



Isotopic characterization of nitrate sources and transformations in Lake Winnipeg and its contributing rivers, Manitoba, Canada

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ABSTRACT

Lake Winnipeg (Manitoba, Canada) is in a eutrophic state from a century of increased riverine loadings from agricultural and urban nitrogen (N) and phosphorus (P) sources. This study investigated seasonal patterns of the isotopic composition of nitrate (NO_3^-) in Lake Winnipeg and its contributing rivers to gain insight into current N nutrient sources and in-lake N dynamics. Elevated NO_3^- concentrations in Lake Winnipeg tributaries between 0.36 and 2.44 mg/L NO_3^- -N were associated with high $\delta^{15}\text{N}$ values between +5.0 and +13.9‰, while $\delta^{18}\text{O}_{\text{NO}_3}$ values were <+15.0‰. The three major riverine inputs had distinctive mean $\delta^{15}\text{N}_{\text{NO}_3}$ values of +8.1‰ for the Red River, -0.6‰ for the Winnipeg River, and +5.0‰ for the Saskatchewan River. The isotopic composition of NO_3^- in Lake Winnipeg was partly controlled by the isotopic composition of the riverine nitrate for instance via the predominant nitrate input to the South basin from the Red River. Nitrate assimilation and late season mineralization of phytoplankton and N_2 fixing cyanobacteria were identified as important additional processes affecting the isotopic composition of lake NO_3^- resulting in low $\delta^{15}\text{N}_{\text{NO}_3}$ values, especially in the North basin. In the South basin, elevated $\delta^{15}\text{N}_{\text{NO}_3}$ values in spring that changed to lower values by summer indicated a dynamic N cycle within the lake. Agreement between $\delta^{15}\text{N}$ values of lake NO_3^- , PON and fish suggests that dissolved nitrate partially affects the flow of nitrogen in the aquatic food webs of Lake Winnipeg.

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Introduction

Over the past 60 years human activity and agricultural industrialization with intensive fertilizer application have greatly affected the nitrogen (N) cycle of terrestrial and aquatic ecosystems (Vitousek et al., 1997). As a consequence of enhanced anthropogenic nitrogen loading, many key aquifers and water supplies, rivers, and estuaries around the world are negatively affected by increased dissolved inorganic nitrogen concentrations (Glibert et al., 2006; Goolsby et al., 2000; Mayer et al., 2002; Rupert, 2008). Consequences of excess dissolved inorganic N entering receiving waters include human health concerns, water quality degradation, eutrophication, red tides, and in some cases hypoxia of coastal oceans and inland lakes (Diaz and Rosenberg, 2008).

Lake Winnipeg, a dual-basin great lake in Western Canada (Fig. 1) is currently in a eutrophic state due to decadal-scale increases in

riverine loadings of agricultural and urban derived nitrogen (N) and phosphorus (P), primarily from the Red River and also from the Winnipeg River (Stewart et al., 2000; Yates et al., 2012). Extensive cyanobacterial blooms in Lake Winnipeg, particularly over the past decade (McCullough, 2009; McCullough et al., 2012), are generally attributed to this increased anthropogenic nutrient loading (Manitoba, 2008). Lake Winnipeg receives discharge and nutrients principally from three major riverine inputs; the Red and Winnipeg Rivers in the South basin, and the Saskatchewan River on the west side of the North basin (Fig. 1). Other potential nutrient sources are from smaller agriculturally affected rivers to the west (e.g. Dauphin and Icelandic Rivers), and the boreal Canadian Shield to the east although these are characterized by comparatively lower discharge and a lack of nutrient monitoring data. The movement and mixing of waters entering Lake Winnipeg follow counter-clockwise mixing gyres – one gyre in the south and two in north basin (Zhao et al., 2012). Mean water residence time is 4.4 years for the entire lake, 1.4 years for the South basin, and 3.6 years for the North basin (Zhang and Yerubandi, 2012; Zhao et al., 2012). Lake Winnipeg drains into Hudson Bay through the Nelson River outflow in the North basin (Fig. 1).

Long-term increases in nutrient loadings to the lake stemmed from the Red River and Winnipeg River, albeit with considerable temporal variability. A 58% increase in the total dissolved nitrogen (TDN) load from the Red River was reported for the period 1978–1999

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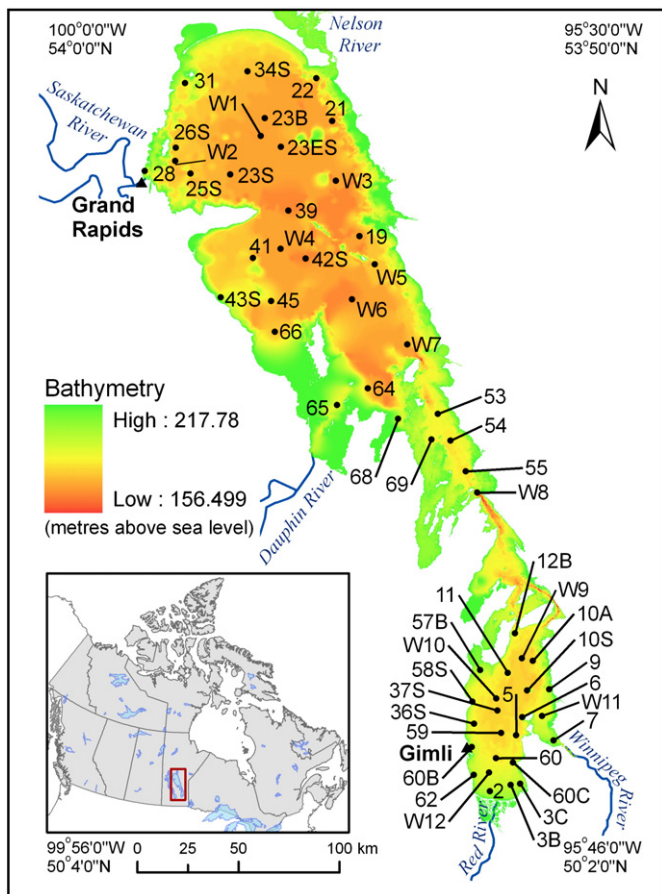


Fig. 1. Map of Lake Winnipeg, Manitoba, Canada, its bathymetry, riverine inputs and outflow, and names of official long-term water quality sampling stations.

(Jones and Armstrong, 2001; McCullough et al., 2012). TDN concentrations in the Red River from 1992 to 2007 were with on average 2.5 mg/L, markedly higher than those of the Winnipeg and Saskatchewan Rivers with 0.52 and 0.49 mg/L, respectively, with no discernible temporal trend. The TDN loading to Lake Winnipeg from all sources averages 90,701 t per year from 1994 to 2007, with 34% coming from the Red River, 25% from the Winnipeg River, and 10% from the Saskatchewan River. The remaining smaller and/or unmeasured rivers plus atmospheric N deposition are estimated to contribute ~30% of the TDN loading to Lake Winnipeg (Environment Canada, 2011). The annual and seasonal riverine nutrient loading to Lake Winnipeg is strongly dependent on the amount of discharge and nutrient levels in each of the contributing watersheds (McCullough, 2001; McCullough et al., 2012; Schindler et al., 2012). In the Red River, for example, the month of April contributes twice the N loading of all other months, owing to higher discharge from snowmelt and spring runoff in the Red River watershed. Other N sources to Lake Winnipeg are less well characterized, but could include nitrate delivered via smaller rivers, diffusive groundwater inputs, and decay and subsequent mineralization of N_2 fixing cyanobacteria (Environment Canada, 2011; Kling et al., 2011).

Monitoring of TDN within Lake Winnipeg by Manitoba Water Stewardship (MWS) revealed distinctive differences between the North and the South basins, averaging 0.65 mg TDN/L and 0.87 mg TDN/L, respectively, from 1999–2007 (Environment Canada, 2011). Similarly, average surface water nitrate (NO_3^- -N) concentrations in the South basin are correspondingly higher than those in the North basin (0.16 mg/L vs. 0.11 mg/L NO_3^- -N). Not surprisingly, highest concentrations of TDN are observed in the South basin near the mouth of the Red River. Seasonally, the TDN concentrations in Lake

Winnipeg increase towards the summer and fall, respectively, in part as a result of biological N_2 fixation by cyanobacteria in the summer months (Environment Canada, 2011).

Identifying the sources of dissolved NO_3^- -N and elucidating its biogeochemical cycling in riverine and lotic systems is challenging, but is crucial to gaining a better understanding of the sources and role of N in nutrient cycling in Lake Winnipeg. Stable isotope techniques are a promising tool for assessing sources of dissolved NO_3^- -N in surface water bodies, especially if the dual-isotope ($\delta^{15}N_{NO_3}$ and $\delta^{18}O_{NO_3}$) approach is used to characterize key sources and assess nutrient transformation processes (Aravena and Mayer, 2010; Kendall, 1998; Kendall et al., 2007; Mayer, 2005). Nitrate from animal manure and human sewage is characteristically typified by high $\delta^{15}N_{NO_3}$ values ranging from +7‰ to +20‰ (Aravena et al., 1993; Aravena and Robertson, 1998; Kreitler, 1979; Wassenaar, 1995). Animal and human waste derived N generally has much higher $\delta^{15}N_{NO_3}$ values than synthetic agricultural fertilizers (~0‰), nitrate generated via nitrification processes in soils, or from atmospheric nitrate deposition (-11 to +8‰) (Kendall, 1998; Kendall et al., 2007). Unfortunately, nitrate derived from ammonium fertilizers, soils, and atmospheric deposition cannot be clearly differentiated on the basis of $\delta^{15}N$ alone because of their overlapping isotopic values (Fig. 2).

Nitrate from atmospheric deposition, however, has very positive $\delta^{18}O_{NO_3}$ values ranging from >+50 to +94‰ (Durka et al., 1994; Kendall, 1998; Kendall et al., 2007; Voerkelius, 1990). Nitrate-containing synthetic fertilizers have $\delta^{18}O_{NO_3}$ values around $+22 \pm 3\%$ (Amberger, 1987; Voerkelius, 1990; Wassenaar, 1995). Nitrate derived from nitrification, for instance, in soils or resulting from mineralization of aquatic phytoplankton and N_2 fixing cyanobacteria followed by ammonification and nitrification has $\delta^{18}O_{NO_3}$ values between -15 and +15‰ (Durka et al., 1994; Hollocher, 1984; Mayer et al., 2001; Rock and Mayer, 2004) that are dependent on the $\delta^{18}O$ of water. Nitrates from manure and sewage have also $\delta^{18}O_{NO_3}$ values between -15 and +15‰ (Aravena et al., 1993; Wassenaar, 1995). Therefore, the combined use of $\delta^{15}N$ and $\delta^{18}O$ values of NO_3^- can provide a diagnostic tool for discerning among four major nitrate sources: (1) atmospheric nitrate deposition, (2) nitrate based fertilizers, (3) nitrate derived from nitrification in soils and aquatic systems (including decay of in-lake organic matter), and (4) nitrate from manure and sewage (Fig. 2).

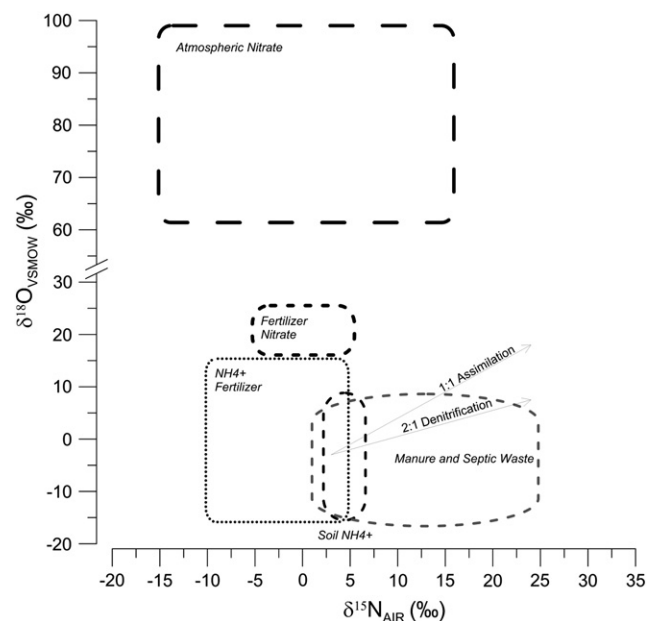


Fig. 2. Typical range of $\delta^{15}N$ and $\delta^{18}O$ values of nitrate derived from natural and anthropogenic N sources (adapted from Kendall et al., 2007).

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