

## **Response to Anonymous Referee #2**

**We thank Anonymous Referee #2 for the time and effort devoted to the review of the manuscript. Below, we reproduce the reviewer's comments and address their concerns point by point. The reviewer's comments are copied below in regular font, with our responses in red.**

**We are responding to this review a long time after it was published on the *Biogeosciences Discussion* online forum because we had to submit a companion paper by Caffin et al. (this issue), cited in this article, before the closure of OUTPACE's special issue on December 31, 2017.**

### **Summary and Overall Impressions**

This is a well-written, interesting, enjoyable paper that quantifies the source of "new" nitrogen to the euphotic zone as well as the flux of nitrogen derived from N<sub>2</sub> fixation with respect to diazotroph community and other factors. Data was collected from three stations in the western tropical south Pacific ocean. Two of these stations were oligotrophic while the third was "ultra" oligotrophic. Nitrogen fixation was overwhelmingly the largest source of N input due in part to very high rates of nitrogen fixation and lower than typical rates of atmospheric deposition. N flux from nitrogen fixation was uncoupled from N inputs, and possible reasons for this are well discussed. This paper is well presented and the "story" is clearly told. Everything is well organized and easy to follow. The conclusions are interesting and important, and I really liked the 5 day averaging and well resolved vertical profiles. Daily variability is high and important to measure! In addition, the comparison between in situ and on deck incubations is useful and important, as most researchers are unable to use in situ arrays. The introduction and discussion sections are very well done, but I do have some concerns about some of the methods and the presentation of the results. My primary concerns are methodological and regard 1) the nitrogen fixation rate calculations including the absence of a reported detection limit and a failure to completely address the failings of the "bubble method" and 2) propagation of error throughout calculations and some additional statistical comparisons. These issues can all using data already in hand, and I think that these changes are unlikely to impact the conclusions of the manuscript. Nevertheless statistical rigor and proper calculations are important. I have also listed some line item concerns below.

### **General Comments**

Throughout the paper, nitrate transport is referred to as a "diffusive flux", but "diffusion" brings to mind molecular diffusion, whereas this flux is really a result of vertical mixing. Please rename this term or, if this is standard terminology, indicate clearly that this is not molecular diffusion but is a physical mixing process.

**Rev. 2 is right and nitrate transport is now referred to as "turbulent diffusive flux" to avoid any confusion with molecular diffusion.**

Regarding atmospheric deposition, was atmospheric deposition of NH<sub>4</sub> considered? What about DON? - These would be "new" N that look like regenerated N. Also, throughout the manuscript, "atmospheric deposition" reflects dry deposition only (as opposed to wet). Please modify the text throughout so that this distinction is clear.

**Rev. 2 argues that NH<sub>4</sub><sup>+</sup> and organic N (particulate and dissolved) should also be considered when quantifying atmospheric deposition. Our quantification of the atmospheric deposition could be underestimated as it only represents dry deposition and that gas and organic forms were not measured. At global scale and depending on the**

location, organic nitrogen could represent up to 90 % of N atmospheric deposition (Kanakidou et al., 2012). Even if we double our estimated deposition flux, atmospheric deposition remained low (< 1.5 %) and consequently represented a minor contribution of the new N input. We have changed “atmospheric deposition” to “dry atmospheric deposition” throughout the text, as recommended.

Kanakidou, M., Duce, R. A., Prospero, J. M., Baker, A. R., Benitez-Nelson, C., Dentener, F. J., ... & Sarin, M. Atmospheric fluxes of organic N and P to the global ocean. *Global Biogeochemical Cycles*, 26(3), 2012

Some of the notation was a bit confusing was used both for integrated N<sub>2</sub> fixation rates (i-N<sub>2</sub>) and density and nitracline density (NO<sub>3</sub>). I know that these are established conventions, but use of the same abbreviation is a bit confusing and these should be clarified somehow.

To clarify this confusion, “ $\rho$ ” is now used to refer only to density and “ $\rho\text{NO}_3$ ” to nitracline density. The special notation of integrated N<sub>2</sub> fixation rates has been deleted as it was not used in the text.

The methods section could use a bit more detail and/or additional references. I have indicated specific problems below.

**We have taken into account the specific problems explained below.**

In section 2.1, a more clear description of which parameters led to the designation of the three stations would be useful, i.e. LD A was oligotrophic and Tricho dominated, etc. A map of station locations would be helpful, to be referenced on p. 4 lines 11-14, which could be Figure 1 or similar.

**A clearer description of which parameters led to the designation of the three stations is proposed, but it appears in the Introduction because most of the parameters concerning the diazotroph domination (i.e. *Trichodesmium* vs UCYN), the trophic gradient (i.e. oligotrophy vs ultra-oligotrophy) and the global N<sub>2</sub> fixation rates of each region (i.e. MA vs SPG) have been explained and published in other papers (Stenegren et al., this issue; Bonnet et al., this issue; Bonnet et al., 2017; Moutin et al., 2017). The detailed description of the whole transect is beyond the scope of this paper.**

**“The WTSP Ocean has recently been identified as a hotspot of N<sub>2</sub> fixation, including N<sub>2</sub> fixation rates >500  $\mu\text{mol N m}^{-2} \text{d}^{-1}$  (Bonnet et al., 2017). The region covered by the OUTPACE cruise is characterized by trophic and N<sub>2</sub> fixation gradients (Moutin et al., 2017). The region covered by the OUTPACE cruise encompasses contrasting trophic regimes characterized by strong differences in top nitracline depths, from 46 to 141 m (Moutin et al., this issue) and representing a large part of the oligotrophic gradient at the scale of the world Ocean (Moutin and Prieur., 2012; their Fig. 9). The westward oligotrophic waters are characterized by high N<sub>2</sub> fixation rates ( $631 \pm 286 \mu\text{mol N m}^{-2} \text{d}^{-1}$ ) mainly associated with *Trichodesmium* (i.e. within the hotspot around Melanesian archipelago waters, hereafter named MA), and the eastward ultra-oligotrophic waters (in the eastern border of the South Pacific gyre, hereafter named SPG waters) are characterized by low N<sub>2</sub> fixation rates ( $85 \pm 79 \mu\text{mol N m}^{-2} \text{d}^{-1}$ ), mainly associated with UCYN (Bonnet et al., this issue; Stenegren et al., this issue).”**

**Then, in the section 2.1 we have mentioned that LD A and LD B were positioned in the MA waters, and LD C in the SPG waters. To be clearer, we refer to Figure 1 on p. 4, lines 11-14, as proposed by reviewer #2.**

Regarding the nitrogen fixation rate calculations, while I understand the reluctance to add equilibrated seawater to the incubations, especially given the oligotrophic nature of the samples, the compromise proposed here (to measure  $^{15}\text{N}_2$  at the conclusion of the incubation) does not adequately reconcile the rates calculated here with the problems associated with the bubble method. It has been shown that the fraction label of the dissolved  $\text{N}_2$  "source pool" changes during the course of the incubation (Mohr 2010 and others). However, the method used in this manuscript assumes a steady fraction label of that pool, based on the fraction label measured at the end of the incubation, inevitably resulting in an underestimation of nitrogen fixation rates. This is problematic because a) different organisms fix at different times of day and b) the rate of change likely varies based on physical and chemical parameters. Problem (a) is especially relevant to this study as the researchers have specifically set out to compare regions with different diazotroph assemblages. At this stage, this problem cannot be addressed directly, but some discussion of the implications of this in light of the conclusions is warranted. Problem (b) can and should be directly addressed by correcting the rate calculations for the rate of bubble dissolution. Examples of this correction can be found in Figure 1 of Mohr (2010) and the Supplemental Figure in Jayakumar (2017). Since these were incubated on an in situ array, differences in temperature with depth could have variably impacted rates of  $^{15}\text{N}_2$  dissolution. This is addressed to some degree in those two publications and should be discussed in this manuscript.

**We totally agree that the  $^{15}\text{N}_2$  dissolution is progressive as shown in Mohr et al. (2010) We have added the following sentence in the discussion section to mention this important aspect:**

**“We are aware the dissolution kinetics of  $^{15}\text{N}_2$  in the incubation bottles may have been progressive along the 24 h of incubation (Mohr et al., 2010), therefore, the  $\text{N}_2$  fixation rates provided here represent minimum values”.**

Atmospheric deposition is really a flux to the mixed layer. Depending on mixed layer depth relative to the nitracline, mixing speeds, and biological uptake rates the upward flux of  $\text{NO}_3$  may reflect a flux to the sub-mixed layer euphotic zone only. Nitrogen fixation rates also show vertical structure. I assume that diazotroph communities have vertical structure (might be nice to put this on Figure 2 if these data were collected, as well as density to show mixed layer depth); Tricho floats (if much of the Tricho is floating on the surface, was it even sampled?). Diatoms can adjust their bouyancy. The 200 m over which everything is integrated is a fairly large region. How does one account for these mini-environments within that 200 m in the budget? Could the vertical structure in N fixation reflect higher  $\text{NO}_3$  concentrations (even though they're undetectable – but the d.l. in this study was pretty high for oligotrophic waters) closer to the nitracline? Some discussion of the vertical structure within the euphotic zone may be informative.

**We focused on the photic layer (from the surface to 0.1 % of surface irradiance), and on what comes from above and below the photic layer, and what is produced within the photic layer. The photic layer that we studied goes down to 125 m, 100 m and 200 m for LD A, LD B and LD C, respectively. We presented integrated rates (per  $\text{m}^2$ ) and considered the whole photic layer, and we are aware of the heterogeneity of the diazotroph communities' vertical structure and the processes that happened in the entire photic layer. Data of diazotroph communities' vertical structure was collected and is**

available on the OUTPACE database (<http://www.obs-vlfr.fr/proof/php/outpace/outpace.php>). In the study we focus on N<sub>2</sub> fixation, a specific paper (Stenegren et al., this issue) focus on diazotroph communities' structure during the OUTPACE transect, which was beyond the scope of the main focus of this paper.

The abstract could use more contextual information and conclusions. In its current form it reads as a bit of a data dump. The interesting conclusions of the paper could be better showcased in this section.

**In accordance with this comment and that of reviewer #4, we have rewritten the Abstract as follows:**

“We performed nitrogen (N) budgets in the photic layer at three contrasting stations representing different trophic conditions in the western tropical South Pacific (WTSP) Ocean during austral summer conditions (Feb. Mar. 2015). Using a Lagrangian strategy, we sampled the same water mass for the entire duration of each long duration (5 days) station, allowing us to consider only vertical exchanges for the budgets. We quantified all major vertical N fluxes both entering the system (N<sub>2</sub> fixation, nitrate turbulent diffusion, atmospheric deposition) and leaving the system (particulate N export). The three stations were characterized by strong nitracline and contrasted deep chlorophyll maximum depths, which was lower in the oligotrophic Melanesian archipelago (MA, stations LD A and LD B) than in the ultra-oligotrophic waters of the South Pacific gyre (SPG, station LD C). N<sub>2</sub> fixation rates were extremely high at both LD A ( $593 \pm 51 \mu\text{mol N m}^{-2} \text{ d}^{-1}$ ) and LD B ( $706 \pm 302 \mu\text{mol N m}^{-2} \text{ d}^{-1}$ ), and the diazotroph community was dominated by *Trichodesmium*. N<sub>2</sub> fixation rates were lower ( $59 \pm 16 \mu\text{mol N m}^{-2} \text{ d}^{-1}$ ) at LD C, and the diazotroph community was dominated by unicellular N<sub>2</sub>-fixing cyanobacteria (UCYN). At all stations, N<sub>2</sub> fixation was the major source of new N (> 90 %) before atmospheric deposition and upward nitrate fluxes induced by turbulence. N<sub>2</sub> fixation contributed circa 13-18 % of primary production in the MA region and 3 % in the SPG water and sustained nearly all new primary production at all stations. The e-ratio (e-ratio = particulate carbon export / primary production) was maximum at LD A (9.7 %) and was higher than the e-ratio in most studied oligotrophic regions (~1 %), indicating a high efficiency of the WTSP to export carbon relative to primary production. The direct export of diazotrophs assessed by qPCR of the *nifH* gene in sediment traps represented up to 30.6 % of the PC export at LD A, while their contribution was 5 and < 0.1 % at LD B and LD C, respectively. At the three studied stations, the sum of all N input to the photic layer exceeded the N output through organic matter export. This disequilibrium leading to N accumulation in the upper layer appears as a characteristic of the WTSP during the summer season, although the role of zooplankton in export fluxes should be further investigated.”

## Technical and Line Item Comments

### Abstract

Line 20: "...all major fluxes..." could be "...all major vertical fluxes..."

**This has been corrected in the new version**

Line 21: Instead of "Thanks to a Lagrangian..." this would be more clear as "Using a Lagrangian..."

**This has been corrected in the new version**

Line 22: "...allowing to consider..." Should this read "allowing us to consider"?

**We have changed this to "allowing us to consider"**

Line 23: Might it be easier to refer to stations as A, B, and C without the 'LD', as it does not seem to have any necessary significance.

**The reference "LD" has been kept in the interests of consistency with all the papers of the OUTPACE special issue.**

Line 26: The N<sub>2</sub> fixation measurements appear to have been depth integrated (units  $\mu\text{mol N/m}^2/\text{d}$ ), but what depth region were these integrated over? Also, what is meant by "extremely high"? In comparison to what, are these high?

**N<sub>2</sub> fixation rates were integrated over the photic layer (i.e. from surface to 0.1% of surface irradiance, as described in the section 'Nitrogen fixation rates' of the Material and Methods), corresponding to surface to 105, 80 and 180 m for LD A, LD B and LD C, respectively. The "extremely high" N<sub>2</sub> fixation rates measured here are in the upper range of the rates reported in the global N<sub>2</sub> fixation Marine Ecosystem Data (MAREDAT) database (Luo et al., 2012), that we have compared our findings with.**

Line 30-31: "N<sub>2</sub> fixation...at all stations." Does this refer only to the LD A, B, and C stations? If not, please state how many stations were sampled in each region for these data.

**In this sentence, we only referred to the three stations LD A, LD B and LD C, on which we focused in this study.**

Line 31: PC and PP have not been defined.

**This has been corrected as follows: "The e-ratio (e-ratio = particulate carbon export / primary production) was maximum at..."**

Line 34: "there contribution" should be "their contribution"

**This has been corrected in the new version**

Line 36-37: "This disequilibrium...summer season..." I don't understand this sentence. Does this mean that this disequilibrium is generally held to be true (confirmed by other studies) or that you found this here and that it was consistent across all stations?

**This sentence means that we observed a disequilibrium of N input vs. N output in the photic layer when we performed the budgets (short time scale), in contrast to Eppley and Peterson (1979), who mentioned that at the year time scale, under ideal steady state, input of new N in the upper zone should be equal to N loss through PON export.**

**Eppley, R. W. and Peterson, B. J.: Particulate organic matter flux and planktonic new production in the deep ocean, *Nature*, 282(5740), 677–680, doi:10.1038/282677a0, 1979.**

Line 37: The mention of zooplankton seems strange. I would delete it.

**The potential role of zooplankton with regard to export seems to be important as suggested in this study, even if difficult to assess. Thus, we prefer to maintain the sentence.**

## **Introduction**

p 2 Line 5: "di-nitrogen" Is this journal format? I have generally seen it written as "dinitrogen".

**It has been corrected to “dinitrogen” in the new version**

p 2 Line 5: "ammonia" At seawater and physiological pH, it is primarily "ammonium" that is present.

**Here, we referred to the equation of the N<sub>2</sub> fixation process which is:**



**Rev. 2 is correct, because although ammonia (NH<sub>3</sub>) is the direct product of this reaction, it is quickly ionized to ammonium (NH<sub>4</sub><sup>+</sup>)**

p 2 Line 7-11: "In the oligotrophic...photic layer." Run-on sentence.

**The sentence has been rewritten as follows: “In the oligotrophic ocean, N availability often limits phytoplankton growth (e.g. Moore et al., 2013) and N<sub>2</sub> fixation sustains a significant part of new primary production (PP, i.e. the production unrelated to internal recycling of organic matter in the photic layer), such as in the North (Karl et al., 1997) and South Pacific Ocean (Moutin et al., 2008), the western Mediterranean Sea (Garcia et al., 2006), or the tropical North Atlantic (Capone et al., 2005).”**

p 3 Line 8: "(see below)" This is not necessary.

**It has been deleted in the new version**

p 3 Lines 9-13: What region are these N<sub>2</sub> fixation rates integrated over?

**In their study, Bonnet et al. (2017) have integrated the N<sub>2</sub> fixation rates over the photic layer, i.e. from surface to 0.1% of surface irradiance, as we did in this study.**

## **Materials and Methods**

p 3 Line 22: "strong thermal stratification" Please provide data or a reference for this statement or delete it. What qualifies as "strong" thermal stratification?

Rev. 2 is right that austral summer condition is sufficient to describe the stratification occurring in this area and we deleted the statement.

p 3 Line 23: "...along a west-east...French Polynesia." Please also indicate the location using lat/long, as the precise locations of New Caledonia and French Polynesia do not spring immediately to mind.

**We understand that the precise locations of New Caledonia and French Polynesia do not immediately spring to mind, thus we have added the location using lat/long in the text “...from New-Caledonia (22°00’S – 166°00’E) to French Polynesia (17°30’S – 149°30’W).”**

p 3 Line 25: "diazotrophs" should be "diazotroph"



**This has been corrected in the new version**

p 3 Line 29: "firstly" should be "first"

**This has been corrected in the new version**

p 4 Line 19: Doesn't PAR stand for "Photosynthetically Active Radiation", not "Photosynthetically Available Radiation", as written here?

**We confirm that PAR stands for “Photosynthetically Available Radiation”, as written in the manuscript.**

p 4 Line 19: Does "fluorescence" refer to chlorophyll a fluorescence, or some other set of wavelengths? Please be specific.

**Here, fluorescence referred to chlorophyll a fluorescence. We have specified this in the new version.**

p 5 Line 3: "In situ Chl a concentration was..." Should be "In situ Chl a concentrations were..."

**This has been corrected in the new version**

p 5 Line 3: Was the AquaTraka III an in situ sensor attached to and deployed with the CTD package? So, this was used instead of the SeaBird chl sensor? Or is the AquaTraka for shipboard measurements? This is a little unclear because I expected to see the SeaBird chl sensor used with the SeaBird package. Please clarify.

**The AquaTraka III sensor is an in situ sensor which was mounted on the CTD and used instead of the SeaBird chl sensor. This has been clarified in the new version. “...fluorescence measurements performed with a AquaTraka III (Chelsea Technologies Group Ltd) sensor mounted on the CTD.”**

p 5 Line 4-7: Please include more details on collection of the NO<sub>3</sub> and PO<sub>4</sub> samples were they filtered? Were they stored or run immediately at sea?

**Two samples for NO<sub>3</sub><sup>-</sup> and PO<sub>4</sub><sup>3-</sup> concentration measurements were collected from Niskin bottles in 20-mL Polyethylene bottles. One sample was directly analyzed on-board and the other poisoned with 50 µl HgCl<sub>2</sub> (20 g L<sup>-1</sup>) and stored for analysis after the cruise in the laboratory. The details have been added in the new version.**

p 5 Line 9: Were these incubations in glass bottles? please indicate. How many replicates were used?

**The PP rates were measured in triplicate using the <sup>14</sup>C tracer method (Moutin and Raimbault, 2002) in 150-mL polycarbonate bottles. The details have been added in the new version.**

p 5 Line 13-17: Were these integrated over the upper 200 m? It looks like that's so, but please state it specifically.

**The integrations were performed from the surface to 20 m below the deepest depth. As the depths of sampling were chosen according to surface irradiance levels to cover the entire photic layer, the deepest depth (0.1 % of surface irradiance) was different at each station: 105 m, 80 m and 180 m for LD A, LD Band LD C, respectively (as mentioned p5**

line 10-12). Thus, the integration was performed from surface to 125 m, 100 m and 200 m for LD A, LD B and LD C, respectively. This has been specified in the new version of the manuscript.

p 5 Line 19: "DIN" should include ammonium, which can account for ~40% of total N deposition (Dentener et al. 2006). I suggest renaming the combined NO<sub>3</sub>/NO<sub>2</sub> term to NO<sub>x</sub> or similar to avoid confusion.

**In accordance with this comment, we have renamed the combined NO<sub>3</sub>/NO<sub>2</sub> term as NO<sub>x</sub>.**

p 5 Line 19-22: Since the reference is "submitted", these methods should be explained in greater detail or additional references given.

**We completely agree with this comment. Thus, we have completed this section in collaboration with Cécile Guieu who performed the measurements. Cécile Guieu will therefore be added as co-author of the manuscript in its final version.**

**The new version of the section is:**

**“N atmospheric deposition (NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup> (nitrite), hereafter called NO<sub>x</sub>) was quantified along the transect after dissolution of aerosols collected continuously during the transect, as described in Guieu et al. (in rev.). Briefly, the sampling device, designed to avoid ship contamination, was installed at the look-out post in the front of the ship, collected aerosols at ~20 L min<sup>-1</sup> on onto polycarbonate, 47-mm diameter, 0.45-μm porosity (previously acid-cleaned with a 2% solution of HCl (Merck, Ultrapur, Germany) and thoroughly rinsed with ultra-pure water and dried under a laminar flow bench and stored in acid-cleaned Petri dishes). Dissolution experiments to determine NO<sub>x</sub> released in surface seawater after deposition were performed on board using acid-cleaned Sartorius filtration units (volume 0.250 L) and filtered surface (5m) seawater. Each sample was subjected to two contact times: the first contact was at one minute, and the second contact was at 24 hours. NO<sub>x</sub> was analyzed using a 1-m long Liquid Waveguide Capillary Cells (LWCC) made of quartz capillary tubing, following the protocol described in Louis et al., 2015. An extrapolated NO<sub>x</sub> from dry deposition was estimated on the basis of a deposition velocity of submicronic particles (0.4 m s<sup>-1</sup>; Vong et al., 2010).”**

**Louis, J., Bressac, M., Pedrotti, M. L., & Guieu, C. (2015). Dissolved inorganic nitrogen and phosphorus dynamics in seawater following an artificial Saharan dust deposition event. *Frontiers in Marine Science*, 2, 27.**

**Vong, R. J., Vong, I. J., Vickers, D., and Covert, D. S.: Size-dependent aerosol deposition velocities during BEARPEX'07, *Atmos. Chem. Phys.*, 10, 5749-5758, doi:10.5194/acp-10-5749-2010, 2010**

p 5 Line 26: "same depths as for NPP" Were these <sup>15</sup>N<sub>2</sub> incubations performed in the same bottles as the <sup>14</sup>C incubations?

**The <sup>15</sup>N<sub>2</sub> incubations were not performed in the same bottles as the <sup>14</sup>C incubations.**



p 6 Line 4: The danger of "trace metal contamination" is mentioned. Was the water for incubations collected using trace metal clean methods?

**Indeed, we mentioned the danger of "trace metal contamination", referring to this danger while preparing the  $^{15}\text{N}_2$ -enriched seawater using the method of Mohr et al. (2010). We did not collect water using in the trace metal clean container for logistical reasons (no space for that) but all bottles, tubing etc were carefully washed with 10% HCl to keep potential contaminations at their minimum.**

p 6 Line 13: "blue screening" I assume that the purpose of blue screening was to alter the quality of incident light on the bottles. Please provide additional information on the change in quality to the incident light achieved with the blue screening. Also, was the quantity of incident light altered (i.e. by using different sizes mesh screen or some other neutral density filter) in the deckboard incubators?

**By "blue screening", we mean transparent blue sheet filters from light blue to dark blue which were fixed on the faces of the incubator and filtered the incident light from 75 to 0.1 % of the incident light, respectively to the depth of sampling. Thus, the quantity of light that reached the bottle was deliberately altered to copy the in-situ light.**

p 6 Line 16: What is meant by "gentle filtration"?

**By "gentle filtration" we mean low pressure (< 0.2 bar) filtration that does not damage and blow up the cells.**

p 6 Line 21-22: Why was the initial PN only measured at two depths rather than for each rate measurement? Has it been determined that two depths are sufficient? If so, please be specific. How exactly were these two measurements used as 'initial' measurements for each rate measurement? Were they averaged and then used for all rates at that stations or for all stations or some other method? please specify. How many replicates were collected per depth for the initial measurements?

**The natural PN  $\delta^{15}\text{N}$  did not vary within the photic layer, that is why we used the average of the 2 values for our rate calculations. In the companion paper Benavides et al. in which we measured aphotic  $\text{N}_2$  fixation, we performed  $\delta^{15}\text{N}$  measurements of PN at each depth because the values were much more variable and could influence our rate calculations, which was not the case in the photic layer.**

p 6 Line 25: How was the nitracline depth calculated? I am unclear on what  $\text{NO}_3$  is. Is this the density where the nitracline occurs?

**The top nitracline depth was graphically determined at the depth where the  $\text{NO}_3$  concentration was detectable. As there were internal waves (Bouruet-Aubertot et al., this issue) at LD A, the top nitracline depth varied daily, but the slope (nitrate concentration against density) was constant. We observed that using the nitrate vs. density profiles instead of nitrate vs. depth profiles, the top of the nitracline was always at the same iso-density. For that purpose, we decided to work with density instead of depth as is usually the case. Thus,  $\rho_{\text{NO}_3}$  corresponds to the density associated with the top of the nitracline.**

p 6 Line 25- p 7 Line 3: Is there a reference for this flux calculation?

**The flux calculation was performed using a typical Fick law where the flux is the result of the turbulent diffusion coefficient ( $K_z$ ) per the concentration/depth gradient ( $d[\text{NO}_3]/dz$ ). As we worked with density, here the concentration/depth gradient ( $d[\text{NO}_3]/dz$ ) was split in the concentration/density gradient ( $d[\text{NO}_3]/dp$ ) per the density/depth gradient ( $dp/dz$ ).**

p 7 Line 6: Why were these depths chosen?

**The first trap was deployed at 150 m as it is more or less the base of the photic layer. The two others depths were chosen for logistical reasons.**

p 7 Line 8: "...buffered solution of formaldehyde..." Please include a reference for this statement.

**The sentence: "The flasks were previously filled with a buffered solution of formaldehyde (final conc. 2 %) and were stored at 4 °C after collection until analysis to prevent degradation of the collected material." was changed by : "The flasks were previously filled with a buffered solution (Sodium borate) of formaldehyde (final conc. 2 % and pH=8) and were stored at 4 °C after collection until analysis to prevent degradation of the collected material."**

p 7 Line 12: Please specify what a "swimmer" is. Were these living organisms found in the 'fresh' trap, or were they in the preserved samples too? If the latter, how were they separated from the rest of the material? Were these a certain size class of organisms? Why were they analyzed separately?

**Swimmers are used to describe zooplankton organisms recovered in the traps (both fresh and preserved samples). The word "swimmers" is used because it is thought that mainly living zooplankton was poisoned and recovered in the traps. They were manually handpicked and analyzed separately because they cannot be considered as settling particulate matter.**

p 8 Line 6: What is meant by "gently filtered"?

**By "gentle filtration" we mean low pressure filtration (<0.2 bar) that do not damage and blow up the cells.**

p 8 Line 11-12: "The C contents...Luo et al. 2012)." This belongs in the results section.

**As this sentence is necessary to validate our method, we have included this sentence in the Methods section.**

p 8 Line 11-16: "As DDAs...asymbiotic)." It is unclear why this information is present; I thought the cell C content was only determined for Tricho and UCYN. Please add a topic sentence to this paragraph explaining for which organisms biovolume and cell C content were determined and which were determined directly v indirectly.

**The cell C content was directly determined for *Trichodesmium* and UCYN, and indirectly for DDAs. This has been specified in the new version.**

## Results

p 9 Line 2: "...with the minimum concentration located at 60 m." It's best to refrain from calling this a minimum because the difference in concentration is slight and there are no replicates.

**We agree with this comment, this part of the sentence has been deleted.**

p 9 Line 5: "...while the DCM was deepening from 25 m to 70 m..." I suggest restating this as "...the depth of the DCM increased from 25 to 70 m during the five days that the station was occupied..." or something similar

**We agree with this comment, thus we have rewritten the sentence as proposed by reviewer #2.**

p 9 Line 6: "...varying simultaneously between 115 and 155 m." This wording is confusing.

**We have changed "simultaneously" to "concurrently".**

p 9 Line 11: "...below 0.2..." Is it below 0.2 or equal to 0.2, as Table 4 indicates? Where is the comparison of total N deposition and NO<sub>3</sub>+NO<sub>2</sub> deposition, as suggested in the Methods?

**We have rewritten this section as follows:**

**"Nitrate dissolution from aerosols occurred rapidly releasing in seawater on average 1.8 nmol.m<sup>-3</sup> (26 ng m<sup>-3</sup>) dissolved inorganic nitrogen. Nitrate appeared to be as nitrate aerosol since no correlation was observed between nitrate and Fe, Si, Na, Cl (K. Desboeufs, pers. Com., 2017), precluding a mixing with ash or sea salt. Extrapolated dry deposition flux (Table 4) was on average 630 ± 329 nmol.m<sup>-2</sup>.d<sup>-1</sup> (3.22 ± 1.7 mg m<sup>-2</sup> yr<sup>-1</sup>)."**

p 9 Line 13: "...0.0-19.3..." What was the detection limit of this method? Instead of reporting "0.0" please report rates as below the limit of detection if they are. Two recent publications depict methods for calculating the detection limit of these rates (Gradoville et al. 2017; Jayakumar et al. 2017). Accounting for a DL will be especially important for LDC where the rates were very low and may be undetectable.

**The minimum quantifiable rate calculated using standard propagation of errors via the observed variability between replicate samples, measured according to Gradoville et al. (2017), was 0.035 nmol N L<sup>-1</sup> d<sup>-1</sup>. We have indicated <QL instead of "0.0" in the new version.**

p 9 Line 17: "...below the quantification limit..." So there is a quantification limit! How was it calculated and what was it?

**The quantification limit has been added in the new version. See previous response.**

p 9 Line 17-19: Please compare the N<sub>2</sub> fixation rates from days 1-3 using the Mann Whitney test, as was done comparing the in situ and shipboard incubations.

**This comparison has been performed, and the rates from day 1 and day 3 were not statistically different. This comparison has been performed for all daily rates at each station, and no statistical difference was found.**

p 9 Line 23: "...the maximum at 40 m at LDC..." Was this small rate actually above the detection limit though?

**The maximum of 2.6 nmol N L<sup>-1</sup> d<sup>-1</sup> measured at 40 m at LD C was above the detection limit.**

p 9 Line 24-27: It is nice to see this comparison! It makes me feel better about all the shipboard measurements in the literature!

**We appreciate the comment**

p 9 Line 31-p10 Line 1: "Strong time...(table 1)." This is repetitive with the previous sentence.

**This sentence has been deleted and merged with the previous sentence. "The averaged NO<sub>3</sub><sup>-</sup> input through vertical turbulent diffusion showed strong time variability with a typical standard deviation of the same order as the mean value (Table 1), and a strong contrast between the western station LD A and the two other stations, with mean values equal to 24.4 ± 24.4 μmol N m<sup>-2</sup> d<sup>-1</sup> at LD A and 6.7 ± 5.3 and 4.8 ± 2.2 μmol N m<sup>-2</sup> d<sup>-1</sup> at LD B and LD C, respectively (Fig. 4)."**

p 10 Line 22: "...maximum export...at LD C." PC export for LDB and LDC do not look significantly different, based on the overlap in standard deviation (Table 2).

**The sentence has been rewritten in the new version: "...maximum export at 150 m at LD A, minimum export at LD B and LD C..."**

p 10 Line 21-24: Why is there a discussion of trends in PC but not PN and PP? Line 24: "...averaged C:N ratios...LDC." Are these significantly different?

**In this study, we decided to focus on the C and N export, associated with N<sub>2</sub> fixation. As POP was also measured, we include it in the Results but not in the Discussion part.**

p 10 Line 31: "below detection limit" What was the detection limit? Elsewhere in the paper, (i.e. for NO<sub>3</sub> and PO<sub>4</sub> concentrations), a "quantification limit" is referenced. What is the difference between that quantification limit and a detection limit? Please define these terms in the Methods section and be consistent with their usage.

**The limit here is a quantification limit, as for NO<sub>3</sub><sup>-</sup> and PO<sub>4</sub><sup>3-</sup> concentrations and N<sub>2</sub> fixation measurements. The quantification limit was referenced in the Methods section. All the terms have been corrected in the new version.**

## **Discussion**

p 11 Line 27: "...atmospheric deposition...lower end of fluxes..." I am curious what the mixed layer depth was at these stations, as atmospheric deposition is really a flux only to the mixed layer, not the entire euphotic zone. Since the flux is so small, it likely doesn't matter for this study, but it would be nice to see those depths in a Table for readers who study atmospheric deposition to the mixed layer.

**We focused on the photic layer, and on what comes from above and below the photic layer. We presented integrated rates (per m<sup>2</sup>) and considered the whole photic layer, and the relatively low atmospheric deposition compared to dinitrogen fixation rates.**

p 12 Line 8-17: "In this study...to significant bias." This is a really good point, and it's nice to see a snapshot of this variability. It does make one wonder, however, if the snapshot had been even longer, how would the results have varied, given the large differences seen between the use of an average  $K_z$  and the instantaneous  $K_z$  values and the seeming randomness of the spikes. Is 5 days long enough? A sentence or two addressing this concern would be useful.

**The differences in estimated fluxes between the stations is mainly attributable to differences in  $K_z$  between the stations. The gradients of concentration varied little between stations (table 1). The turbulent diffusion contrast is a robust result despite the variability of the signal ( $K_z$  and therefore the flux). This is explained by the fact that shear instability, the dominant process triggering turbulence at long stations, is more intense in LDA because of higher shear, by a factor of about 3. Due to this, we suppose that if this had been studied longer than 5 days, the results would have been in the same order of magnitude. This point has been studied in depth in Bouruet-Aubertot et al. (this issue), that we refer to: "The contrasted  $\text{NO}_3^-$  input observed at the three stations results from the high variability in turbulence along the west-east transects (Bouruet-Aubertot et al., this issue)."**

p 12 Line 22: "N m<sup>-4</sup>" What is a m<sup>-4</sup>?

**The unit of the gradient is unit of concentration divided per unit of depth, expressed in  $[\mu\text{mol N m}^{-3}]/[\text{m}]$  and simplified as  $\mu\text{mol N m}^{-4}$**

p 15 Line 11: "such as DON and/or zooplankton export" Please elaborate. Dissolved compounds are not exported like particles are; they do not sink. Or is this a reference to conversion of fixed N to the DON pool, followed up uptake and subsequent export? Both a direct DON flux and a delayed N flux following DON uptake are worth mentioning. Also, what about fixed N that is released as ammonium? Ammonium and many simple DON compounds (amino acids, urea...) cycle very quickly and likely would be taken up before they could be mixed downward. Also, it is unclear why DON and zooplankton export are lumped together like this... Is this referring to an active zooplankton flux (which is a completely different process than the DON pathway) or to the sinking out of dead zooplankton later?

**In this sentence, we assessed the hypothesis that could explain the imbalance between new and exported N. We presented 3 hypotheses, and the third one is "processes other than particle export, such as DON and/or zooplankton export". Those two come within the same hypothesis because they refer to an "export", but yes, they are not exported via the same process. They are lumped together in this sentence, but below in the section, DON and zooplankton export are discussed separately.**

**We are aware that DON is not exported as particles and does not sink. The export of DON (whatever the source of DON: i.e. phytoplankton or zooplankton) by turbulent diffusion was estimated by Moutin et al. (this issue) and the important result is that it cannot explain the imbalance. As reviewer #2 refers to the conversion of fixed N to the DON pool, note that Caffin et al. (this issue) have studied the transfer of the fixed N to the dissolved pool ( $\text{NH}_4$  and DON) and to plankton communities. This N transferred to the dissolved pool, was re-uptaken and subsequently exported as reviewer #2 mentioned, and finally recovered in the sediment traps particulate matter.**

p 16 Line 1: "zooplankton itself is" should be "zooplankton themselves are" Please make similar corrections throughout.

**This has been corrected in the new version.**

## Conclusion

p 16 Line 16-17: "contributed to ~15-21%" and "...and to ~3%..." Delete "to".

**This has been corrected in the new version**

## Tables and Figures

The tables seem to be out of the order that they are mentioned in the text. Please check this.

**This has been corrected in the new version**

Table 1. Please define K<sub>z</sub> in the figure legend. For this and all figures and tables, please indicate what the error is. Standard deviation? Standard error? n = ?

**This has been corrected in the new version**

Table 2. What is "DW<sub>matter</sub>"? Please indicate this in the table legend or with a footnote to the table. Why are the errors italicized in this table and Table 3 but not in Table 1? Please standardize this across tables. If PC, PN, and PP should be PC flux, PN flux, and PP flux, please indicate that. Please propagate the error of PC, N, and P into the C:N:P ratio calculation.

**We have specified dry weight (DW) in the table legend. The errors have been standardized across tables in regular font. We have indicated that PC, PN, and PP are PC flux, PN flux, and PP flux in the table. The C:N:P ratio were presented without propagation error to keep the table and the message clear and to give a global overview of the ratio at each depth. We allowed ourselves to do that as the standard deviation was presented in each of the previous terms.**

Table 3. Please apply the comments from Tables 1 and 2 to this table. Also, please use a standard number of significant digits for all measurements (i.e. LD A 300 m is inconsistent).

**This has been corrected in the new version**

Table 4: Please include the standard deviation or error of these values by propagating the error from the measurements. please define all non-obvious terms (i.e.  $d[NO_3]/dp$ ,  $NO_3$ ) and the calculation for the e-ratio in a footnote so that the reader does not have to dig through the text to find them. Is N<sub>2</sub> fixation the in situ rate or the shipboard rate? Please use consistent significant digits (i.e. for  $NO_3$  diffusion and export N 150 m).

**The objective of this table is to show a global overview of the budget and the different fluxes, that is why we do not include standard deviations. All the standard deviations are given in the text or/and in other tables. We have deleted the term  $d[NO_3]/dp$  from the table as it is not a useful information. Here, we present in situ rates.**

Fig 1: This figure seems needlessly complicated and the vertical component does not seem spaceworthy. The advantage of the vertical component is to show where the production arrays and



traps were deployed, but it's difficult to tell the exact depths in this figure. A simple map may be preferable.

**The purpose of this figure is to give a global map of the surface chl a, show the station position and show where the production arrays and traps were deployed, in a same picture. We understand that it's difficult to tell the exact depths, that is why we have added the exact depth in the legend. We have decided to keep this figure in this version.**

Fig. 2: Please indicate on the figure itself which units correspond to which parameter, as it is a bit confusing in current form (particularly for phosphate). It may be instructive to use a different scale for the N fixation rates in the third panel, since they can't be seen on this scale.

**This has been corrected in the new version**

Fig 3: This is a cool figure! Nice dataset! I am a little confused on how 1% of surface PAR was calculated at night. Should there be breaks in the dotted line for nighttime?

**The 1 % of PAR was calculated only at midday (yellow crosses) and then linked to the previous and following day by the yellow dotted line. Even during the night 1 % of PAR is present at depth.**