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Noble gas enrichments in porewater of estuarine sediments and their effect on the estimation of net denitrification rates

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Abstract

The concentration and the isotopic ratios of noble gases He, Ne, Ar, Kr and Xe were measured in porewater trapped in shallow sediments of the estuary of the St-Lawrence River, Québec, Canada. The gases are atmospheric in origin but most samples have gas concentrations 1.7–28 times higher than those expected in solution in water at equilibrium with the atmosphere. Elemental fractionation of heavier noble gases Kr and Xe compared to Ar strongly suggests that noble gases were adsorbed on sediments or organic matter and then desorbed into porewaters due to depressurization, as collected samples were brought to the surface. Atmospheric Ar in porewater is used as a reference to measure the N_2-fluxes at the water–sediment interface. Ignoring the Ar enrichments observed in porewater could lead to a severe underestimation of the denitrification rate in oceans and estuaries.

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1. INTRODUCTION

The concentration of dissolved atmospheric noble gases (ANG) in groundwater (Air Saturated Water or ASW) and seawater (Air Saturated Seawater or ASSW) is determined by the physical conditions – air pressure, water/soil temperature and salinity – prevailing during gas exchange with the atmosphere (Mazor, 1972). Therefore, ANG can be used for paleoenvironmental and paleoclimatic reconstructions (Kipfer et al., 2002 and references therein). Recently, these applications have been extended to porewater preserved in sedimentary rocks (Osenbrück et al., 1998; Rübel et al., 2002) and in unconsolidated lacustrine and oceanic sediments (Brennwald et al., 2003, 2005; Strassmann et al., 2005). Vertical profiles of dissolved noble gas concentrations in lacustrine porewater are used for the quantitative reconstruction of past salinity and water levels in lakes (Brennwald et al., 2003). These profiles may be controlled by vertical diffusion/advection within the sediment column and thus serve as proxies to study the transport dynamics of solutes and fluids in the sediment (Strassmann et al., 2005; Chaduteau et al., 2009). Finally, noble gases can trace secondary gas exchange processes in the sediment. Brennwald et al. (2005) showed a distinct depletion of the ANG concentrations in lacustrine porewater of Soppensee, Switzerland. Poorly soluble, lighter noble gases escaped from the porewater into gas bubbles, due to ebullition of biologically produced methane. This phenomenon helped to quantify the release of potential greenhouse gases from lacustrine sediments during the Holocene (Brennwald et al., 2005).

Being inert, noble gas profiles can be compared against reactive species to quantify the processes of production/consumption and the input/output fluxes of those elements at the water–sediment interface. For example, the N_2/Ar ratio is measured in porewater to determine the net rate of denitrification (N_2 production from reduction of nitrates) in marine and estuarine settings, assuming the Ar concentration to be in solubility equilibrium with the atmosphere (Nielsen, 1992). Using a rigorous approach to the problem, the processes controlling the ANG concentration and distribution in sedimentary porewater need to be fully

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elucidated. For example, degassing in lacustrine sediments, as described by Brennwald et al. (2005), has to be taken into account to adjust the apparent N_2 losses for ebullition (calculated from the Ar losses) and to determine the amount of excess N_2 resulting from denitrification.

Here, we show that noble gases, Ne, Ar, Kr and Xe (and He in some samples), collected in porewater of the St-Lawrence estuary, Québec, Canada are enriched by a factor of 1.7–28 times the expected ASSW values. Physical adsorption of noble gases on sediments and/or organic matter and their successive desorption and release into porewater during the core recovery is likely the cause of these enrichments. The relative Ar enrichment, if not accounted for, would severely underestimate the net denitrification rate in estuaries and oceans.

2. SAMPLING AND ANALYTICAL METHODS

Seafloor sediments were collected in the St-Lawrence estuary, Québec, Canada (Fig. 1) during an oceanographic cruise in May 2007, onboard the research vessel Coriolis II. The primary goal was to test whether the noble gases measured close to the water–sediment interface could be used for calibrating C and N_2-fluxes in the organic-rich sediments of the estuary, these two elements being studied by research teams from McGill University, Concordia University, Université de Montréal and the GEOTOP research center at the Université du Québec à Montréal. Sediment cores, up to 36 cm long, were taken at stations 20, 21, 22, 23 and 23S located in the Laurentian Channel (Fig. 1). Samples from stations 20 and 21 have been analyzed for their porewater noble gas content and reported here.

Sediment cores were collected in transparent acrylic tubes of 62 cm length and external diameter of 10.5 cm (Fig. 2), which could be mounted on a standard 6- or 8-tubes multiple corer. Seven holes were drilled in the plastic tube, with a spacing of 6 cm, and then closed with a NPT plug (Fig. 2). Once the multi-corer was onboard, the acrylic tube was closed at both extremities with two machined PVC pistons to fit the system with a whole-core squeezer (Bender et al., 1987). The upper PVC piston has a valve to purge the air contained in the upper part of the core (Fig. 2). After installing the core on the squeezer, the lowermost NPT plug was removed and a standard copper tube (ca. 5 cc volume; Table 1) with two stainless steel pinch-off clamps (Weiss, 1968) was installed using a male NPT to tube fitting (Fig. 2). During core squeezing, the air contained in the copper tube was expelled and then the clamps were closed

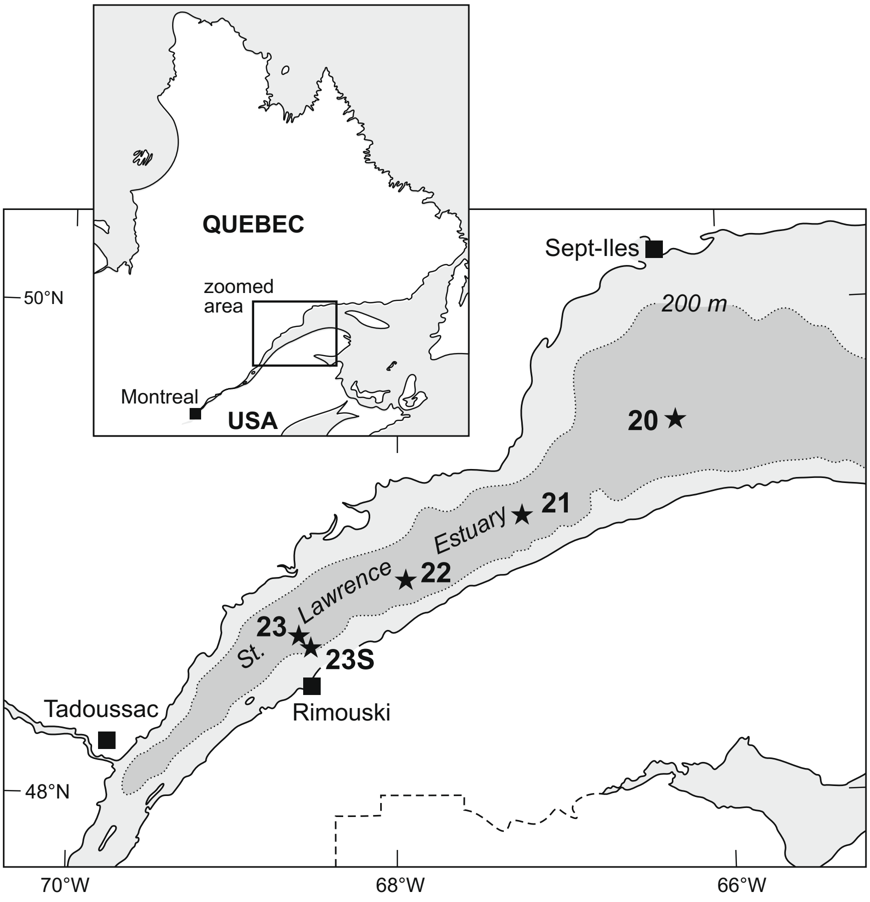

Fig. 1. Map of the St-Lawrence estuary, including the sampled stations in the Laurentian Channel. Coordinates of the stations are as follows: St20, 49°25.43′ N, 66°19.41′ W, St21, 49°05.55′ N, 67°16.94′ W; St22, 48°52.88′ N, 67°59.82′ W; St23, 48°42.07′ N, 68°38.98′ W; St23S, 48°38.84′ N, 68°35.80′ W.

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