Interactive comment on "Contrasting decadal trends of subsurface excess nitrate in the western and eastern North Atlantic Ocean" *by* Jin-Yu Terence Yang et al.

Anonymous Referee #1

Review of 'Contrasting decadal trends of subsurface excess nitrate in the 2 western and eastern North Atlantic Ocean' by Yang et al.

This is a nice contribution that I recommend be published. I do have some comments that should be addressed and these are detailed below. Overall the manuscript is well written and the figures are clear and complete.

We thank Referee #1 for the positive evaluation and insightful comments. We have addressed the concerns raised by this referee in the revised manuscript, and thoroughly describe all changes made in our responses. Where no change to the text has been made we provide a full justification.

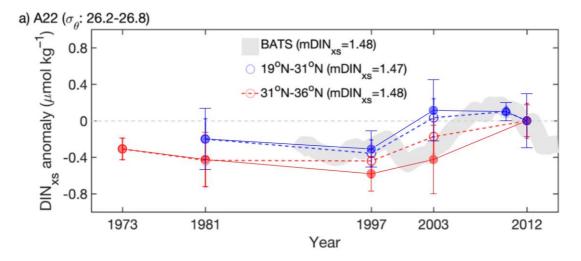
Line 45: 'an evidence' – change to just 'evidence'?

Please include a short discussion on the potential for any bias as a result of not having reliable concentration data <0.1 μ mol kg⁻¹ for DIN and 0.01 μ mol kg⁻¹ for DIP. (Change made): A short discussion about possible biases in results has been added to the revised manuscript (lines 116–119). Our analysis for estimation of excess DIN focused exclusively on data collected from 200–600 m depth, where nutrient concentrations were greater than 1.4 μ mol kg⁻¹ for DIN and 0.08 μ mol kg⁻¹ for DIP. More explicitly, the lower ends of the DIN and DIP concentrations in these targeted waters are several-fold higher than the detection limits of DIN and DIP. As low DIP concentration (< 0.1 μ mol kg⁻¹) may result in uncertainties (Martiny et al. 2019), to eliminate any potential bias in the DIN_{xs} estimates we did not use those DIP and accompanying DIN data (accounting for 1.4% of the total 1955). Removal of the low DIP concentration data did not alter our finding (e.g., the trend of increasing excess nitrate in the western subtropical NAtl).

Lines 125–128: Please make it clearer whether this procedure was conducted by the authors of the current manuscript, or if this is a correction made prior to publication of the datasets the authors use. I also do not fully understand what this correction does? Please explain in clear terms why this correction need to be performed (i.e., why do the datasets need this correction to be made comparable in the first place?). Some details for this might be shifted from the supplement to the main text. An alternative option would be to state explicitly that this is discussed in more detail in the supporting information, but it would be useful if the key reason behind the corrections could be summarized succinctly in the main text. (Explanation provided and changes made): We agree that we did not clearly describe data calibration issue in the original manuscript. In the revised manuscript (lines 136–143) we have included a section describing the data calibration and methods, which reads:

The GLODAPv2 product includes data obtained from > 700 cruises during the period 1972–2013. These large datasets collected in different years and by different investigators may contain some systematic and analytical errors. To remove these systematic errors, quality control of the data was performed by Key et al. (2015) and by Olsen et al. (2016), largely based on comparison of repeated measurements made for waters deeper than 2000 m at the same locations. Any biases found were corrected by applying adjustment factors to the raw datasets, and the adjusted datasets were reported in the GLODAPv2 product (Key et al., 2015; Olsen et al., 2016).

The corrections noted in italics above were performed by two groups (Key et al., 2015; Olsen et al., 2016). More recent data, collected during the 2010s, were not thoroughly compared with the data obtained in the 2000s or earlier. In addition, we removed the influence of remineralization of organic matter by considering the changes in AOU among different cruises. Therefore, we applied additional minor adjustments to all of the datasets obtained over three decades. In the absence of these small corrections (summarized in Table S2) we still found consistent increasing rates of excess DIN using the original uncorrected datasets (solid symbols); for example, since 1997 along the transect A22.



Paragraph stating line 130: What is the cause of this inter-dataset difference? Analytical measurement errors?

(Change made): In the revised manuscript (lines 136–138) we have stated that the difference in the N and P concentrations was largely caused by use of different analytical instruments and analysts.

Line 151: "In addition, the effect of seasonal variations on DINxs signals at this depth layer is generally insignificant," Please clarify, which layer are the authors referring to? Two different layers are discussed earlier in the paragraph.

(Change made): In the revised manuscript (lines 169) we have indicated that the effect of seasonal variations in DINxs signals found at 200–600 m depth was insignificant, because this depth range is typically deeper than the winter mixed layer in the study area.

Line 190: "Based on multiple cruises along each transect, changes in DINxs were discernable over the decadal periods; these changes were most pronounced between 200 m and 600 m (Fig. 2)" How many data time points are these rate calculations based on? I understand this varies depending on the cruise line. I think it is important to include this information somehow on Figure 2.

(Change made): Data from three cruises that occurred between the GO-SHIP and WOCE time periods were used to calculate the DINxs change in Figure 2. For the A22 transect we used 418 and 187 data points for the depth intervals 200–600 m and 1200–1500 m, respectively. For those depth intervals we used (respectively) 371 and 208 data points for the A20 transect, and 1168 and 613 data points for the A16N transect. We have added the number of data points used to the caption of Figure 2.

Line 197: "Moreover, the Δ DINxs values remained close to zero in the intermediate waters (1200–1500 m) in the western and eastern subtropical NAtl (Fig. 2). This observation

confirms that the marked changes in DINxs largely occurred in the subsurface waters." This does not seem to be the case for the A16n line (i.e., deeper waters show the same trend as the surface waters here).

(Change made): We agree that the DINxs changes in the subpolar region along the A16N transect occurred in both the subsurface and intermediate waters, whereas changes in the subtropical regions were only found in the subsurface waters. In the revised manuscript (lines 215-219) we have clarified that the smaller decrease in the DIN_{xs} values in the intermediated waters north of 45° N along the A16N transect relative to those in the subsurface waters was probably associated with the deep winter convection and formation of the North Atlantic Deep Water in the subpolar NAtl. In this subpolar region there is a close connection between the subsurface and intermediate waters, which probably led to the DINxs decrease in the subsurface and intermediate waters. In contrast, changes in DINxs in the subtropical region were only found in the subsurface waters.

Line 203: "layer of the DINxs maximum decreased since 1997" Do the authors mean 'increased' instead of 'decreased'? (Change made): We have changed "decreased" to "*increased*".

Paragraph starting line 220: Please attempt to describe N deposition rates quantitatively. i.e. to back up statements such as 'pronounced increase' and 'considerable AND input' (Change made): Quantitative information on AND has been added to the main text (lines 241–243), which now reads: "Model results have showed that the total AND over the NAtl basin in 2000 varied between 35–70 mmol N m⁻² year⁻¹, reaching higher values along the US coastal areas (Duce et al., 2008)."

Paragraph starting line 234: Do the authors use the mean rate of deposition for the coastal AND sampling sites? Can an error bar therefore be added to the deposition trend in Fig. 5? This would help support the statement 'trend . . . commonly found at AND monitoring sites' (Change made): The revised Figure 5 shows the 5-year moving means for wet NO_x deposition (orange solid curve) along the coast of the USA. Note that the range of the 95% confidence intervals (indicated by the orange shading) indicates the error in those mean deposition values.

Line 264: ". . .although the mismatch between the observed time lag and the ventilation age of water masses may be due, at least in part, to the biological processes." For full clarity, please briefly specify the biological processing being referred to here. (Change made): The biological processes involve planktonic assimilation of anthropogenic N during photosynthesis, and subsequent gravitational sinking and bacterial oxidation of organic matter at depth (lines 264–270). The oxidation of organic matter containing anthropogenic N may contribute to the elevation of the N:P signals at depth. In the revised manuscript (lines 285–289) we have briefly described these biological processes.

Lines 274–278: But here anthropogenic nutrient input is from a different continent? Please clarify.

(Change made): In the revised manuscript (lines 300–302) we have stated that the source of nutrient to the Mediterranean Sea is the "European continent". This is intended to indicate a similar phenomenon found in the other oceans, and highlights anthropogenic factors having a lag period of 20 years in their effect on subsurface nutrient dynamics.

Line 320–322: Would the detection limit of phosphate in surface waters be low enough to detect this change due to increased N_2 fixation?

(Change made): We agree that the change in surface DIP may be biased because of its very low concentration. We have rewritten this sentence (lines 347-350), which now reads: "...if N_2 fixation activity had increased during the study period we would have expected more DIP in the surface ocean to be remineralized in the thermocline, leading to an increase in the subsurface concentration of DIP (Kim et al., 2014a), but no subsurface increase was observed (Fig. 4)".

Line 429: "particularly in the USA" Rephrase to "particularly from the USA"? (Change made): We have changed "particularly in the USA to "*particularly from the USA*".

Line 732: "To ensure consistent comparisons between atmospheric N deposition rates and seawater DINxs anomalies, the seawater DINxs anomaly values were shifted by approximately 15 years." Please state exact time shift and if it was added or subtracted. (Change made): In the revised manuscript (line 738) we have stated that "the seawater DIN_{xs} anomaly values were shifted backward by 15 years."