Hydrological and land use control on N export sensitivity to climate in three adjacent watersheds

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Abstract
Although fluctuation in air temperature can cause changes in snowpack depth in winter and stream discharge in summer, the response of N export to the climate is poorly understood. We investigated the response of N concentrations to hydrological process and climatic conditions in three adjacent watersheds with different land use. Hydrological events contributed to 58-64% of the annual discharge and 62-78% of the annual TN loading in three headwater streams, and wet years tended to trigger larger discharge and N export. Stepwise multiple regression revealed that stream discharge was the best predictor of N concentrations, but variability in stream N concentrations also corresponded to changes in air temperature and snowpack depth throughout the winter, and were sensitive to precipitation in the summer. The agricultural watershed contributed more N loadings and higher concentrations than the forested watershed, but the mole ratios of Si to TN were much lower in the agricultural watershed and most of them were below the threshold value (2.7) for eutrophication during hydrological events, posing a high threat to coastal water.

Key Words
Nitrogen export; stream discharge; climate; land use; watershed.

Introduction
Nitrogen (N) export from a watershed at temporal and spatial scales has been widely reported and an understanding of N export is improved, for example, the well-known hydrological flushing of N, especially NO_3^--N, during the snowmelt season and storm events (Creed and Band 1998; Zhang *et al*. 2007). Long-term studies on Adirondack forested watershed showed that temperature fluctuation in winter played a key role in interannual variation in the export of NO_3^--N (Park *et al.* 2003) and DOC (Park *et al.* 2005). Therefore, an interaction between temperature and streamflow might be an important regulator for the export of N. However, the impact of climatic conditions responsible for affecting the snowpack depth and streamflow is usually neglected and poorly understood.

The Shibetsu area in northern Japan has a hemi-boreal climate, characterized by warm summers and cold winters with a substantial portion of snow-covered period. Our previous studies found that there was a significant positive correlation between NO_3^--N concentrations and the proportion of upland (Hayakawa *et al.* 2006) and that N export was controlled by hydrologic processes during storm events (Jiang *et al.* 2009, under review). Therefore, we investigated the response of N concentrations to climatic variation in three watersheds with different land use to further understanding the coupling impact of climatic variation and hydrological processes on N export at a spatial scale.

Methods
Watershed description
Three headwater stream watersheds adjacent to each other (agriculture-dominated watershed, AW; forest-dominated watershed, FW; and mixed agriculture-forested watershed, AFW) were selected for this study, which are located in the Shibetsu watershed in eastern of Hokkaido, Japan. The characteristics of watersheds are given in Table 1.

Watershed monitoring, sampling, and analysis
Base flow water samples were grabbed from the stream outlet in 1 L polypropylene bottles once a month from March to November during 2003-2005. Automated water samplers (ISCO ™ 3700) were installed at the outlet of watersheds, and water samples were collected during storm events and snowmelt season. Daily stream water level at every 15 min was recorded using a water sensor and a logger. Meteorological data was obtained form Japan Meteorological Agency (http://www.jma.go.jp). Chemical analysis including total N

(TN), total dissolved N (TDN), NO_3^--N, NH_4^+-N, DOC, SO_4^{2-}, Si, and basic cations were analyzed at the laboratory following the standard methods.

Table 1. Location, watershed characteristics, and discharge statistics.

Watershed	Site coordinates		Area km^2	Land use %		Discharge (2003-2005) m^3/s		
				Agriculture	Forest	Mean	Maximum	Minimum
AW	43.544N	144.865E	9.3	90.3	9.3	0.24	2.58	0.11
AFW	43.525N	144.833E	28.3	52.2	45.9	0.99	50.40	0.11
FW	43.540N	144.762E	76.0	14.7	85.1	2.63	23.34	0.84

Results

Climatic and hydrologic conditions

Stream discharge of the three watersheds all peaked sharply during storm events, and quickly responded to the increase in mean daily air temperature at the beginning of the snowmelt season (Figure is not shown). All stream discharge values showed a strong positive correlation with precipitation, and an inverse correlation with snowfall and snowpack depth in a whole period.

Nitrogen concentration

A significant difference in discharge and concentrations of N species was found among years and watersheds. The largest discharge and N concentrations were observed in wet year (2003). The forested watershed FW had the largest discharge with the lowest N concentration. By contrast, the agricultural watershed AW had the lowest discharge with the largest N concentration. NO_3^--N was the dominant N species for all the watersheds.

Response of N concentrations to climatic condition and discharge

N concentrations in headwater streams generally showed a synchronized response to runoff and precipitation during the study period. In winter months, N concentrations displayed a similar trend with the fluctuations of temperature, increasing significantly on the several consecutive days with above-freezing temperatures and increasing runoff at the beginning of snowmelt events (Figure 1, only watershed AW is shown, the other two are similar). To better understand the relationship, we separated the results for winter and summer. A positive correlation of N concentrations with runoff and temperature, and a negative correlation with snowpack depth were found in each watershed during the winter (table is not shown). Stepwise multiple regression selected discharge, temperature, and snowpack depth as significant predictors of N concentrations in each watershed (Table 2). In the summer, N concentrations showed a positive correlation with discharge in all watersheds, and a positive correlation with precipitation was found in watershed AW and AFW, but positive correlation with temperature for watershed FW (table is not shown). Overall, discharge was found to be the best predictor of N concentrations (Table 2).

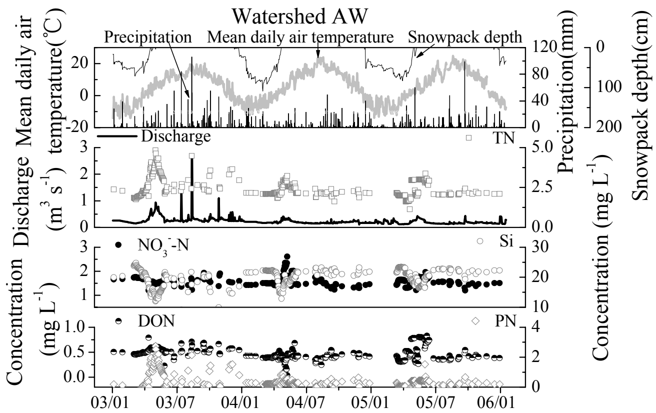

Figure 1. Response of N species concentrations to discharge and climatic conditions.

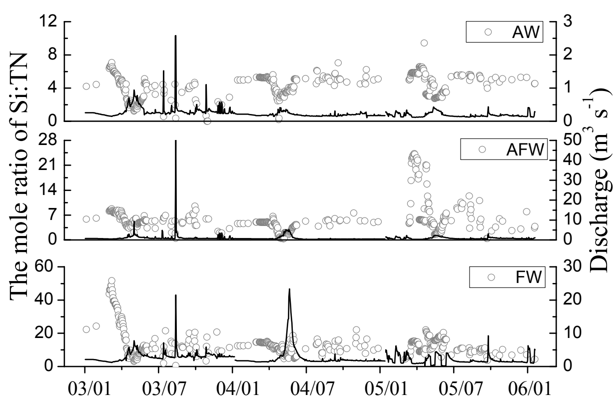

Figure 2. The mole ratio of Si to TN in three headwater stream watersheds.

Table 2. Results of stepwise multiple regression between N concentrations and climatic and hydrological variables. Asterisks indicate significant difference (*p<0.001; **p< 0.01; *p<0.05). Q: discharge; SD: snowpack depth; T: mean daily air temperature; P: precipitation; SLD: sunlight duration.**

		Winter	Summer
AW	TN	Y=1.650+3.23Q***-0.002SD***+0.007T***	Y=1.636+2.906Q***
	TDN	Y=2.197-0.003SD***+0.003T***	Y=1.984+0.363Q***-0.004P***-0.004SLD*-0.002T*
	NO_3^--N	Y=1.786-0.003SD***-0.364Q***	Y=1.541-0.003P***+0.11Q***
	NH_4^+-N	Y=0.027+0.001T***-0.037Q***	Y=0.01-0.00005T*
	DON	Y=0.417+0.319Q***-0.0008SD***	Y=0.417+0.23Q***-0.003T***
	PN	Y=-0.58932+3.384Q***+0.001SD**	Y=-0.317+2.691Q***
AFW	TN	Y=1.314+1.498Q***-0.010SD*	Y=1.439+0.041P***+0.053Q**+0.028SLD*
	TDN	Y=1.210+0.193Q***-0.012**	Y=1.289+0.016P***+0.013SLD*-0.022Q*
	NO_3^--N	Y=0.982+0.075Q***-0.008T***	Y=1.079-0.018Q***
	NH_4^+-N	Y=0.025+0.001T***	-
	DON	Y=0.196+0.124Q***-0.006T*	Y=0.219+0.015P***+0.01SLD*
	PN	Y=0.093+1.327Q***-0.011SD*	Y=0.151+0.025P***+0.075Q***+0.015SLD*
FW	TN	Y=0.165+0.129Q***+0.004SD***-0.006T*	Y=0.23+0.144Q***+0.01T***
	TDN	Y=0.212+0.055Q***+0.003SD***-0.004T*	Y=0.364+0.022Q***+0.004T***
	NO_3^--N	Y=0.084+0.002SD+0.045Q-0.006T	Y=0.332-0.02Q***+0.004T***
	NH_4^+-N	Y=0.023+0.001T***-0.001Q***	Y=0.019-0.002Q***
	DON	Y=0.125+0.008Q**	Y=0.014+0.044Q***
	PN	Y=-0.044+0.072Q***+0.001SD**	Y=-0.134+0.123Q***+0.006T**

Nitrogen loading

The annual discharge and N loading were much larger in 2003 than that in 2004 and 2005 for all watersheds (Figure is not shown). The forested watershed FW showed the lowest N export. Hydrological events contributed the most of annual loading, of which TN accounted for 62, 78, and 62% of annual loading with 58, 64, and 62% of annual discharge in watersheds AW, AFW, and FW, respectively.

Implication of N export to surface water

A strong positive correlation between NO_3^--N and SO_4^{2-} concentrations was found for all watersheds and all periods (table is not shown), indicating that higher N export during hydrologic events also led to higher SO_4^{2-} export, which might enhance the potential for acidification and pose a high threat to coastal water.

The mole ratio of Si:TN in river water is a useful predictor for eutrophication, and the value below 2.7 is considered as the threshold. Our results showed that the mole ratios of Si:TN were below 2.7 during the snowmelt season and rain events in watersheds with high agricultural proportion (AW and AFW), posing a potential hazard of entrophication to water bodies (Figure 2).

Conclusion

Hydrological events had control over N export in the three headwater streams, and wet year tended to trigger larger discharge and N export. Discharge was the best predictor of N concentrations; however, stream N concentrations were also sensitive to temperature, precipitation, and snowpack depth. Agricultural watershed exhibited remarkably high N loadings to streams and higher concentrations than that of forested watershed, and the mole ratio of Si to TN was below the threshold value for eutrophication during hydrological events, posing a high threat to coastal water.

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