

Anonymous Referee #1

We kindly thank Reviewer #1 for the review and taking the time to provide constructive comments on our manuscript. We considered all comments in the revised version of our manuscript. Our answers to each comment are detailed in bold.

REVIEWER COMMENT: This manuscript details a nice study of N-species distributions in the subterranean estuary (STE). It adds a rather regionally-unique site to the large body of coastal groundwater nutrient studies available for lower latitudes on the North America east coast. I think there are two weaknesses of this paper that should be addressed before publication:

First, the assignment of endmembers is critical for interpreting non-conservative mixing behavior. The “fresh groundwater” endmember seems poorly matched to the STE study site because the chemical composition is not similar to any of the low salinity regions within the sampled STE. In addition, it’s not clear from the few transect contour plots shown, but some of the data suggest that there may be more than two endmembers that contribute to mixing patterns within this STE. Have the authors considered the possibility of shallow and deep fresh groundwater endmembers? They may have similar salinity, but spatially separated and chemically-distinct signatures. This clearly complicates the interpretation, but it may be more accurate. For example, for NO_x, Figs 2 and 3 show a fresh, low NO_x landward EM; a fresh, high NO_x deep seaward EM; and a shallow, saline low NO_x seaward EM. This interpretation means that Fig 4 may not show extensive removal of NO_x, but simply dominant mixing of low NO_x fresh groundwater and seawater.

AUTHOR’S RESPONSE: We agree that a great difference in nitrogen concentration occur between the defined “fresh groundwater end-member” and the range of concentrations measured in the surficial STE. We have three additional response elements: the first one is based on water stable isotopic signature of the water masses, the second is based on the hydrogeological knowledge of the Cap-aux Meules aquifer, and the last is due to the dataset of nitrogen concentrations we obtained from different wells.

1) We investigated the origin of water masses present along the STE using water stable isotopes ($\delta^{18}\text{O}$, $\delta^2\text{H}$; these data have been presented in a manuscript submitted to the Journal of Hydrology). Figure 1S presents the stable isotopic signatures of groundwater and seawater samples. Beach groundwater samples plot along a line defined by:

$$\delta^2\text{H} = 5.6 \delta^{18}\text{O} - 10,$$

which is located slightly below the local groundwater – seawater line. The absence of a distinct isotopic composition in the beach groundwater samples suggests that infiltration and recharge of the beach aquifer occurred under modern climatic conditions. Furthermore, the absence of a significant depletion in ^{18}O between the fresh beach groundwater and inland groundwater samples, suggests a common origin. The depletion in ^2H observed along the salinity gradient is related to processes of evaporation, particularly in the saline samples located in the upper recirculation cell.

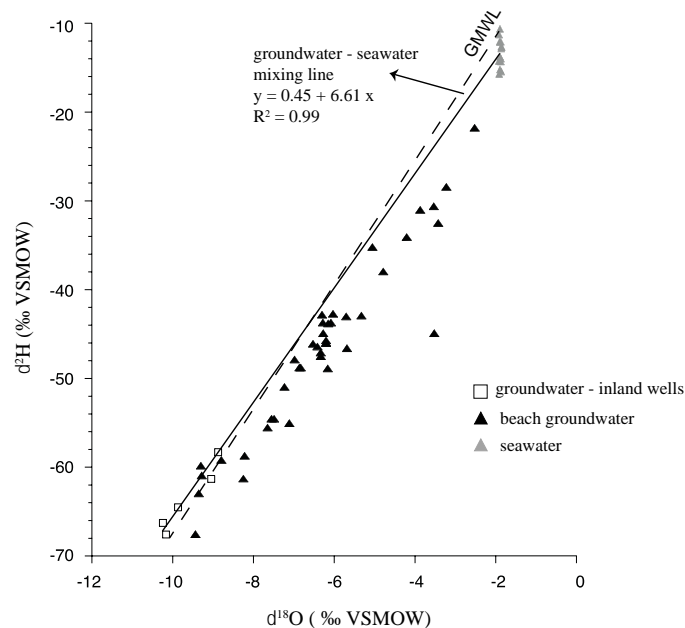


Fig 1S: Isotopic composition of groundwater samples showing the meteoric groundwater–seawater mixing line and the linear relation between samples from the beach system with their respective equations. The global meteoric water line (GMWL (Craig, 1961)) is also reported. (Confidential figure from Chaillou et al., submitted to Journal of Hydrology).

2) Concerning the hydrogeological context, there is no indication of confined deep aquifers. The seaward portion of the inland aquifer is considered isotropic and homogeneous (see Comte and Banton, 2007; Lemieux et al., 2015). However, we cannot exclude that fine sediments or organic-rich layers act as local impermeable horizons that change the water flow along the STE (see Evans and Wilson, 2017). Excepted the organic horizon located at ~30 cm below the Holocene sands in the landward part of the STE (i.e., at the top of the beach), there is no sedimentological indication of such conditions in the sedimentary core we collected using vibracoring technique at the high tide mark (i.e., ~2.5 m depth, Chaillou et al., 2014).

3) The different NO_x concentrations reported in the fresh inland groundwater were measured in different private and municipal wells located between 2000m (i.e. P5, a municipal well) and 50m (i.e. PC, a private well) from the studied transect. The nitrate concentrations ranged from 14 to 94 μM. The concentration in the seaward well PC was about 20 μM, which is close to the concentrations measured in the fresh beach groundwater samples (see figure 3A, concentration >15 μM, or the “fresh rich-NO_x EM”).

So, based on the water stable isotope signatures and hydrological context, we considered this surficial STE as a continuum between fresh inland groundwater and ocean where meteoric groundwater transit to the sea. This vision does not exclude that denitrification occurred along the transit, before the studied transect or deeper (as outlined by the second reviewer).

RC: Second, the authors argue variously for N species removal and enrichment. They provide a lot of detailed and well-written general discussion about all the possible sources of this non-conservative behavior, but very limited evidence for which processes are probably responsible for trends at their site. This would be a much stronger contribution if the authors could provide more concrete evidence for occurrence of particular geochemical processes.

AR: We, indeed, provided a lot of details about the possible transformation of nitrogen species in the subterranean estuary without any evidence of one dominant mechanism. In this revised version of the manuscript, we provide a more accurate and robust discussion on the occurrence of particular processes. Based on our large dataset, we are able to discriminate the occurrence of dominant mechanisms to explain 1) the nitrate attenuation and 2) the ammonium production.

1) *Nitrate attenuation.* Denitrification is central to the nitrogen cycle with respect to the sub-surface groundwater environment and involves the reduction of nitrate *via* a chain of microbial reduction reactions to N₂. In aquifers, lack of organic carbon to provide energy to heterotrophic micro-organisms (denitrifying bacteria that use organic carbon as the electron donor) is usually identified as the major factor limiting denitrification rates. Here, organic carbon content is not limited regarding DOC concentrations (DOC > 1.5 mM; see Couturier et al., 2016). Denitrification is probably the dominant N-pathway in this groundwater: sub-surface environments with high concentrations of labile organic matter and reducing conditions are likely to be particularly significant zones for denitrification. This process occurs all along the transit, from the aquifer to the discharge zone, upstream of the study transect and deeper as mentioned by the referee #2.

In addition to the absence of oxygen and the presence of organic carbon, reduced iron facilitates the occurrence of denitrification. Iron-oxides play a key role in the biogeochemical processes in this STE. The tidal input of oxygen induces oxic/anoxic oscillation and then the reductive dissolution of Fe-oxyhydroxides leading to total dissolved iron concentrations as high as 1 – 1.6 mM. There is some evidence that groundwaters containing Fe²⁺ contain little or no nitrate (Korom, 1992): Fe²⁺ supports autotrophic denitrification.

In this new version, the spatial distribution of Fe²⁺ will be added (see Fig 2S) and the discussion focuses on these two complementary processes.

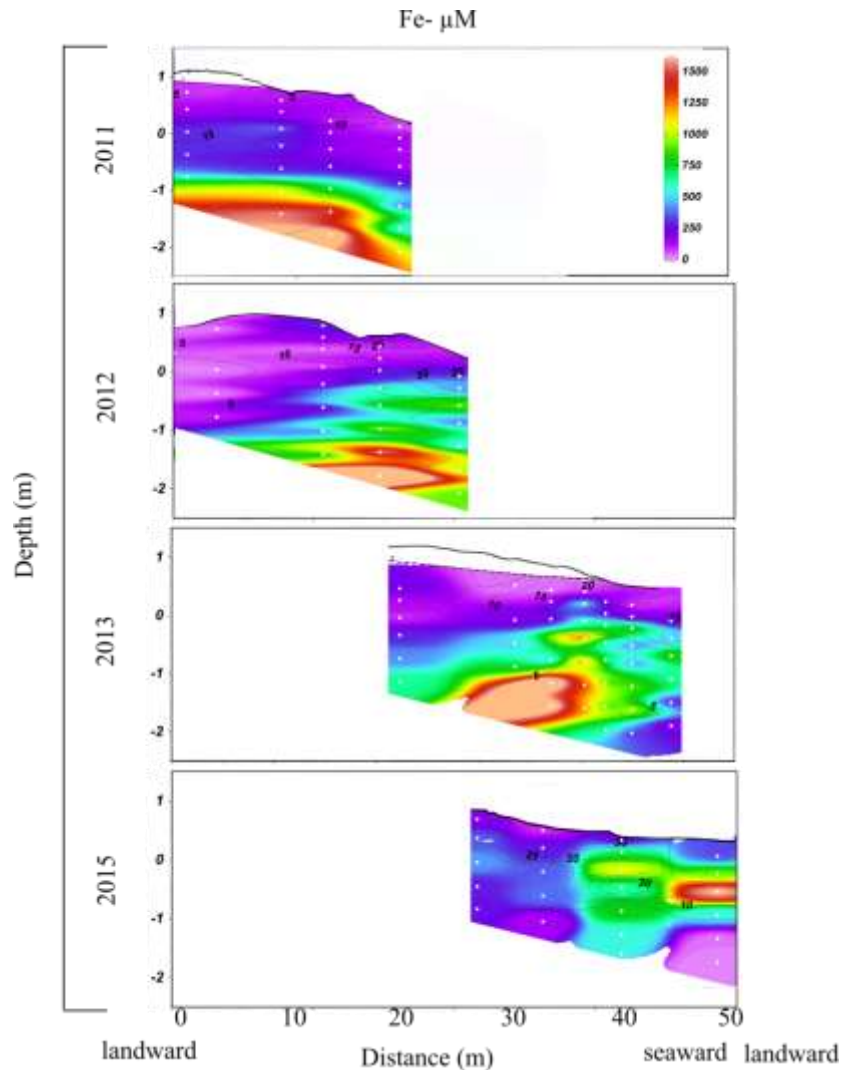


Fig.2S: Cross-sections of the transect showing the topography and distributions of Fe^{2+} in 2011, 2012, 2013 and 2015. Black contour lines refer to salinity. Depths are relative to mean sea level (i.e., 0 m is mean sea level). Contour lines were derived by linear interpolation (kriging method) of data points; the interpolation model reproduced the empirical data set with a 97% confidence level. White dots represent the depths at which samples were collected using multi-level samplers.

2) Ammonium production. The problem that we met here was to explain the origin of the high NH_4^+ concentrations measured along the shallow STE. NO_x cannot support these concentrations. As explained before, there is no evidence of additional water masses input enriched in NH_4^+ and no problem of local sewage inputs (as septic tank seeps) was observed. The only pathway to form such concentrations of NH_4^+ is the mineralisation of DOM and POM. POM content in Holocene sands and Permian sandstone aquifer is low (~5%). However, we measured very high DOM concentrations (Couturier et al., 2016) associated to high DON concentrations (>500 μM). DON concentrations are particularly high (~ 47 μM) at the high tide

mark, in the landward region of the upper saline recirculation cell. High DON concentrations are associated to high NH_4^+ concentrations (Figure 3S).

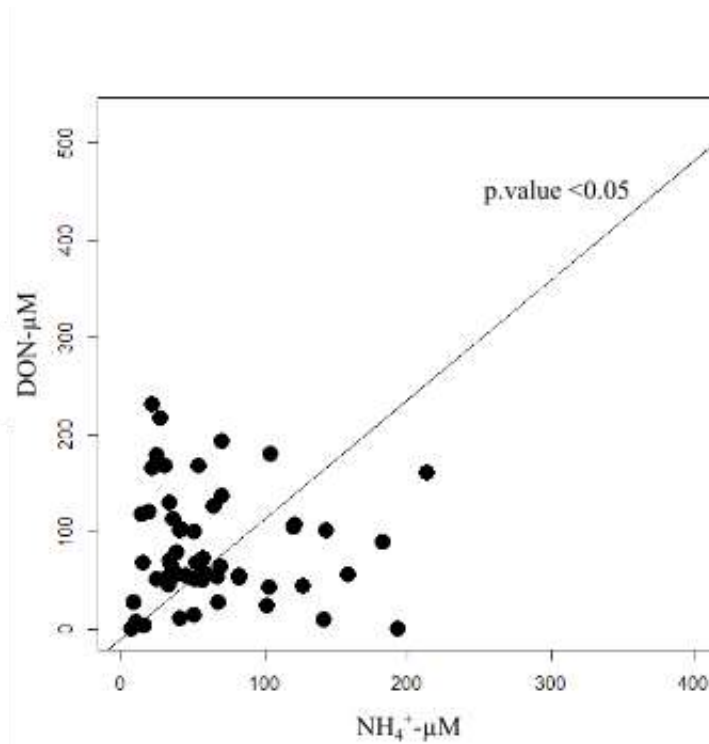


Fig 3S: Relationship between NH_4^+ and DON in the beach groundwater in 2012.

RC: L7 p7 and elsewhere. Suggest reporting dissolved oxygen in molar units instead of percent to facilitate comparison with other chemical constituents.

AR: We agree with the reviewer. The concentrations of oxygen range from 0.1 to 386μM. It appears that oxygen is completely consumed *via* the oxidation of DOC and is probably a minor oxidant for reduced metabolites produced in the sub-oxic / anoxic part of the STE. The replenishment of oxygen is not sufficient.

RC: L18 p7. I disagree with the “fresh endmember” choice. Is this really representative of water entering the study STE, especially since the NO_x and DON mixing lines don't seem consistent with the STE samples (Fig 4)? Seems like the best choice would be from within the site boundaries.

AR: There is indeed a difference between the defined fresh end-member N value and concentrations in the beach groundwater sample. Values in fresh end-member were measured in 4 different private and municipal wells located between 2000m and 50m from the landward part of the STE. NO_x concentrations ranged from 14 to 94 μM. The concentration in the closest wells (i.e., 50m) of the STE is about 20 μM, which is close to the concentration measured in the fresh beach groundwater samples (i.e., 5-12 μM, see figure 4S).

As explained above, we think this fresh end-member is quite representative of water entering the STE, water stable isotope signature confirms that this fresh end-member is a source of water to the STE. We chose this end-member as we wanted to explain not only the N transformations and source within this shallow surficial subterranean estuary, but also show the transformation along the continuum between fresh inland groundwater and ocean (this idea is more clearly presented in the introduction of the new version of the manuscript). In this case, we found more appropriate to take this end-member. However, we do not exclude that denitrification occurred along the transit, before reaching the study STE as suggested by the referee 2.

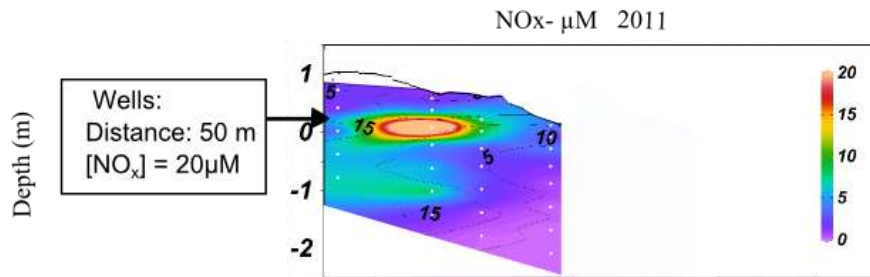


Fig.4S: Cross-sections of the transect showing the topography and distributions of NO_x. Black contour lines refer to salinity. Depths are relative to mean sea level (i.e., 0 m is mean sea level). Contour lines were derived by linear interpolation (kriging method) of data points; the interpolation model reproduced the empirical data set with a 97% confidence level. White dots represent the depths at which samples were collected using multi-level samplers.

RC: The spatial and salinity patterns almost seem to suggest 3-EM mixing, with were seasonal or spatial/depth differences greater? In L24 p8, note that assessment of removal or addition depends on 2- vs. 3-EM mixing. Hard to evaluate this further without seeing the spatial distribution of NO_x and NH₄ similar to Salinity in Fig 2.

AR: We already discussed and justified our 2-EM mixing in the previous comments (p.1-2). The spatial and depth difference observed between the different study periods are controlled by 1) the multi-samplers location along the cross-shore transect (we focused on the top of the beach in 2011 and 2012 and on the intertidal area in 2013 and 2015); 2) the beach morphology that changes over the seasons due to spring tides, storms and ice covers. For example, in 2015, we obtained a post-storm-type profile, with no seaward accumulation in comparison with 2011 and 2012 profiles (see the morphology of the beach surface in Fig. 5S). Because the upper saline recirculating cell moves along a seaward – landward direction (which lead to the displacement of the “biogeochemical reactor”), we need to “normalize” the target species distribution. Here, salinity is used as a conservative proxy of the mixing to obtain a global view of N-species distribution along the STE whatever the recirculation cell location. Nevertheless, we agree with the referee 1 and we will add a figure with the spatial distribution of NO_x and NH₄⁺ similar to salinity (see Fig 5S).

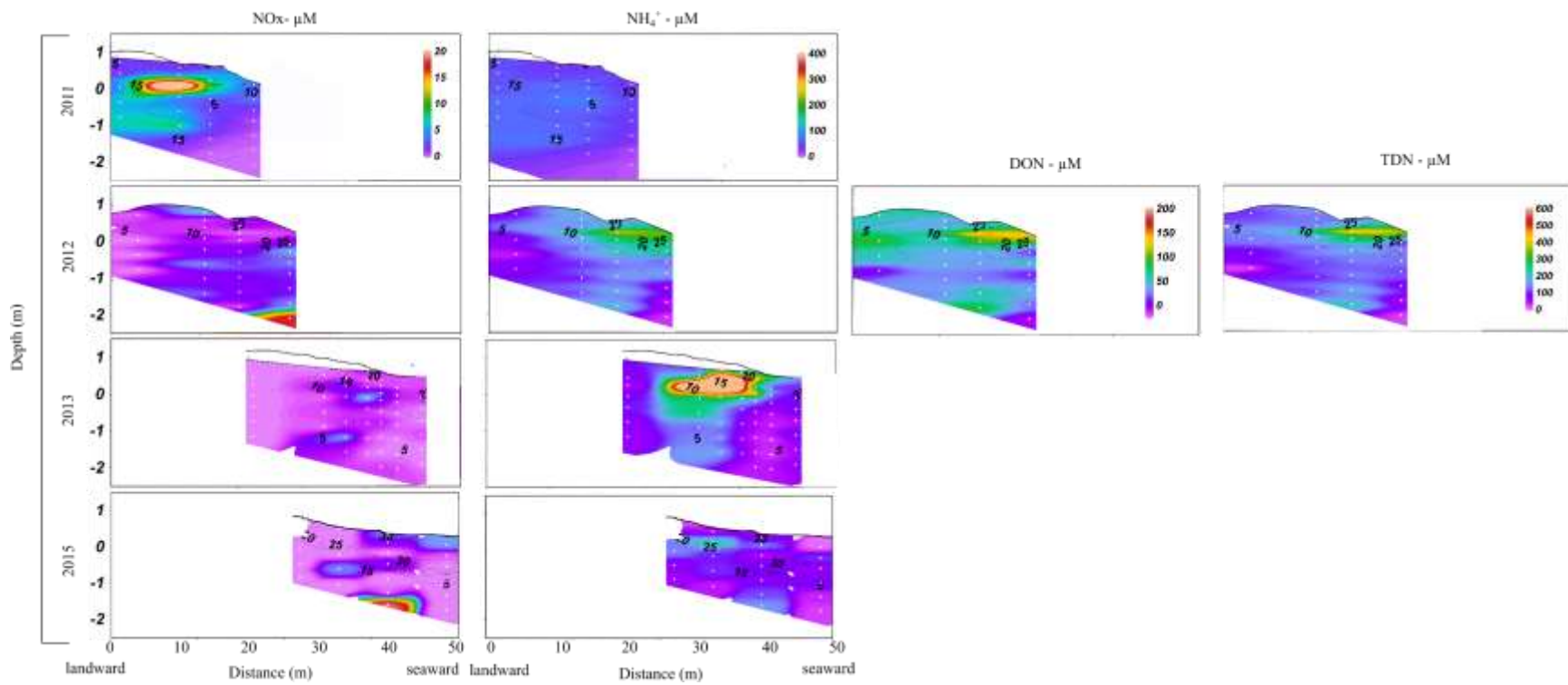


Fig 5S: Cross-sections of the transect showing the topography and distributions of NO_x^- , NH_4^+ in 2011, 2012, 2013 and 2015 and DON, TDN in 2012. Black contour lines refer to salinity. Depths are relative to mean sea level (i.e., 0 m is mean sea level). Contour lines were derived by linear interpolation (kriging method) of data points; the interpolation model reproduced the empirical data set with a 97% confidence level. White dots represent the depths at which samples were collected using multi-level samplers.

RC: The distances in Figs 1-3 don't match. I also don't like Fig 3 (even though 2012 was apparently the most complete with respect to N species) because it doesn't appear to capture the most landward "inventory" site. Or maybe it's just because the distances are all mixed up.

AR: As mentioned by the referee, distances don't match on Figures 2 and 3; distances in 2012 should be 35 m, and 20m in 2013. We will correct Fig 1-3 with the correct distances in the revised manuscript. Also, as discussed in the last point, we will add the spatial distribution of NO_x and NH₄⁺ in 2012, 2013 and 2015. The figures are presented above.

RC: L25 p8. Does NH₄ really decrease under high O₂ conditions? How was this evaluated? It's not at all apparent from the data shown in Fig 4.

AR: We evaluated this point using linear regression ($R^2=65\%$, $p.value<0.05$), but we agree with the referee that the decrease is not really apparent on Fig. 4 due to high variability in the groundwater beach.

RC: L3 p9. How much water was pumped before stabilization of GW quality parameters (L3p6)? How much volume was pumped for samples? Could co-existing NO_x and NH₄ be an artefact of sample volumes that overlap redox boundaries?

AR: We pumped no more than 700mL for stabilization of GW quality parameters and samples. We made preliminary tests in 2011 to avoid overlap. We estimated that, considering the permeability and porosity of the sediments ($K= 11.4 \text{ m d}^{-1}$; $\phi=0.25$; Chaillou et al., 2016), and the size of the sampling pore ($\phi=0.9 \text{ cm}$), the maximal volume is about 1 to 1.5 L (depending of the distance between the sampling pores). We cannot totally exclude the overlapping of redox boundaries due to sampling artefacts and sampling pore depth. However the co-occurrence of NO_x and NH₄⁺ were found under oxygen depleted conditions, each year in same location (i.e., below saline circulation cell and mixing zone). We assume it is not an artefact.

RC: The discussion in section 4.1 doesn't really say much about the current study site. So how variable do the authors think that this STE is with respect to salinity and redox conditions? Does the STE structure change temporally relative to the snowmelt period? How did the June sampling periods relate to snowmelt during the study years?

AR: We think that salinity and redox conditions are quite constant over the year in the spring period (see comments on the spatial distribution). In the Îles-de-la-Madeleine Archipelago, the presence of snow in May is common. Maximal aquifer recharge occurs between April and June (see figure 6S from (Chaillou et al., 2013).

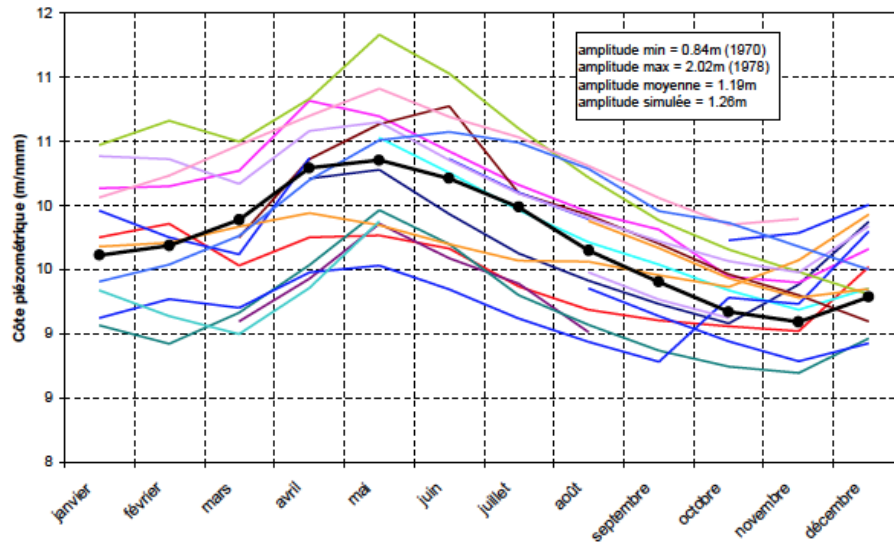


Fig 6S: Water tables from municipal wells of Cap-aux-Meules Island. The black line is the mean simulated level from (Madelin'Eau, 2004).

We assume that high water table level, due to the snowmelt, leads to a dominance of fresh groundwater inland in the STE and spatially limit the landward extension of saline circulation cell. There is no information on the impact of snowmelt on the STE structure. According to our knowledge, our studies are the first work on boreal and snowmelt-affected STE. In a recent work, Heiss and Michael, (2014) showed that the size of the circulation cell and area of the mixing zone were mostly affected by seasonal water table oscillations. The intertidal circulation cell expanded horizontally and vertically as the inland water table declined, displacing the fresh discharge zone and lower interface seaward. When water table is maximum, the beach groundwater was mostly fresh as we observed in the Martinique Beach in June. Calculations based on Darcy flow combined with Rn-based mass balance SGD estimation (Chaillou et al., submitted to the Journal of Hydrology) show that fresh groundwater mainly contribute to total SGD; the seawater infiltration is minimum and only limited to surficial intertidal Holocene sands (~ 30 cm depth). We think this period is important for chemical exports to the ocean.

As pointed by the referee, the section 4.1 is not essential and the information will be added to the section 2.1 “Study area”.

RC: L16 p11. If DNRA depends on Corg availability, would it also be expected to be depressed due to high lignin/low labile DOC?

AR: We agree with the reviewer. It is also expected that DNRA will be depressed due to high lignin/low labile DOC. It is why we think those heterotrophic and autotrophic denitrifications are dominant processes to explain the loss of nitrate as previously discussed.

RC: The discussion of biogeochemical N-transformations is rather speculative. In Section

4.2.1, the NO_x distribution suggests removal (or maybe not, if there are more than two endmembers, see above). If removal, then reduction to N₂ and reduction to NH₄ are suggested as possibilities. This discussion is well-written, but doesn't really lead to a useful conclusion. In the end, it's not clear to me if any of the discussed pathways are any more likely than the others. ("More work is needed" is not a very satisfying summarizing sentence.)

AR: We agree with the referee. In a previous comment (p.3), we pointed out the main pathways. We will add complementary information in the revised manuscript and present a better structure for our discussion.

In Section 4.2.2, I agree that the NH₄ source is probably organic matter oxidation, especially considering the sediment layer containing 20% OM. It's not clear to me why it is important whether the NH₄ come from decomposition of DON or POM/sediment OM, or the importance of the pathway by which organic carbon is oxidized (sulfate reduction, Eq 5, or something else). It would however be useful if the authors could say something about where the NH₄ enrichment occurs in this STE and why (e.g., in Fig 3, at the 15m profile).

AR: We agree and as suggested by the referee we will provide information on the location of the NH₄⁺ enrichment (see Fig. 5S, add in the revised version). Production of NH₄⁺ is mainly located in the mixing zone where salinity ranges from 10 to 20. The highest NH₄⁺ concentrations are observed upstream of the saline circulation cell where the highest DON concentrations are found.

RC: L8, p13. I don't see these "hot spots", except maybe deep and seaward in Fig 3. I would find this discussion much more interesting if the authors showed and discussed their results in more detail, and limited the speculation. None of the discussion in this section is specific to this location or to the results found in this study.

AR: We will provide additional figure of NO_x distribution in 2012, 2013 and 2015 in order to show these hot spots in the revised manuscript (see fig. 5S). We agree that this section is rather speculative. We will add this section to the section 4.2.1 on the nitrate attenuation.

We use the term "hotspot" to describe local production of nitrate in a nitrate depleted environment (i.e., in 2013 and 2015, 190 cm and 50 cm depths). We attributed this local production of nitrates to local and sporadic production. Some local production may occur due to micro-environment and non-steady state conditions.

RC: The nutrient inventory approach to flux estimation seems a bit misguided, given the work by e.g., H. Michael and C. Robinson showing the finer structure in transport through the STE (i.e., not just plug flow). For the nutrient inventories, it appears (Fig 1) that the data are from 2013? Or are data from different years somehow combined? If so, what is the spatial region considered for combining different profile locations?

AR: We agree that some studies now succeed in describing finer structure of transport through STE (Abarca et al., 2013; Heiss and Michael, 2014; Robinson et al., 2014). These studies showed SGD are mostly dependent of tide level and water table oscillations. Using radon 222 and water stable isotopic analyses, Chaillou et al., (in revision) described in more details transport through the STE of Martinique Beach. However, variability in geochemical constituent and non-conservative transport are still difficulties to afford accurate nutrient fluxes.

We think there is a misunderstanding of our explanation of inventories approach as we are not just using plug flow. Nutrient inventory approach has the advantage to capture the entire nutrient variability on the depth at given position (see the work of Gonnee et al., 2013).

Nutrients inventories are reported from 2013 for NO_x and NH_4^+ , since water flux calculations were made in 2013 using Darcy's Law. Unfortunately, DON and TDN values are not available in 2013, thus we used data from 2012 to estimate DON and TDN inventories. As location of some multi-level samplers is superimposed from year to year, we used the same multi-level samplers to performed inventories in 2012 and 2013.

RC: L12, p15. The “filter” interpretation depends on how you account for dilution. Presumably, the inventories would somehow have to be normalized to salinity? Also, here and the discussion on p16, the non-conservative behavior interpretation (and also flux) depends on endmember assignment. I would argue that the STE is not a sink for terrestrial N because any removal relative to the upland well endmember must have occurred prior to reaching the “onshore” profile (or according to Fig 3, the farthest onshore profile). If anything, the inventories in Fig 5 suggest that groundwater is enriched in N at the HTM profile, and the STE is therefore a source of N to the coastal ocean.

AR: We agree that the non-conservative behavior interpretation depends on end-member assignments. As discussed previously (p.1-2), we are confident with the choice of these two end-members.

We agree that inventories at the HTM profile suggest enrichment, however, at the LTM, inventories show an attenuation of nitrogen species. This is all the difficulties to estimates accurate fluxes to the ocean. The Fig. 5 needs more precision about the discharge zone, the arrow is not representative enough as fresh groundwater discharge should occur after the saline circulation cell. We will correct this figure in the revised manuscript.

Considering Q_{inland} and Q_{beach} measured, and the concentration in fresh inland groundwater as well as the inventories, we observed a loss of total N in the discharge zone. We agree with the reviewer that most of the NO_x species removal must have occurred prior the STE. But results also showed a removal of DON through the beach groundwater (Fig.4). DON concentration comes from the fresh inland groundwater and Couturier et al., (2016) showed that DOM in fresh inland groundwater and in beach groundwater exhibited terrestrial signature.

However we may temper our conclusion. Indeed, total N loss was estimated to 14 mol m⁻¹ y⁻¹ which represents a loss of 13%. Input of DIN concentration is almost similar to the DIN exported. However, the interesting point is the change in nitrogen exported to the coastal water.

RC: L2, p17. It's not clear how relevant these comparisons are. For example, the comparison with the St Lawrence River would probably be more appropriate if the authors scale their shoreline fluxes to the shoreline length for the entire nearshore region receiving the river input (assuming this is justified).

AR: We agree, this comparison was poorly adapted. In the revised manuscript, we will compare our results with nutrients exportation by STE. Thus our results are lower than others reported studies such as Anschutz et al., 2016; Li et al., 2009; Weinstein et al., 2011.

RC: Should check significant figures in the tables.

AR: We are going to check and correct significant figures in the tables

RC: Fig. 4. Suggest using different symbol shapes in addition to color to distinguish oxygen level categories.

AR: We agree, we are going to used different symbol shape and color to distinguish oxygen level (Fig. 7S).

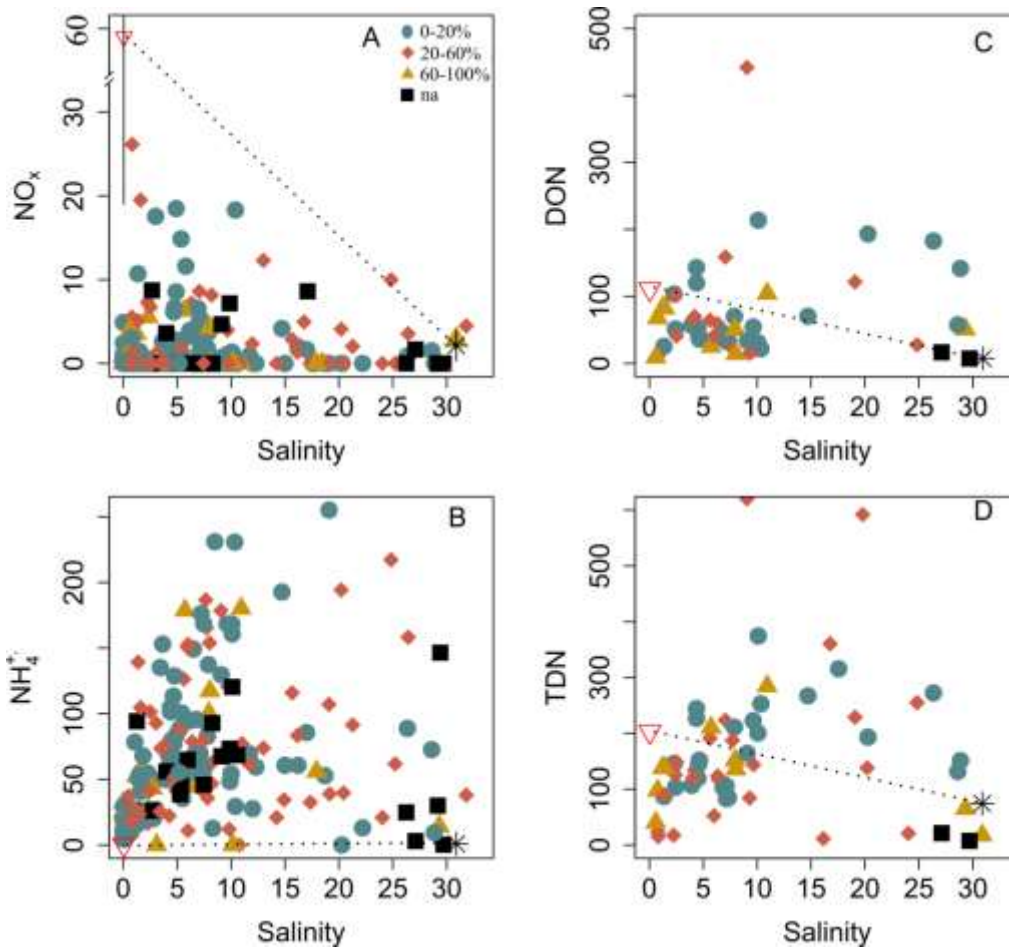


Fig.7S: Mixing plot of NO_x and NH_4^+ groundwater concentration in $\mu\text{mol L}^{-1}$ collected in 2011, 2012, 2013, and 2015 (A, B) and DON and TDN in 2012 (C, D) within the STE relative to the salinity grouped for different DO saturation from 0-20%, 20-60% and 60-100%. Black square were used when no data on DO saturation were available. Red triangles are mean groundwater end-member values and black squares are mean seawater end-member values. Standard deviations are black lines associated with end-members. Dashed lines represent the theoretical mixing line between groundwater and seawater end-members.

RC: Fig. 5. “saline saltwater” is rather redundant wording.

AR: We will correct the grammar and check for redundancy in the revised manuscript

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:

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