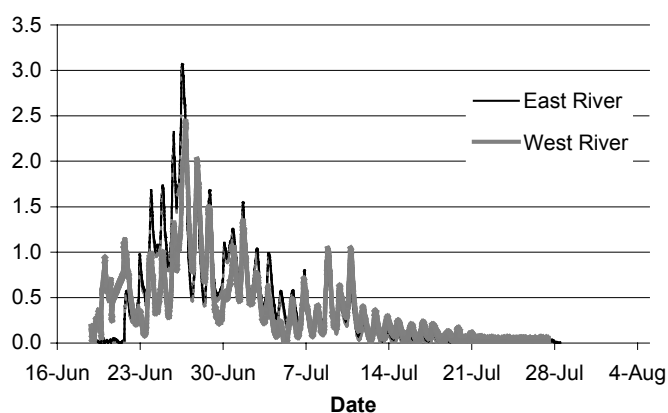


Figure 2. Mean hourly discharge ( $\text{m}^3/\text{s}$ ) for West and East rivers 2006.

### 3.2 N Species concentrations and mass fluxes

The dominant N species in both rivers was DON, which comprised approximately 85% of the seasonal nitrogen mass flux from both rivers (Table 2). The mean DON and DIN concentrations and total mass fluxes for the two catchments were also very similar, but the mean concentrations of the different inorganic N species (especially nitrate ( $\text{NO}_3^-$ ) and ammonium ( $\text{NH}_4^+$ )) for the two rivers were somewhat different

(Table 2). In both streams nitrite ( $\text{NO}_2^-$ ) was the least abundant of the inorganic N species, but it was slightly more abundant in the East river than in the West. The West river exhibited a slightly higher mean ammonium concentration (0.018 ppm N) than the East (0.011 ppm N), while the East river had much a higher mean concentration of nitrate (0.024 ppm N) than the West (0.011 ppm N, Table 2). Due to the differences in the size of the two catchments, the West catchment is found to export more nitrogen per unit area (30.6 kg DON/km<sup>2</sup> and 5.6 kg DIN/km<sup>2</sup>) than the East catchment (21.1 kg DON/km<sup>2</sup> and 3.9 kg DIN/km<sup>2</sup>).

Table 2: Arithmetic mean concentrations and seasonal mass flux of nitrogen species and DOC in the West and East rivers.

	DOC ppm	DON ppm	DIN ppm	N- $\text{NO}_3^-$ ppm	N- $\text{NO}_2^-$ ppm	N- $\text{NH}_4^+$ ppm	DOC: DON	DOC kg	DON kg	DIN kg
West	2.63	0.180	0.031	0.011	0.002	0.018	14.6	3770	245	45
East	2.50	0.200	0.040	0.024	0.006	0.011	12.8	3820	248	42

The West river generally had lower mean DIN and DON concentrations than the East, and ammonium concentrations were often higher than nitrate in the West river (Figure 3). For all the N species, concentrations in the West river were at a maximum at the onset of melt, and decreased exponentially as discharge increased to peak flow (Figure 3). After the peak DON and DOC concentrations continued to decrease slightly, then stabilized or increased slightly towards the end of the season. The DIN concentrations were somewhat different, the DIN concentration was highest in the first sample, the concentration dropped significantly in the second sample, then the DIN concentrations increased with discharge until just before peak (Figure 3a). Figure 4 illustrates that the nitrate concentrations follow a trend that is similar to DOC and DON, and that  $\text{NH}_4^+$  is the species driving the increase in DIN with discharge in the early season (Figure 4a). Correlation analysis revealed that in the West river,  $\text{NO}_3^-$  and  $\text{NH}_4^+$  were well correlated with DOC (correlation coefficient  $r = 0.81$  and  $r = 0.69$ ). However, DON was only weakly correlated with DOC ( $r = 0.59$ ). In the West river  $\text{NO}_3^-$  was most strongly correlated with DOC ( $r = 0.81$ ) and  $\text{K}^+$  ( $r = 0.81$ ).

In the East river, concentrations are also at a maximum at the onset of melt, and although the DOC and DON concentrations are variable near the peak in discharge, they remain high on average (Figure 3b). In this stream both DON and DIN concentrations increased gradually after approximately July 14<sup>th</sup>, while DOC concentrations decline. Figure 4b shows that it is an increase in nitrate concentrations at the end of the season that is driving the increase in DIN. In the East river there is only a weak correlation between DOC and DON ( $r = 0.44$ ) and little to no correlation between DON and  $\text{NO}_3^-$  and  $\text{NH}_4^+$  ( $r = 0.22$  and  $r = -0.49$ , respectively). Here  $\text{NO}_3^-$  was most strongly correlated with the inorganic ions  $\text{Cl}^-$  ( $r = 0.92$ ),  $\text{Na}^+$  ( $r = 0.86$ ) and  $\text{K}^+$  ( $r = 0.83$ ), and showed no correlation with DOC ( $r = -0.06$ ).

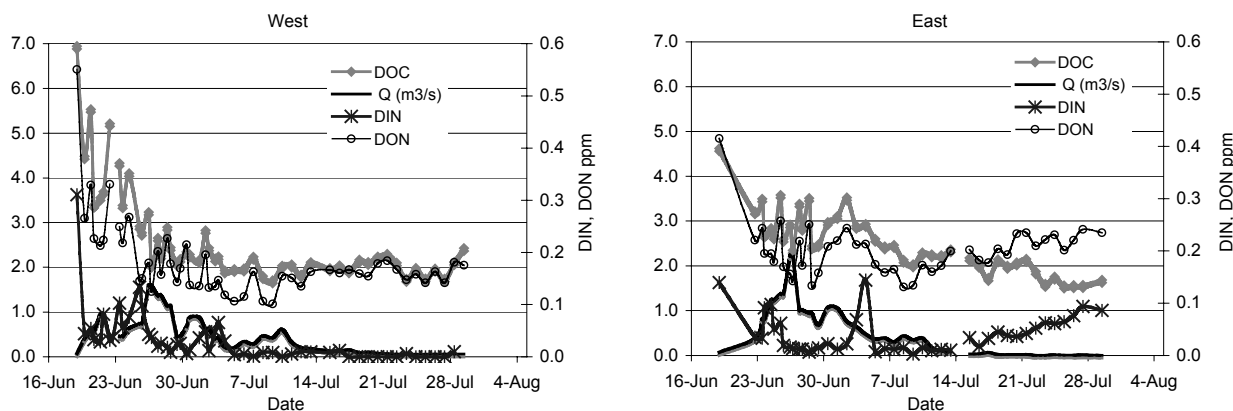


Figure 3. Seasonal variations in concentrations of DOC, DIN and DON plotted with mean daily discharge (m<sup>3</sup>/s) for (a) West and (b) East rivers.

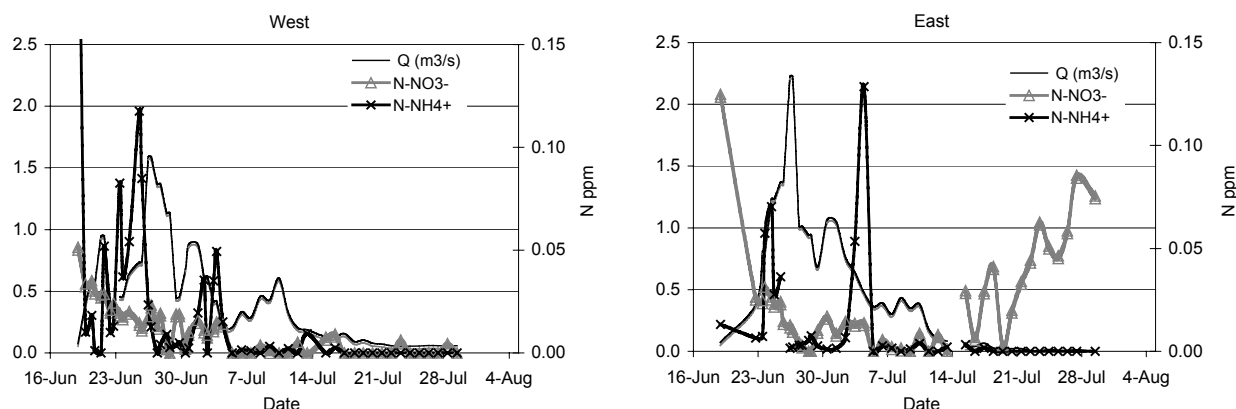


Figure 4. Seasonal variations in concentrations of N from  $\text{NO}_3^-$  and N from  $\text{NH}_4^+$  plotted with mean daily discharge ( $\text{m}^3/\text{s}$ ) for (a) West and (b) East rivers.

### 3.3 Controls on N species and seasonal variations in concentrations

Water flux exerts first order control over the export of solutes from the terrestrial system. However, as nitrogen is a limiting nutrient in most ecosystems, nitrogen losses in stream waters will only occur if the nitrogen cannot be utilized or controlled by the biota in the terrestrial system (Perakis, S. 2002). In principle this means that N can be lost in forms that are unavailable to organisms, such as complex or large organic molecules, or else if there is a spatial or temporal disconnect between the hydrological supply and biological demand of bioavailable forms of N (such as  $\text{NO}_3^-$  and  $\text{NH}_4^+$ ) (Perakis, S. 2002, Pinay, G., *et al.* 2002)

The ultimate source of DON is from soluble or leachable soil organic matter, as DON yields in stream water are usually closely related to the composition of soil organic matter and the flow of water through the shallow soil horizons (Cooper, R., *et al.* 2007, Hood, E., *et al.* 2003a). The sinks for DON are primarily mineralization reactions (conversion of DON to  $\text{NH}_4^+$  by ammonification, and to  $\text{NO}_2^-$  and  $\text{NO}_3^-$  via nitrification) and percolation through mineral soils which promotes the retention of DON via sorption reactions and removal by biotic uptake (Hagedorn, F., *et al.* 2000, Perakis, S. 2002).

The differences in the composition of the N species, and the substantial differences in the relationship between N species and other solutes in the two streams suggest that N losses are controlled by different factors in these two catchments. The West catchment chemistry suggest that most of the N export from this catchment (DON,  $\text{NH}_4^+$  and  $\text{NO}_3^-$ ) is related to DOC. Therefore, it appears that the N exports are related to the flushing of the shallow soil horizons, which provide a source of leachable DOM and also  $\text{NH}_4^+$ , and  $\text{NO}_3^-$  that may have built up in soils as a result of mineralization (decomposition) over the course of the previous summer. Note that the DOC and DON concentrations remain stable while the  $\text{NH}_4^+$  and  $\text{NO}_3^-$  concentrations drop to zero for most of the summer (after July 5<sup>th</sup>). This suggests that although there is still some hydrological connection between the West river and the organic soil horizons in the catchment, either there is limited production of inorganic N (ammonification and nitrification) in these soil horizons, or that any  $\text{NH}_4^+$  or  $\text{NO}_3^-$  that was available at this time (the start of the growing season) was immobilized by catchment biota. The East river displays a very different scenario of control on N export. Here there was only a weak correlation between DOC and DON concentrations, and the dominant inorganic N species ( $\text{NO}_3^-$ ) co-varied with solutes derived from weathering, and atmospheric deposition (e.g.  $\text{K}^+$ ,  $\text{Na}^+$  and  $\text{Cl}^-$ ). These results suggest that  $\text{NO}_3^-$  may be largely derived from mineral soil horizons in this catchment. The increase in the DON and  $\text{NO}_3^-$  concentrations at the end of the summer in the East river, suggest that there was a spatial disconnect between the source of nutrients and areas where there was a biological demand for them (Figure 4). The decrease in the DOC:DON ratio (from ~15-6) and the increase in DON concentrations indicate that there was a change in the dominant source of DOM in the East River over the course of the course of the summer (Figure 5). The higher N content of the DOM likely indicates a more labile DOM, possibly in the form of fulvic acids produced by microbial activity

(McKnight, D. M., *et al.* 1994). Note that in contrast the West river, DOC:DON ratios remain relatively high during recession of flow (Figure 5)

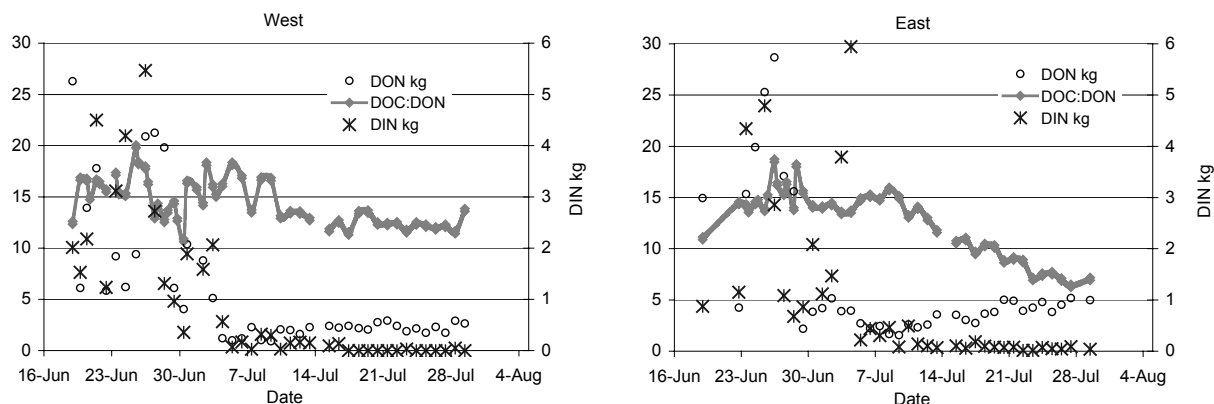


Figure 5. Seasonal DOC:DON ratios, DIN and DON mass fluxes (kg) for (a) West and (b) East rivers.

### 3.4 N structure and losses relative to other environments

Despite differences in the seasonal patterns of nutrient concentrations in the river, the two catchments have nearly identical seasonal DON, DIN and DOC yields (Table 2). These yields were on the order of 250 kg DON, 45 kg DIN and 3800 kg DOC. When converted to fluxes on a catchment area basis these mass fluxes convert to 475 kg C/km<sup>2</sup>, 31 kg N-DON/km<sup>2</sup> and 5.6 kg N-DIN/km<sup>2</sup> for the West and 328 kg C/km<sup>2</sup>, 21 kg N-DON/km<sup>2</sup> and 3.9 kg N-DIN/km<sup>2</sup> from the East catchment. The results show that DON is by far the most important vector for N losses from these High Arctic catchments (DON > 80% of total N) (Table 2). The mean concentrations of DON in these High Arctic streams (0.180 to 0.200 ppm), are generally higher than those reported for studies in the Colorado Rocky Mountains (0.023-0.084) and the total DON fluxes from the Cape Bounty catchments (20-30 kg/km<sup>2</sup>) are within the range reported for these alpine and subalpine catchments (18-60) (Hood, E., *et al.* 2003b, Williams, M. W., *et al.* 2001). However, the export of inorganic N from these High Arctic streams is 2 orders of magnitude lower than DIN export in the catchments in the Colorado Front Ranges (Hood, E., *et al.* 2003b, Williams, M. W., *et al.* 2001). The very high DIN exports in these catchments are largely attributed to the high rates of atmospheric N deposition in these mountain ranges. When compared to unpolluted headwater catchments in temperate South American forests, we find that the concentrations of both inorganic and organic N in runoff are higher than in these forested watersheds (Perakis, S. S. and Hedin, L. O. 2002). The mean concentrations for DON, N-NO<sub>3</sub><sup>-</sup>, and N-NH<sub>4</sub><sup>+</sup> for the East and West rivers combines are 192 ppb N, 17 ppb, and 15 ppb, respectively. The mean concentrations for the 13 South American study areas were 58.6 ppb DON, 1.9 ppb N-NO<sub>3</sub><sup>-</sup>, and 4.9 ppb N-NH<sub>4</sub><sup>+</sup> (Perakis, S. S. and Hedin, L. O. 2002). Similar to the Arctic streams where DON accounted for >80% of total N load in streams. DON represented >90% of the total N exported in streams from the unpolluted South American catchments. However, due to the high rates of precipitation (500-6000 mm/yr) in these South American locations, the annual net flux of DON from these areas are estimated for be between 20-350 kg DON/km<sup>2</sup> /yr (Perakis, S. S. and Hedin, L. O. 2002). The author is unaware of any studies of DON and DIN species concentrations and export from Arctic rivers with which to compare these results.

## 4. CONCLUSIONS

The study finds that although the West and East catchments yield the same seasonal mass of DON and DIN, the controls on DON and DIN in the two watersheds are clearly different. While the West catchment appears to have a consistent source of dissolved organic matter (as indicated by the relatively constant

DOC:DON ratios), there appears to be a gradual change in the composition of the DOM in the East river as flows recede in the summer. The drop in flux of inorganic nitrogen species to near zero values in the West river after the first week in July, suggests efficient retention (biological uptake) of inorganic N in this catchment. In contrast, the rise in the concentrations and fluxes of NO<sub>3</sub><sup>-</sup> and DON in the East river over the month of July, suggests biological demand for nitrogen is lower, or spatially disconnected with the N sources in this watershed. Therefore, although the controls on N losses vary across the landscape, the two watersheds likely have similar constraints on the upper limits of total N yields.

DON represents more than 80% of the total mass of N exported from these two arctic catchments, suggesting that DON may be a key source of N for ecosystems downstream. The fact that the mass flux of this DON (and other DIN) species occur long before the beginning of the growing season, suggests that the delivery of these nutrients may be well in advance of the peak of biological demand in aquatic ecosystems downstream. The total mass fluxes of DIN and DON from these small Arctic catchments are not trivial when compared to catchments in more temperate environments. The results of this study seem to suggest that N should not be a limiting factor for aquatic biological productivity, but the timing and composition of the nitrogen being delivered may be the more important constraints on productivity in these ecosystems. There is a need to understand the extent to which the magnitude, composition and timing of the N fluxes from Arctic systems vary with changes in hydrology and climate in order to support the conclusions drawn from this preliminary study.

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