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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 #2

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The paper "Contrasting decadal trends of subsurface excess nitrate in the western and eastern North Atlantic Ocean" presents an impressive compilation of nutrient data from the North Atlantic (NA) Ocean. The main result is a DIN excess increase in subsurface waters of the western NA during the last 3 decades, which might be explained by anthropic atmospheric nitrogen deposition. Some other results, interesting interpretation and discussion follows, but according to my analysis the main result should be discussed before, relying on the accuracy of DIP measurements necessary for DIN excess calculation. I will begin with this major concern and follow with other comments.

Major concern:

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Page 5 (106-109): the analysis of nutrient data was based only on concentrations greater than 0.1 μmol kg-1 for DIN and 0.01 μmol kg–1 for DIP. These concentration levels approximate the detection limits of DIN and DIP for the analytical methods used in the field observations (Zhang 109 et al., 2001; Hydes et al., 2010).

A similar colorimetric method is used to measure nitrate and phosphate in seawater using autoanalyzers, and there are no apparent reasons to consider a detection limit which is 10 times lower for phosphate than for nitrate. It would have been the case if the cell used to measure phosphate compare to nitrate had been 10 times longer, but it was not the case at least for the WOCE and previous cruises. Considering Redfield proportion (N:P=16:1) or what you have considered (N:P=15:1), it is clear that more than 10 times increased precision is useful to correctly interpret biogeochemical processes in the Sea. It is the reason why many efforts were done to lower the quantification limits for phosphate measurements, using nanomolar methods particularly in surface waters. A recent paper stipulates that "The underlying reason for a limited understanding in the distribution of surface DIP is that the standard methodology has high variance and low interlaboratory accuracy, below ~100 nM" (Aoyama et al., 2016; Martiny et al. 2019). Even if 100 nM may appear as a higher limit, it is the one you have considered for nitrate, and following my argument in the first sentence of this paragraph, it may be a plausible accuracy value. I ask you therefore to determine the uncertainties of DIN excess values, considering a 100 nM uncertainty in DIP measurements, in order to see if the increasing trend in DIN excess is still observable in this condition. This point is my major concern. It should at least be put forward first in your discussion.

Other comments:

Line 36: You use DIN for nitrogen; therefore, it would be preferable to use DIP for phosphate.

Lines 48-52 It would be preferable to reinforce the demonstration of the main result rather than propose new hypotheses far from the main result.

Line 55 Nr means nothing for me. You could use DIN, defined as the sum of nitrate NO_3^-, nitrite NO_2^- and ammonium NH_4^+ where nitrite is usually negligible.

Lines 59-60 Anthropogenic nitrogen deposition (AND) to the contemporary ocean is comparable in magnitude to marine biological N_2 fixation: add reference(s).

Line 64 Delete pollutant nitrogen, replace with DIN.

Lines 93-94 The sentence at the end of this paragraph uses an older reference than the statement just before, making it a bit confusing.

Lines 102-109 This refers to my major concern explained in the upper part of this report.

Lines 114-115 Which is a major source region of anthropogenic nitrogen according to a model-derived atmospheric NOx deposition (Dentener et al., 2006; Fig. 1). It is not data but model-derived prediction. This information is important and needs to be added here and not only in the figure legend. If you want to look at a comparison between prediction and data in another context, I invite you to read this short interesting paper (Grüber, 2016).

Lines 138-140 The finding that the subsurface deltaDINxs signals were considerably greater than the detection limit of DIN is a strong indication that our data adjustments probably did not influence the temporal trend of DINxs. I agree with the adjustments, but deltaDINxs signals depend on DIN and DIP, and the accuracy will largely differ depending on what you choose as a quantification limit for DIP (see my first comment).

Line 145 Deficit? Don't you mean excess?

Line 166 Fig. S4. See Line 109 in the SM, after (b), A20 is missing.

Line 178 I am not sure that the introduction of anomalies here helps the readers. You will then compare anomalies and anomalies of excess, which have completely different uncertainties.

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Lines 183-200 It is the main result which needs to be reinforced with a discussion on DINxs uncertainties. I wonder if a result/discussion part on DIN evolution rather than DINxs (depending on DIP concentration which may be harder to measure with enough accuracy) evolution will not be more straightforward and easier to publish.

Line 189 Fig. S6. See Line 131 in the SM, different and no difference.

Lines 201-202 Fig. 3 presents DINxs and not deltaDINxs.

Line 203 (measurement year)?

Lines 221-222 which is subject to considerable AND input from the North American continent. Add reference(s).

Line 222 Recent studies suggest that the reduced form of nitrogen… Do you mean NH_4^+?

Line 224 Thus? Marine nitrogen fixation is an autochthonous process which influences DINxs!

Line 225 Therefore?

Line 226 Emissions? Only deposition. If you add significant amount of NH_4^+ by the atmosphere, it will certainly influence DINxs values. I am not able to follow your reasoning there. NO_x is introduced without being defined.

Lines 266-278 All this part will be clearer if DIN inventory is used instead of DINxs inventory.

Line 315 In this region (boxes 1–3). Please, add Fig. 5a.

Line 317" The atmospheric NO_x deposition. Replace with "the modelled atmospheric NO_x deposition".

Line 346 The formation of the STMW is generally enhanced when the NAO index becomes negative. Please, add reference(s).

Line 356 Fig. S7b. Please also represent the DIN anomaly, and for each graph use the proportion you have defined (N:P=15:1) in the axes to better represent N and P. Refer to my remark for Fig. S12.

Line 370 but little change in DIP was observed (Fig. S12). If you correctly represent the axes using N:P=15:1, as defined as a relevant ratio by yourself, you might not conclude that little change in DIP was observed. For DIN represented between 12.5 and 14.5 μmol kg-1, please represent DIP between 0.83 and 0.97 μmol kg-1. It is the only way to graphically compare N and P evolutions. Are the evolutions of DIN and DIP different? or are the uncertainties between DIN and DIP measurements, at the level needed, different?

Line 364-409 All this part is speculative. A paper focusing on proving the main result would be more interesting.

References cited:

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