

Anonymous Referee #2

We kindly thank Reviewer #2 for the review and taking the time to provide constructive comments on our manuscript. We considered all comments and suggestions in the revised manuscript. Our answers to each comment are presented in bold.

REVIEWER COMMENT: This paper examines N cycling processes in the subterranean estuary of an island in the Canadian Archipelago. It employs nutrient data collected over multiple years combined with previously published estimates of groundwater flow to try and quantify N removal and addition processes in the STE as well as fluxes to the coastal ocean. Overall the paper is generally well written and the data set are valuable and unique. However, I have two main issues, one having to do with interpretation and another with flux methodology.

Regarding the former, and as noted up front in the title, the study focused on the shallow portion of the STE (upper 2-2.5 m). The general lack of NO₃ within this zone as compared to the relatively high NO₃ measured in inland fresh groundwater is used to invoke substantial denitrification or other N removal process during groundwater transport to the coast. The problem with this is that their shallow sampling scheme did not allow them to capture the local fresh-saline groundwater interface (even at the furthest seaward multi port piezometer). The authors therefore cannot rule out that a NO₃ plume exists beneath the reach of their piezometers. This conclusion should be cut from the paper (or at least tempered with much of the discussion relating to it removed).

AC: We agree with the reviewer that we cannot effectively totally rule out that a NO₃⁻ plume occurs beneath the sampling location or, as suggested by reviewer#1, that a third end-member occur in the STE (i.e., a fresh and rich-NO_x EM) as in some others studies (Kroeger and Charette, 2008; Weinstein et al., 2011).

However, in this study we aim to consider the nitrogen transformations along a continuum between fresh inland groundwater and seawater by way of hydrodynamics conditions. We focus on the difference between the fresh inland groundwater input to the STE and the exportation to the coastal ocean *via* the discharge zone. We, thus, observed nitrogen transformation in relation with fresh groundwater and the upper saline circulation cell. Fresh inland groundwater, which comes from the aquifer, transports rich NO_x groundwater to the STE. Even if nitrates concentrations observed in wells (~60 μM) are higher than in beach groundwater and in the seawater, this is below the Guidelines for Canadian Drinking Water Quality. It's a low anthropogenic system where nitrates concentrations are weak. At the depth defined by our sampling approach, loss of nitrogen appears in the shallow STE, leading to depleted nitrogen exportation to the coastal ocean *via* the discharge zone.

Introduction and objectives will be refined in the revised manuscript and our conclusion will be tempered.

The second main issue is on the definition of Q_{inland} vs. Q_{beach} and how they're used to derive N fluxes through the STE. Based on the description, they should both be equivalent, but are based on different datasets? If Q_{beach} is an estimate of the fresh SGD, then how can the shallow circulated seawater be (and its associated N load) be included in the flux calculation? If the focus is entirely on the fresh SGD plume, then Q_{in} should equal Q_{out} , therefore the use of two different Q values to derive N fluxes in with N fluxes out is inappropriate. Please provide further details (even though data from other papers is used, this paper needs to stand alone even if finer details can be looked up elsewhere) and also clarify what the main focus of the mass balance is (are saline SGD N fluxes, which are typically dominant, meant to be ignored, excluded?). Overall I support the publication of this paper if these two main issues can be suitably addressed.

AC: Q_{inland} and Q_{beach} used in the manuscript were calculated by Chaillou et al., 2016. We provide further details in the revised manuscript as follow:

Hydrogeologic data from municipal and private water wells were used to estimate the inland Darcy velocity (v_{inland}) as $v_{inland} = -K \times i$, where K is the hydraulic conductivity of the aquifer and i is a mean annual hydraulic gradient from the land to the coast. K and i values were estimated from hydrogeological reports. To convert these results (cm/d) to volumetric freshwater flux (m³/d), the cross-sectional flow area was determined using GPS measurements of 1200 m of shoreline. Furthermore, based on the Ghyben-Herzberg and Glover relationship (Cooper et al., 1964), the freshwater / saltwater interface was estimated to about 73 m below the water table of the aquifer at the nearest well from the coastline. Hence, a flow depth of 73 m is used to estimate the inland groundwater flux at the coastline (Q_{inland}). Q_{inland} is then the theoretical inland groundwater export from the Permian sandstone aquifer. This rate assumes a uniform hydraulic conductivity (K) at the head of the bay and an isotropic shallow aquifer. This flux agreed quite well with a fresh groundwater flux estimate based on a mass-balance approach developed in the same area by Madelin'Eau, (2004), a private company. In the same manner, a specific discharge of local groundwater was estimated for the beach system (v_{beach}) using Darcy's Law with K^* is the hydraulic conductivity (m day⁻¹) of the unconfined beach aquifer material and dh/dl is the hydraulic gradient (m) measured using three monitoring piezometers perpendicular to the shoreline ($L \sim 30$ m, from the top of the beach to the low tide mark). Also based on the Ghyben-Herzberg and Glover relationship (Cooper et al., 1964), the top 3.2 m of the aquifer at high tide mark is fresh (except for the narrow surficial saltwater lens). Hence, we used a flow depth of 3.2 m to estimate the fresh groundwater flux (Q_{beach}) through the beach in May 2013. This flux is two times higher than the Q_{inland} . This flux was ~ 2 times higher. This difference is not surprising since SGD is highly variable on a seasonal scale: the snowmelt period is characterized by a rapid elevation of the water table in this region. In addition, the proximity of seawater and tides change hydrostatic pressure and contribute to water-level elevation in the unconfined beach aquifer compared to the regional aquifer (Pauw et al., 2014).

These explanations will be summarized and added in the manuscript (in the "study area" section).

In this approach, we used Q_{inland} and N-species concentrations in wells to estimate groundwater-born fluxes, or the volume of matter potentially exported to the

coastal zone (it is a common view used at global scale, see Beusen et al., (2013). The N-species flux within the beach were evaluated using Q_{beach} . Here the inventory was based on the entirety of the groundwater column, including the few saline samples collected in the upper saline lens. Because the water table is high (see Heiss and Michael, 2014) and based on water stable isotopes data (see Fig S1, RV1, from Chaillou et al., submitted), we assume that beach groundwater is mainly fresh (~50-70% of total SGD is fresh SGD; Chaillou, pers. Comm.). Vertical inventory allow to estimate a total N-species discharge from the shallow surficial STE.

RC: P1 Line 23: The paper has a general issue with overuse of significant figures. For example, the N fluxes here cannot possibly be accurate to for significant figures (two is probably appropriate). Same with the concentration data (e.g. 6 sig-figs used on p 12, line 10). Please correct throughout the paper.

AC: We agree, we will correct significant figures for the N fluxes and concentration data in the revised manuscript.

RC: P. 2 Line 16: sea-level has recently been shown to be a control on mixing zone dynamics: Gonnee, M.E., A.E. Mulligan, and M.A. Charette. (2013) Climate-driven sea level anomalies modulate coastal groundwater dynamics and discharge. *Geophysical Research Letters*, 40, 2701-2706.

AC: Sea level (and reference associated) will be added as an additional control of the mixing zone.

RC: P. 3. Line 24: See Saenz et al for an example of Anammox occurrence in the STE: Sáenz, J.P., E.C. Hopmans, D. Rogers, P.B. Henderson, M.A. Charette, K. Casciotti, S. Schouten, J.S. Damsté, and T. Eglinton. (2012) Distribution of anaerobic ammonia-oxidizing bacteria in a subterranean estuary. *Marine Chemistry*, 136-137, 7-13.

AC: Reference will be added.

RC: P5 Line 5: At what depth is the boundary between the beach (sand) aquifer and the sandstone aquifer? Was the inland well sampled within sand or the sandstone unit?

AC: The boundary between the sand and the sandstone aquifer is located at a depth of 20cm (Chaillou et al., 2014). Inland wells were sampled within the sandstone aquifer.

RC: P7 Line 18: Inconsistent use of super/subscripts throughout.

AC: Corrected

RC: Fig. 3A: concentration color bars (legend) do not match those in use on the figure/figure contours. Would be ideal if these plots could have the salinity contours as an overlay.

AC: We agree. The legend of Fig 3A will be corrected, and as suggested, salinity contours line will be added as an overlay of these plots (see Fig.5S- see RV1).

To sum up all our response to the referee, we will address some revisions in the new manuscript:

- Specify our objectives as we present N transformations along a continuum between fresh inland groundwater and ocean through a shallow surficial aquifer

- Precise the definition of the two end-members
- Complete information about the calculation of volumetric fluxes and the nutrient approach

Additional references:

- Beusen, A.H.W., Slomp, C.P., Bouwman, a F., 2013. Global land–ocean linkage: direct inputs of nitrogen to coastal waters via submarine groundwater discharge. *Environ. Res. Lett.* 8.
- Chaillou, G., Couturier, M., Tommi-Morin, G., Rao, A.M., 2014. Total alkalinity and dissolved inorganic carbon production in groundwaters discharging through a sandy beach. *Procedia Earth Planet. Sci.* 10, 88–99. doi:10.1016/j.proeps.2014.08.017
- Chaillou, G., Lemay-Borduas, F., Couturier, M., 2016. Transport and transformations of groundwater-borne carbon discharging through a sandy beach to coastal ocean. *Can. Water Resour. J.* 38, 809–828. doi:10.1080/07011784.2015.1111775
- Cooper, H., Kohout, F., Henry, H., Glover, R., 1964. Seawater in coastal aquifers, Geological. ed.
- Heiss, J.W., Michael, H.A., 2014. Saltwater-freshwater mixing dynamics in a sandy beach aquifer over tidal, spring-neap and seasonal cycles. *Water Resour. Res.* 50, 6747–6766. doi:10.1002/2014WR015574
- Kroeger, K.D., Charette, M., 2008. Nitrogen biogeochemistry of submarine groundwater discharge. *Limnol. Oceanogr.* 53, 1025–1039.
- Madelin'Eau, 2004. Gestion des eaux souterraines aux Îles-de-la-Madeleine Un défi de développement durable Rapport final.
- Pauw, P.S., Oude Essink, G.H.P., Leijnse, a., Vandenbohede, a., Groen, J., van der Zee, S.E. a. T.M., 2014. Regional scale impact of tidal forcing on groundwater flow in unconfined coastal aquifers. *J. Hydrol.* 517, 269–283. doi:10.1016/j.jhydrol.2014.05.042
- Weinstein, Y., Yechieli, Y., Shalem, Y., Burnett, W., Swarzenski, P.W., Herut, B., 2011. What is the role of fresh groundwater and recirculated seawater in conveying nutrients to the coastal ocean ? *Environ. Sci. Technol.* 45, 5195–5200.