

Response to Reviewer 2

R2.1 General comments:

This is a well-written and relatively concise manuscript that uses $\delta^{15}\text{N}$ of nitrate data to trace the distribution of Pacific versus Atlantic waters in the Northwest Atlantic. This type of analysis is not new, the manuscript by Granger et al. (2018) previously laid the groundwork. However, while the focus of these two papers is similar, the current manuscript presents new relationships that estimate the fraction of Pacific water (based on both N^ and the $\delta^{15}\text{N}$ of nitrate) using a more extensive dataset and discuss some possible applications (food-web studies, paleoceanographic reconstructions, etc...).*

- We thank the reviewer for taking the time to read the manuscript and for offering their comments and constructive criticisms.

R2.2 *I have some minor comments to improve the manuscript. First, it would be best to separate the results from the discussion to improve clarity and focus, if at all possible. Most of the text before section 3.4 could be moved to a Results section, as these sections are mostly descriptive, the remainder could be re-organized into a proper discussion section.*

- We generally agree with this suggestion. Sections 3.1 to 3.3 contain mostly descriptive results and can be re-titled as “Results” (with existing subheadings), with some moving of the more inferential statements to a new Discussion section. Sections 3.4 to 3.8 will be re-titled as “Discussion” with some minor re-organization.

R2.3 *Second, some analytical detail or background information in the discussion needs to be added (see specific comments below).*

- Please see our responses to points R2.5 to R2.9 below. These details will be added to section 2.2 of the Materials and Methods.

R2.4 *Finally, I wonder if the $\delta^{18}\text{O}$ data could be explored in more detail. These data are shown in Figure 3, but poorly discussed in the manuscript.*

- Respectfully, a deeper exploration of the $\delta^{18}\text{O}_{\text{NO}_3}$ data would require a more detailed discussion of the factors that modulate $\delta^{18}\text{O}_{\text{NO}_3}$ variability, which would go beyond the scope of the ms. The primary focus is on $\delta^{15}\text{N}_{\text{NO}_3}$ variability because it is preserved in organic materials and therefore important in isotope ecology and paleoceanography contexts. The $\delta^{18}\text{O}_{\text{NO}_3}$ data help to *support* the interpretations of $\delta^{15}\text{N}_{\text{NO}_3}$ variability (Figures 5 and 6 and associated discussion) and provide additional insights as for example the interpretations of sedimentary denitrification in BBW (Lehmann et al. 2019), but the $\delta^{18}\text{O}_{\text{NO}_3}$ in isolation are not a diagnostic tracer for Pacific water. We propose to rephrase the final paragraph of the introduction to clarify that the focus is on $\delta^{15}\text{N}_{\text{NO}_3}$.

Specific comments:

R2.5 Materials and methods:

Lines 96-107: Is using different types of filters (0.45 μm versus 0.22 μm) affect nutrient concentrations? Was this tested?

- The use of different filter sizes was an unintended consequence of obtaining samples opportunistically during different expeditions. However, most organisms that could impact nutrient concentrations between the point of collection and subsequent freezing and analysis are larger than 0.45 μm . We did not perform any specific tests, but there is no indication that any data were impacted by the small difference in filter pore size. We also note that 0.45 μm filters are used regularly for nutrient measurements.

R2.6 *Lines 121-122: Was USGS 32 used to correct $\delta^{15}\text{N}$ data? Since its $\delta^{15}\text{N}$ is much different from the $\delta^{15}\text{N}$ of the samples, I assume this would be problematic.*

- The reviewer is correct in noting that the USGS 32 standard lies beyond the range of sample $\delta^{15}\text{N}_{\text{NO}_3}$ values. It is used in a 3-point calibration as part of routine operating procedures in the Dalhousie lab. Omitting USGS 32 from the calibration curve had negligible impact (<0.2‰) on sample $\delta^{15}\text{N}_{\text{NO}_3}$ values.

R2.7 Lines 123-125: Why was NO_2^- not removed? Even small NO_2^- concentrations can affect the $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ of NO_3^- , especially at low NO_3^- concentrations. What was the lowest NO_3^- concentration for samples analyzed for isotopic composition? Since nitrate concentrations are generally high below the mixed layer, and for most water masses discussed in this manuscript, I don't think this is a major concern.

- The reviewer is correct in their last point about higher concentrations below the mixed layer in the water masses of concern. The NO_2^- concentrations in this zone were always less than $0.36 \mu\text{M}$, as shown in Figures S3 – S7, which is too low relative to the much higher NO_3^- concentrations ($< 6 \mu\text{M}$) to affect the isotopic compositions. We report some isotope data from mixed layer waters with NO_3^- concentrations as low as $0.7 \mu\text{M}$, but since the focus is on waters below the mixed layer, this is not a major concern. We will clarify this point in the revision. We will also clarify that “ NO_3^- ” actually refers to the sum of NO_3^- and NO_2^- throughout the manuscript.

R2.8 Another concern is that the “denitrifier” method is sensitive (to some extent) to the $\delta^{18}\text{O}$ value of the sample water because of O exchange with water during the conversion of NO_3^- to N_2O . This is an issue if the $\delta^{18}\text{O}$ of the samples and standards are drastically different or in polar regions where the $\delta^{18}\text{O}$ of water is greatly variable due to mixing between freshwater from rivers and glaciers (with a low $\delta^{18}\text{O}$ around -20‰) and seawater ($\delta^{18}\text{O}$ of about 0‰). Was this taken into account while analyzing their samples? See Kobayashi et al. (2021) for more detail.

- We did not apply a correction for the $\delta^{18}\text{O}$ of seawater. However, the $\delta^{18}\text{O}$ in the sample waters range only from -2.2‰ to 0.2‰ (Lehmann et al. 2019) which would have a minor impact on the $\delta^{18}\text{O}_{\text{NO}_3}$ results. We will note this in the revision.

R2.9 Finally, what was their blank size (i.e., for the bacteria method)?

- The blank size constituted $< 2 \%$ for the routine 20 nmol target analyses, and $< 5 \%$ for the low concentration 5 nmol target surface water analyses. We will state this in section 2.2.

R2.10 Lines 144-145: The authors should make a clearer distinction between regenerated (calculated using Redfield P: O_2 ratio and AOU) versus preformed PO_4^{3-} here. Preformed nutrients are those that were present in solution when the parcel of water sank from the surface and are characteristic of different water masses: pre-formed $[\text{PO}_4^{3-}] = \text{measured } [\text{PO}_4^{3-}] + \text{regenerated } [\text{PO}_4^{3-}]$

- Agreed. We will articulate a clearer distinction between preformed and regenerated PO_4^{3-} in the revision.

R2.11 Lines 160-161: Explain what cause that kink in the NO_3^- vs PO_4^{3-} relationship at low nutrient concentrations (i.e., nitrate assimilation).

- We will expand on the sentence at line 160 to explain the kink in the NO_3^- vs PO_4^{3-} relationship for Pacific water in the Canada Basin.

Results and Discussion:

R2.12 Lines 367-371: I am curious about the isotope effect for nitrate assimilation derived from these relationships and how it compares to previous field studies (e.g., Altabet et al. (2001)?

- We calculated the apparent fractionation for NO_3^- assimilation for the few stations with a sufficient number of shallow water $\delta^{15}\text{N}_{\text{NO}_3}$ data, based on the slopes of $\delta^{15}\text{N}_{\text{NO}_3}$ versus the negative natural logarithm of the fraction sub-euphotic zone NO_3^- concentrations. The fractionation factors were smaller (0.8 to 3.9‰) than the generally assumed 5‰ fractionation for assimilation, but consistent with observations in Granger et al. (2010). However, we choose not to report these results, as they do not address the objectives of the manuscript.

R2.13 Lines 415-416: Why isn't the correlation showed for $\delta^{18}\text{O}-\text{NO}_3^-$? Could $\delta^{18}\text{O}$ of NO_3^- also be used as a complementary tool to trace these different water masses? This aspect should be better discussed.

- With respect to the first point, we will add the correlation matrices (as in figure S8) for $\delta^{18}\text{O}_{\text{NO}_3}$ to the supplementary figures. With regard to discussion of the $\delta^{18}\text{O}_{\text{NO}_3}$ data, please see our response to point R2.4 above.

R2.14 Lines 427-429: The authors should explain why no change in water-column $\delta^{15}\text{N-NO}_3^-$ is expected during sedimentary denitrification (i.e., discuss the suppressed net “community” isotope effect for sedimentary denitrification due to diffusion limitation and complete NO_3^- consumption in the sediments).

- We will add a sentence to expand on our existing explanation for why sedimentary denitrification does not impact water column $\delta^{15}\text{N}_{\text{NO}_3}$.

R2.15 Line 468: Could a similar equation be derived for $\delta^{18}\text{O}$ of NO_3^- as well? However, $\delta^{18}\text{O}$ of NO_3^- would not be useful for food-web or paleoceanographic studies as the O atom is not conserved during N incorporation into organic material.

- The reviewer is correct in their assessment that $\delta^{18}\text{O}_{\text{NO}_3}$ has little utility with respect to foodweb or paleoceanographic applications, in the sense that oxygen is not conserved during incorporation into biological materials. This is our rationale for not showing the relationship between $\delta^{18}\text{O}_{\text{NO}_3}$ and fPW. We will add a sentence to make this clearer.

R2.16 Lines 471-478: This argument needs to be discussed better since as for N^* , it is not possible to disentangle different co-occurring processes (nitrification, denitrification, N_2 fixation) using $\delta^{15}\text{N}$ of NO_3^- data solely. These co-occurring processes were disentangled in the BBW because of the additional insights from $\delta^{18}\text{O}$ of NO_3^- data.

- We propose to expand this section, tightening up our argument for the utility of $\delta^{15}\text{N}_{\text{NO}_3}$ as a proxy for fPW.

R2.17 Line 483: This section title is vague. I would rename it “Using our $\delta^{15}\text{N-NO}_3^-$ relationship to establish a baseline $\delta^{15}\text{N}$ for food-web and paleoceanographic studies.” This section could also be merged with sections 3.7 and 3.8.

- We will retitle section 3.6 accordingly. But we prefer to maintain the current structure of sections 3.6, 3.7 and 3.8.

R2.18 Line 565: Change to “fraction of Pacific water”

- We agree that writing “fraction of Pacific water” instead of the abbreviated “fPW” in the conclusions will assist the reader. We will make this change.

R2.19 Table 1. Add number of samples analyzed for each water masses (n).

Indicating a range of depths for each water masses would be better than showing the average (given the large standard deviation).

- We will incorporate both of these suggestions.

R2.20 Figure 3. Are $\delta^{18}\text{O}$ of NO_3^- values shown at about 200 m depth (177) and 500 m depth (ROV5) outliers? It is unclear why there is no corresponding increase in the $\delta^{15}\text{N}$ of NO_3^- at these stations/depths. Were these samples measured in duplicate?

- These are most likely analytical outliers, especially given the lack of a corresponding increase in $\delta^{15}\text{N}_{\text{NO}_3}$. Unfortunately, neither of these samples was measured in duplicate. In the interests of data transparency, we choose to show these data. However, we will remove the dashed lines connecting these points to the rest of the profile data, which should help to improve figure clarity. We will explain this in the figure caption.

R2.21 Figure 5. I think it would make sense to separate the symbols based on depths for this figure (e.g., as in Figure 2: surface waters impacted by nitrate assimilation (open symbols) versus deeper waters (filled symbols)).

- Respectfully, we disagree with this suggestion to reformat the symbols in Fig. 5. However, to improve clarity we propose to rescale the symbol sizes representing NO_3^- concentrations. This will make it easier for the reader to infer which samples were impacted by NO_3^- utilization.

2.22 Figure 7. The R^2 as well as p -values should be added.

- We will add r^2 and p-values to the plot, as requested.