Groundwater Research Report
WR10R005

GROUNDWATER NITRATE PROCESSING IN DEEP STREAM SEDIMENTS

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WR10R005: Groundwater nitrate processing in deep stream sediments

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PROJECT SUMMARY

Title: Groundwater nitrate processing in deep stream sediments
Project I.D. WR10R005

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Background/need: Elevated nitrate concentration in ground water is a pressing environmental problem in many regions of the world, including Wisconsin (Browne et al. 2008, Rupert 2008, Saad 2008). The nitrate concentration of ground water in many areas of the Central Sand Ridges Ecoregion of Wisconsin exceeds the recommended limit for drinking water (10 mg NO_3-N L^{-1}) set by the Environmental Protection Agency. Current federal policy mandating the use of biofuels (e.g. ethanol produced from corn) and world demands for food may lead to further increases in nitrate concentrations and loads in groundwater. Identification of hot spots of nitrogen processing will improve the ability of scientists to predict nitrogen retention and loss from watersheds and will aid land and water managers who need to make decisions that balance nitrogen removal with needs of other stakeholders. The proposed project addresses the following priorities of the University of Wisconsin System: Interactions of groundwater and surface water including chemical transformations in the hyporheic zone.

Objectives: The main objectives of the proposed project were: 1) To determine if nitrogen processing in groundwater associated with deep stream sediments is widespread throughout a river network, 2) To determine if high nitrate concentration in groundwater saturates denitrification in stream sediments.

Methods: We identified eight study sites on streams and rivers in the Waupaca River Network in Central Wisconsin. Sites were chosen that spanned a large range in groundwater nitrate concentration (<0.01 to 9 mg NO_3-N/L on average), were located in upwelling reaches, and had fine sediments present. We measured denitrification on sections at 5 cm intervals from four to five sediment cores (to a depth of 20 to 30 cm) collected from each stream to determine how denitrification rates vary by depth, among cores, and among streams. The organic matter content of the sediment cores, as well as the nutrient and dissolved oxygen concentrations of the groundwater used in the denitrification incubations, will be used to develop regression models for predicting denitrification rates in stream sediments. Three peeper samplers and piezometer nests were deployed in each stream to determine fine-scale vertical profiles of nitrate and chloride concentration in the groundwater to a sediment depth of 90 cm. Our combined approach (denitrification measurements and nitrate profiles) has resulted in some of the most high-resolution estimates of groundwater nitrate processing in stream sediments.

Results and Discussion: Mean denitrification rates were higher in shallow sediments than deeper sediments. However, core sections deeper than 5 cm accounted for about 70%, on average, of the total denitrification (integrated throughout the entire core). The magnitude of denitrification rate differed strongly among sites. At many sites denitrification rates were higher in shallower sediments, while other locations showed similar denitrification rates at various sediment depths or higher denitrification rate in deeper sections. Denitrification rate increased linearly with groundwater nitrate concentration at low concentrations (< 2 mg NO_3-N/L) but denitrification varied considerably at high groundwater nitrate concentrations (> 5 mg NO_3-N/L), a pattern that suggests nitrate saturation.

For most of the study sites nitrate concentration was higher in deep groundwater than in shallower groundwater. At most sites including the Tomorrow River Site I, Bear Cr., Emmons Cr. and the Crystal River nitrate concentration tended to decline to very low concentrations as groundwater moved from deeper to shallower sediments, while chloride concentration changed much less. Two piezometer nest locations showed that groundwater nitrate remained high as water moved from deeper to shallower sediments. At two nest locations at Tomorrow River Site II chloride and nitrate concentrations were both higher in the deep groundwater than in the shallow groundwater. Finally, all piezometer nest locations at Hartman Cr. and the Waupaca R. revealed nitrate concentrations at or below the detection limit for both deep and shallow groundwater. The ratio of NO_3-N:Cl^- was lower in shallow groundwater than in deep groundwater at 14 of 18 of the locations in which the nitrate concentration in the deep groundwater was above the detection limit. This result suggests that nitrate was removed in most cases as groundwater upwelled from deep to shallower sediments.

Conclusions/Implications/Recommendations: The denitrification results and nitrate profile results both suggest that nitrate removal from groundwater is widespread in deep sediments of streams and rivers in the Waupaca River Network. Our results suggest that estimates of nitrogen processing based exclusively on shallow sediment cores or on whole-stream injections of nitrate may underestimate stream ecosystem N-removal by not capturing nitrogen processing that occurs in deep sediments. We think that processes in deep sediments will need to be considered when modeling nitrate removal at the network and watershed scales. Failing to account for nitrate removal in deep sediments could lead to errors when closing nitrogen budgets at these scales. Our results also emphasize the importance of healthy intact sediments for groundwater nitrate removal in nitrate-contaminated stream ecosystems.

Related Publications: none currently (a manuscript is in preparation)

Key words: nitrate, groundwater, denitrification, sediments, streams, sand plains, biogeochemistry, river network, scale

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PROJECT COMPLETION REPORT for WR10R005: Groundwater nitrate processing in deep stream sediments

Introduction-

Humans have dramatically altered the nitrogen cycle during the past several decades, doubling the amount of fixed nitrogen worldwide (Galloway et al. 2008, Schlesinger 2009). Global increases in fertilizer production and application and increases in nitrogen oxide generated by burning fossil fuels are major causes for increases in the amount of available nitrogen in ecosystems. These changes have resulted in increases in the concentration and fluxes of available nitrogen in rivers (Howarth et al. 1996, Donner et al. 2002) and increases in the concentrations of available nitrogen in groundwater in many parts of the world, including Wisconsin (Browne et al. 2008, Rupert 2008, Saad 2008). Elevated nitrate in groundwater has implications for human health (Kross et al. 1992) and contributes to nitrogen loading in river and lakes where groundwater discharges to surface water. When available nitrate reaches high levels, the ability for ecosystems to process this nitrogen can become saturated (Aber et al. 1997, O'Brien et al. 2007). For example, Mulholland et al. (2008) showed that stream water nitrate concentration saturated denitrification in streams at the continental scale. It is less clear if elevated nitrate concentration in groundwater saturates nitrate retention and removal mechanisms in stream sediments.

Because the supplies of available nitrogen to ecosystems have been increasing and are projected to continue to increase, there is growing interest in processes that can retain or remove available nitrogen in streams and rivers (Alexander et al. 2000, Mulholland et al. 2008). Processes contributing to nitrate retention in streams include assimilatory uptake by autotrophs and by heterotrophic microbes (e.g. Stelzer et al. 2003) and dissimilatory uptake, including denitrification, by microbes (Burgin and Hamilton 2007). Denitrification has been shown to be influenced by nitrate concentration, carbon supply, and oxygen status (Arango et al. 2007, Groffman et al. 2009). It is well known that processes in riparian zones (e.g. Hedin et al. 1998), in hyporheic zones (where groundwater and surface water mix) (Hill and Lymburner 1998) and in the surface water of streams and rivers (Mulholland et al. 2008) can retain and remove substantial amounts of available nitrogen. Much less is known about the role of deep sediments beneath the stream channel (below the hyporheic zone) in nitrogen processing. Many studies of nitrogen processing in streams do not include deep sediments. For example, most studies of denitrification in streams only include denitrification measurements from surficial sediments (cores less than 5 cm deep) (e.g. Arango et al. 2007, Herrman et al. 2008). In groundwater-fed streams groundwater typically passes through substantial quantities of sediment before discharging to the stream. Previous studies have suggested that available nitrogen is retained along upwelling flow paths in deep sediments (Duff et al. 2008, Puckett et al. 2008, Stelzer et al. 2011). However, most previous studies have not included process-oriented measurements in deep sediments (but see Fischer et al. 2005, Inwood et al. 2007) or have not included the fine-scale vertical profiles of available nitrogen necessary to infer where nitrogen retention occurs in deep sediments. We have reported fine scale changes in nitrate and chloride concentration from a single stream in the Waupaca River Network (in the Central Sand Ridges Ecoregion of Wisconsin) that suggests nitrate processing can be substantial in deep sediments associated with streams (Stelzer et al. 2011). In the current study, we determined the applicability of these

findings to a network (the Waupaca River Network) of streams and rivers spanning a 100-fold range in groundwater nitrate concentration. We addressed the following questions:
1. Is nitrogen processing in groundwater associated with deep stream sediments widespread throughout a river network?
2. Does high nitrate concentration in groundwater saturate denitrification in stream sediments?

Procedures and Methods-
We identified eight study sites on streams and rivers in the Waupaca River Network (Fig. 1). Sites spanned a large range (100-fold) in groundwater nitrate concentration, were in upwelling reaches, and had fine sediments (silt, sand) present. The study took place during late spring through early fall of 2010. We addressed our research questions by completing the following tasks: 1) We measured denitrification rates on sectioned sediment cores to determine if denitrification rate varied with sediment depth, 2) We determined if denitrification rates saturates at high groundwater nitrate concentrations, and 3) We measured fine-scale variation in groundwater nitrate and chloride concentrations using both peepers and piezometer nests.

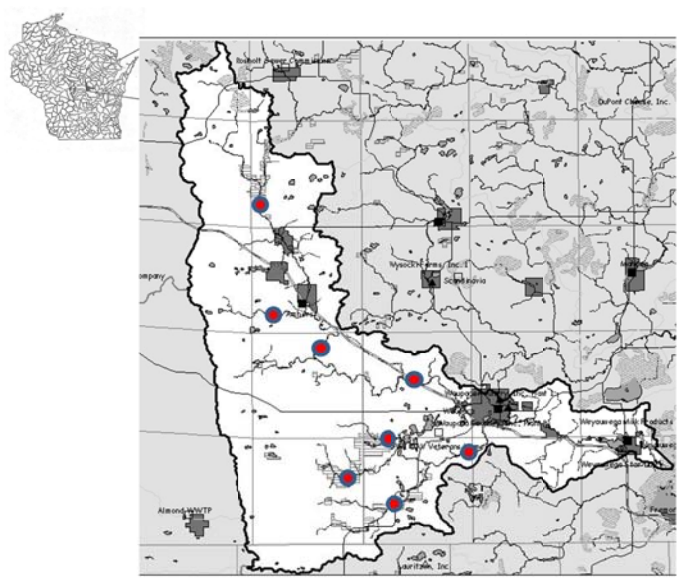
Fig. 1. Map of Waupaca River Watershed with sampling sites indicated (modified from map by the Wisconsin Department of Natural Resources)

Denitrification rate measurements-
Four to five sediment cores (7.6 cm diameter, 20 to 30 cm length) were collected in upwelling locations (as determined by measurements of vertical hydraulic gradient) at each site. Each core was divided into 5-cm sections and placed in Whirl-Pak bags for transport to the Aquatic Ecology Laboratory at UW Oshkosh. Within 24 to 48 hrs denitrification rates were measured using the chloramphenicol-amended (1 mg ml^{-1}) acetylene block method (Richardson et al. 2004, Groffman et al. 2006) in the laboratory. Incubations (and core storage prior to incubation) were carried out in a Fisher Isotemp Model 307C incubator set to the ambient temperature of groundwater at the time of sediment core collection. Groundwater was pumped from piezometers adjacent to the location of each sediment core (see below) and was added to the sediments prior to the incubations. Denitrification rates were calculated as the rate of nitrous oxide (N_2O) production during 90 minute incubations. Subsamples of sediments from each core section will be analyzed for organic matter content and bulk density using standard methods. Samples from groundwater used in the incubations were analyzed for nitrate and dissolved organic carbon. Sediment and groundwater parameters will be used in multiple regression models to determine the drivers of denitrification in deep stream sediments in the Waupaca River Network. Nested ANOVA was used to compare denitrification rates among sites and among core sections (core section was nested within site in the models).

Groundwater Nitrate Profiles- Piezometers in groups of six (nests) were installed at three upwelling locations at each site. Piezometers were constructed of CPVC (1.2 cm inner diameter) with the terminal 4.5 cm screened (3 mm holes covered with 100 µm Nitex mesh). Modified Pore Water Hesslein Samplers (peepers) were deployed within each piezometer nest. The piezometers were installed at different depths within each nest so that the nitrate and chloride concentrations in relatively deep groundwater (35 to 90 cm) could be characterized while the peepers provided nutrient concentrations at 1.3 cm vertical intervals in the 1 to 25 cm range. Together, water samples collected from the piezometers and peepers provided a high-resolution profile of nitrate and chloride in the sediments to about 70-90 cm. Groundwater nitrate and chloride concentrations were used to calculate NO_3-N:Cl^- ratios. Unpaired t-tests were used to compare the NO_3-N:Cl^- ratios of deep groundwater (from the 6 piezometers) to those in shallower groundwater (from the 6 deepest peeper samples) for each piezometer nest-peeper complex. We predicted that the NO_3-N:Cl^- ratio would be higher in deep groundwater than in shallow groundwater if nitrate removal was occurring in the deep sediments.

Results and Discussion-

Mean denitrification rates were higher in shallow sediments than in deeper sediments (Table 1, ANOVA P <0.01). However, core sections deeper than 5 cm accounted for about 70%, on average, of the total denitrification (integrated throughout the entire core). The magnitude of denitrification rate differed strongly among sites (ANOVA P <0.01, Fig. 2). At many sites (Fig. 2a, b, e, f) denitrification rate was higher in shallower sediments, while other locations showed similar denitrification rates at various sediment depths (Fig. 2c) or higher denitrification rate in deeper sections (Fig. 2d, g). Denitrification rates tended to be much higher on average at locations with high concentrations of groundwater nitrate such as Bear Cr., Tomorrow River Site II, and Radley Creek (Fig. 2). Denitrification rate increased linearly with groundwater nitrate concentration at low concentrations (< 2 mg NO_3-N/L) but denitrification varied considerably at high groundwater nitrate concentrations (> 5 mg NO_3-N/L), a pattern that suggests nitrate saturation (Fig. 3).

Table 1. Denitrification rates (mean, SD, N) by core section for the sites in the Waupaca River Network.

Core Section (cm)	Mean (ug N2O-N/cm2/h)	SD	N
0-5	2.04	2.78	33
5-10	1.98	3.68	33
10-15	0.94	1.81	33
15-20	0.68	1.39	33
20-25	0.71	2.08	30
25-30	0.80	1.97	17

For most of the study sites nitrate concentration was higher in deep groundwater (sampled with the piezometers) than in shallower groundwater (sampled with the peepers) (Fig. 4). At most sites including the Tomorrow River Site I, Bear Cr., Emmons Cr. and the Crystal River nitrate concentration tended to decline to very low concentrations (at or below the detection limit) as groundwater moved from deeper to shallower sediments, while chloride concentration changed much less (e.g. Fig 4a,c,e,g,l). Two piezometer nest locations showed that groundwater nitrate remained high as water moved from deeper to shallower sediments (Emmons Cr.-Fig. 4h, Radley Cr.-Fig. 4r). At two nest locations at Tomorrow River Site II (Fig. 4m,n) chloride and nitrate concentrations were both higher in the deep groundwater than in the shallow groundwater. This may reflect that different groundwater flow paths were sampled by the peepers and piezometers at these locations or that chloride did not behave conservatively. Finally, all piezometer nest locations at Hartman Cr. and the Waupaca R. revealed nitrate concentrations at or below the detection limit for both deep and shallow groundwater. The ratio of NO_3-N:Cl^- was lower in shallow groundwater than in deep groundwater at 14 of 18 of the locations in which the nitrate concentration in the deep groundwater was above the detection limit (one-tailed t-tests, $P<0.05$). This result suggests that nitrate was removed in most cases as groundwater upwelled from deep to shallower sediments.

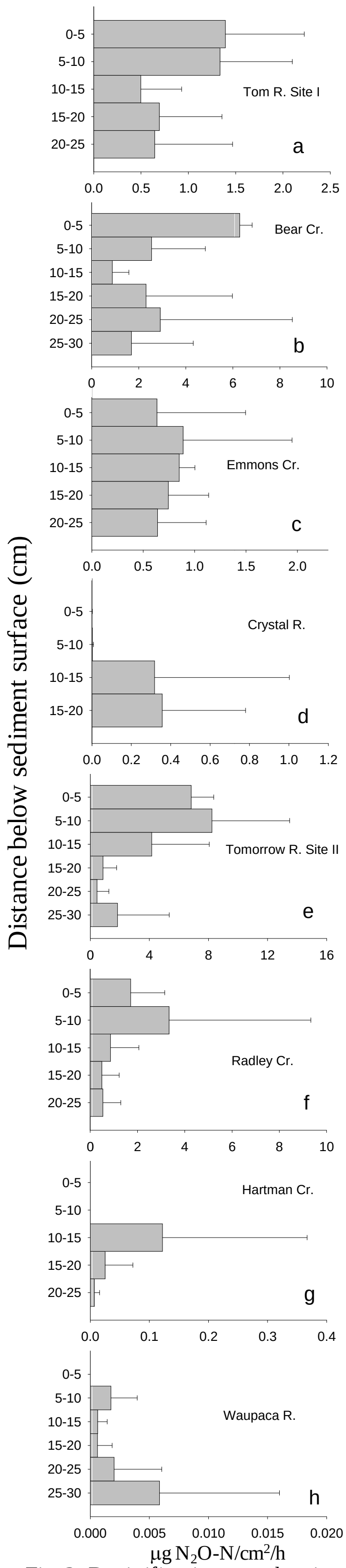

Fig. 2. Denitrification rates by site and by core section in the Waupaca River Network

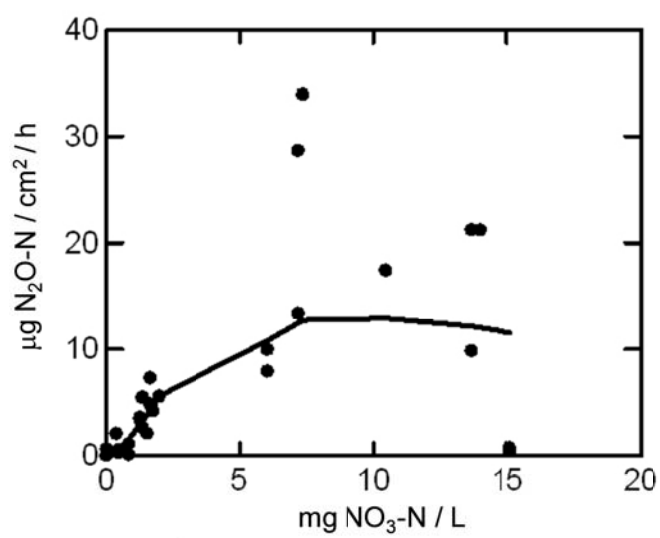

Fig. 3. Denitrification rates (integrated across core sections) plotted against groundwater nitrate concentrations associated with each core for sites in the Waupaca River Watershed.

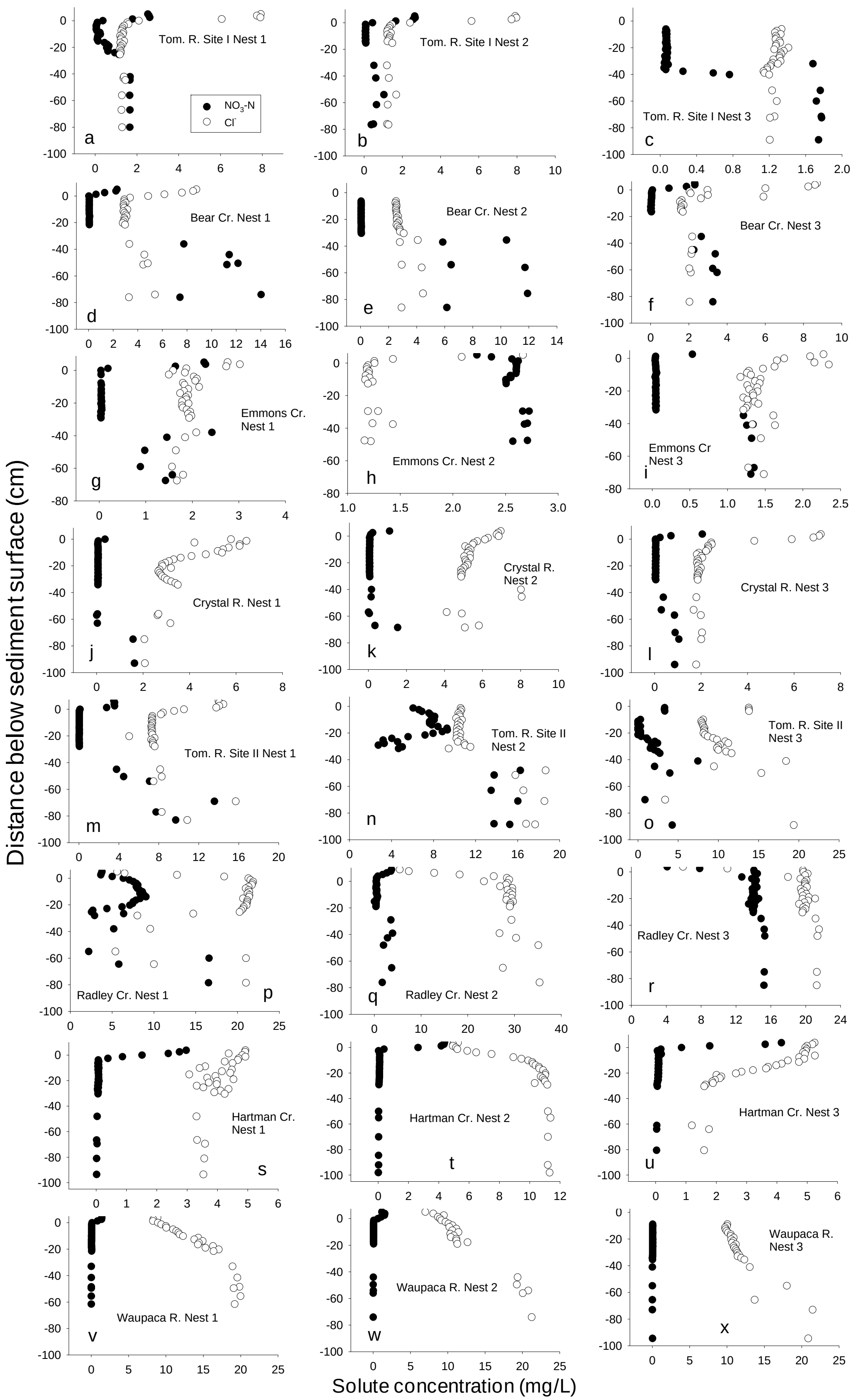

Fig. 4. Nitrate and chloride depth profiles for each piezometer nest at the 8 sites.

Conclusions and Recommendations-

The denitrification results and nitrate profile results both suggest that nitrate removal from groundwater is widespread in deep sediments of streams and rivers in the Waupaca River Network. Our results suggest that estimates of nitrogen processing based exclusively on shallow sediment cores or on whole-stream injections of nitrate may underestimate stream ecosystem N-removal by not capturing nitrogen processing that occurs in deep sediments. We think that processes in deep sediments will need to be considered when modeling nitrate removal at the network and watershed scales. Failing to account for nitrate removal in deep sediments could lead to errors when closing nitrogen budgets at these scales. Our results emphasize the importance of healthy intact sediments for groundwater nitrate removal in nitrate-contaminated stream ecosystems. If stream sediments become degraded because of toxin exposure or physical removal (e.g. dredging) ecosystem services they provide, such as nitrate removal, may be compromised.

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APPENDIX A:

Presentations at state and national conferences

Stelzer, R.S., L.A. Bartsch. 2011. Nitrate processing in deep sediments of a Central Wisconsin river network. Abstract of an oral presentation at North American Benthological Society Annual Meeting, Providence, RI.

Stelzer, R.S., L.A. Bartsch. 2011. Denitrification of groundwater nitrate in a Central Wisconsin river network. Abstract of an oral presentation at American Water Resources Association (Wisconsin Section) Annual Meeting, Appleton, WI.