Responses to Review #1

We thank Reviewer #1 for taking the time to read the submitted manuscript and for his/her comments. Below is the original response from Reviewer 1 (in italic/bold), with our own responses interspersed within. The original text of the manuscript is written in blue typefaceand the changes to the text are shown in red.

1. ***Fumenia and coauthors have presented biogeochemical data from the OUTPACE expedition that took place in the western tropical South Pacific Ocean. Presented data are important but it lacks a robust analysis. There are too many speculative statements to list them all. I try to list some of main points below***

Response: We thank Reviewer#1 for mentioning the important dataset presented in our manuscript. Reviewer#1 emphasizes the lack of a robust analysis. The dataset presented in our manuscript is supported by a robust chemical analysis. Only the chemical data (NO_3^-, PO_4^{3-}) above the quantification limit (QL = 0.05 µM) were taken into account to calculate the derived ratio (N*). The same remark is valid for the chemical data, including PON, POP and the derived ratio PON:POP. Furthermore, high analytical repeatability was found for NO_3^-, PO_4^{3-} and for organic particulate matter (see tables R4 below). Therefore, the increases in both PON, PON:POP and N* observed in the Melanesian Archipelago surface waters are supported by robust chemical analysis. Concerning data extracted from the GLODAPv2 database, only data which have passed the two-quality controls, as recommended by Olsen et al. (2016), have been used in this study. Furthermore, a statistical analysis (t-test and Pearson test) has been added in the revised text of the manuscript (see the details below in the answer to comment 3).

General comment: The title of our paper has been rephrased.

Modified text:
The effect of N_2 fixation on the thermocline waters of the South Pacific Ocean traced by excess nitrogen

2. *Authors have tried to interpret their biogeochemical data (such as N*) based on community structure (such as lines 385-388 on page 10, "In the SW, the N* variations. . .. .). But the authors present no record of biology data, so all such statements (including the ones are PON:POP stoichiometry) remain speculative!*

3. *The manuscript seems like more or less a data reporting manuscript with no robust statistical analysis.*

Original text (lines 385-388): "If in the SW Pacific, the N* variations are solely due to diazotrophic activity, these results suggest that the decrease in PO_4^{3-} and the associated increase in N* in the MA waters are controlled by diazotrophic organisms. The high concentrations of PO_4^{3-} in the SW of the GY suggest a low use by diazotrophs, as N_2 fixation is hypothesized to be limited by dissolved iron (DFe) availability in GY waters (Moutin et al., 2008; 2018; Blain et al., 2008; Guieu et al., 2018) maintaining a low N* anomaly in this region.

Response: We apologize for this confusing section (4.1.1). The whole of the paragraph above has been deleted from the revised manuscript. In the revised text, the Discussion has been rewritten in a clearer and more concise way. The Discussion has been reviewed, and awkward and ambiguous sentences corrected. Section 4.1.1 has been deleted, and we have focused the Discussion on the effect of intense N_2 fixation on the particulate organic N pools, and on the N* distribution, first in the WTSP, and then in the thermocline waters at the scale of the South Pacific Ocean.

But we disagree with the reviewer concerning the speculative statements of our study. Our results show significant N_2 fixation rates in the surface waters and at the top of the upper thermocline waters of the Melanesian Archipelago. In agreement with the high diazotrophic activity observed in the region, relatively high PON concentrations (0.32-1.18 µmol kg^{-1}), an excess of PON concentration relative to that expected from Redfield stoichiometry, and positive N* in the surface waters and in the upper and lower thermocline waters of the Melanesian Archipelago were observed. In contrast, the surface waters and the top of the upper thermocline waters of the South Pacific Gyre (170° W-160° W) were characterized by low N_2 fixation rates and low PON concentrations.

As requested by Reviewer#1, a statistical analysis (Pearson test) has been added. We showed a significant relationship between N_2 fixation rates and PON in excess (r²=0.86, p<0.001), and between N_2 fixation rates and N* values (r²=0.60, p<0.001). Our results clearly show that the intense N_2 fixation observed in the WTSP significantly influences the dissolved inorganic (N excess) and particulate organic (PON excess) N pools of the surface and thermocline waters in the WTSP. Following an analysis of South Pacific nutrients database (GLODAPv2), we find a strong positive N* signature in the thermocline waters of the WTSP, the Coral/Tasman Seas, and the southern part of the subtropical gyre between 23° S and 32° S. Together with a basin-scale analysis of water mass circulation, we

hypothesize that the outstandingly high N_2 fixation rates observed at the surface influence the N status of thermocline waters across the whole South Pacific Ocean.

Several additional statistical tests have been added to provide a robust analysis of our data, as requested by Reviewer#1. The equality of mean concentrations in the Melanesian Archipelago and in the gyre was tested as follows. Firstly, a Fisher-Snedecor test was used to establish the equality of variances. Secondly, a Student's test (level significance $p = 0.01$) was used when variances were equal and Satterthwaite's approximate t-test was performed otherwise. The results are presented in the supplementary material for surface waters, the UTW and the LTW (see tables R1, R2 and R3 below).

Table R1. Results of Student's test or Satterthwaite's approximate t test (marked by a red asterisk) in the surface waters of the Melanesian Archipelago (MA) and of the gyre (GY); t is the t value, DF is the degree of freedom, p is the p-value. The results marked by a black asterisk have a p-value < 0.01, n is the number of samples for the MA and the GY areas.

		Surface Waters			
		n	t	DF	p < 0.01
N_2 fixation	MA/GY	57/10	9.16*	60.75	*
PO_4^{3-}	MA/GY	19/20	7.18*	24.97	*
N^*_{surf}	MA/GY	20/21	7.88*	28.24	*
PON	MA/GY	77/20	15.03*	92.31	*
POP	MA/GY	77/20	15.49*	80.53	*
PON : POP	MA/GY	77/20	5.08	95.00	*
NOPexcess	MA/GY	77/20	8.22*	94.90	*

Table R2. Results of Student's test in the UTW of the Melanesian Archipelago (MA) and of the gyre (GY), t is the t value, DF is the degree of freedom, p is the p-value. The results marked by a black asterisk have a p-value < 0.01, n is the number of samples for the MA and the GY areas.

		UTW			
		n	t	DF	p < 0.01
N_2 fixation	MA/GY	24/7	0.70	29	
NO3-	MA/GY	70/18	1.24	86	
PO_4^{3-}	MA/GY	85/25	1.49	86	
N :P	MA/GY	70/17	2.60	85	*
N*	MA/GY	70/17	8.95	85	*
PON	MA/GY	97/25	2.48	120	*
POP	MA/GY	97/25	2.40	120	*
PON : POP	MA/GY	97/25	0.39	120	

Table R3. Results of Student's test in the LTW of the Melanesian Archipelago (MA) and of the gyre (GY), t is the t value, DF is the degree of freedom, p is the p-value. The results marked by a black asterisk have a p-value < 0.01, n is the number of samples for the MA and the GY areas.

		LTW			
		n	t	DF	p < 0.01
NO3-	MA/GY	38/9	0.76	45	
PO_4^{3-}	MA/GY	38/9	1.39	45	
N :P	MA/GY	38/9	2.64	45	*
N*	MA/GY	38/9	5.72	45	*
PON	MA/GY	39/6	1.96	43	
POP	MA/GY	39/7	1.10	44	
PON : POP	MA/GY	39/6	1.42	43	

4. Reviewer: All the figures and tables list some data with repetition but no rigorous analysis.

Response:

We agree with Reviewer#2 regarding the excessive repetition and excessive details provided in the manuscript. In the revised paper, the Results section has been rewritten in a clearer and more concise way. Furthermore, the manuscript has been shortened and additional statistical tests were taken into account in the interpretation of our results. In addition, the aims of the study have also been clarified and new scientific knowledge highlighted.

General comment: We propose a new version of the results section (3.1, 3.2 and 3.3) in the revised manuscript.

Modified text:

3.1 Water masses and general biogeochemical trends

Figure 2a shows Θ-S diagrams (0-2000 dbar) for all 18 stations sampled during the OUTPACE cruise. The temperature range observed on the OUTPACE section varied from 29.9 °C at the surface to 2.0 °C at 2000 dbar depth (Fig. 2a). Salinity varied from 34.3 to 36.1 (Fig. 2a). The MLD ranged between σ21.9 and σ22.6, with an average value of 22.2 ± 0.2 kg m^{-3} (corresponding to 19 ± 9 dbar), (Fig. 2b). The temperature and salinity observed in the surface waters were 29.4 ± 0.4 °C and 35.1 ± 0.2, respectively (Fig. 2a). A salinity maximum (S_{max}) was observed in the UTW between σ24 and σ25.4, centered on σ24.7 (Fig. 2a). Between σ25.4 and σ26.7 (249-501 dbar, Table 3), a strong decreasing gradient of temperature and salinity marked the presence of the LTW.

In the surface waters of the Melanesian Archipelago, N_2 fixation rates ranged from 2.4 to 42.2 nmol N L^{-1} d^{-1} (average rate 15.4 ± 10.4 nmol N L^{-1} d^{-1}; Fig.3, Table 1). Rates decreased drastically towards the

gyre waters, with values ranging between <0.01 to 2.3 nmol N L^{-1} d^{-1} (Fig.3, Table 1). N_2 fixation rates showed high values at the top of the UTW above $\sigma23.5$ in the Melanesian Archipelago, and homogeneous low values between $\sigma23.5$ and $\sigma25.4$ (Fig.3). Conversely, the N_2 fixation rates were very low in the UTW of the gyre waters (Fig.3; Table 2). Below $\sigma25.4$, the N_2 fixation rates were undetectable (Fig.3; Table 2).

3.2 Inorganic nutrient pools

The concentrations of NO_3^- were below the quantification limit in the surface waters for all sampling stations (Fig. 4a). The concentrations of PO_4^{3-} were below the quantification limit or low in the surface waters of the Melanesian Archipelago, with a mean value (for measurable samples) of 0.07 ± 0.01 µmol L^{-1} (Fig. 4b, Table 1). Significant higher PO_4^{3-} concentrations ($p < 0.01$, Table S3) were observed in the surface waters of the gyre, with a mean value of 0.15 ± 0.01 µmol L^{-1} (Fig. 4b, Table 1). The NO_3^- concentrations became quantifiable below $\sigma23.5$. The NO_3^- and PO_4^{3-} concentrations increased gradually in the upper and lower thermocline waters. The bottom waters (> $\sigma27.5$, 1000 - 2000 dbar, 2000 dbar corresponding to the deeper depth sampled) had relatively homogeneous maximum concentrations, with values ranging between 35.1 and 36.5 µmol L^{-1} and between 2.40 and 2.55 µmol L^{-1} for NO_3^- and PO_4^{3-}, respectively (Fig. 4a; Fig.4b).

The NO_3^-: PO_4^{3-} ratio increased sharply in the UTW, and was significantly higher ($p < 0.01$, Table S4) in the Melanesian Archipelago than in the gyre (Fig. 4c, Table 2). In the LTW, the NO_3^-: PO_4^{3-} ratio increased slightly and was significantly higher ($p < 0.01$, Table S5) in the Melanesian Archipelago than in the gyre (Fig. 4c, Table 3). In deep waters (below 27.5), the NO_3^-: PO_4^{3-} ratio reached stable and uniform values, ranging between 13.8 and 15.2 (Fig. 4c). During the OUTPACE cruise, the distribution of NO_3^- *versus* PO_4^{3-} concentrations followed a linear regression ($r^2 = 0.99$) with a slope of 15.0, which was close to the Redfield ratio of 16:1 (Fig. 4d). In the upper waters, NO_3^- concentrations become depleted before those of PO_4^{3-}. In this layer, a deficit in NO_3^- compared to the value expected from a Redfield point of view (dashed black line) was observed for all data (Fig. 4d).

3.3 Organic pools

The concentrations of PON and POP showed maximum values in the surface waters of the Melanesian Archipelago, with values ranging between 0.32 and 1.18 µmol L^{-1}, and between 0.02 and 0.06 µmol L^{-1}, respectively. They were significantly higher ($p < 0.01$, Table S3) in the Melanesian Archipelago than in the gyre (Fig. 5a, Fig. 5b; Table 1). In the UTW, a significant attenuation of POM concentrations was observed. The mean concentrations of PON and POP remained, however, significantly higher ($p < 0.01$, Table S4) in the UTW of the Melanesian Archipelago than those of the gyre (Fig. 5a, Fig. 5b, Table 2).

POM concentrations decreased to reach minimum and relatively constant values in the LTW. A significant relationship (p<0.001; Pearson test) was obtained between PON concentrations and N_2 fixation (r^2=0.56). The PON concentrations in excess showed a contrasted distribution in the surface waters of the studied areas. Indeed, in the Melanesian Archipelago, the highest values were observed between the surface and the top of the UTW (< σ22.5, ≈ 0-25 dbar), with a maximum of 0.37 µmol L^{-1} (Fig. 5c). In the gyre, the PON concentrations in excess exhibited values around zero. Below σ23.5, the PON concentrations in excess showed a similar distribution in both areas studied, with values close to those calculated based on Redfield (Fig. 5c). A significant relationship (p<0.001; Pearson test) was obtained between PON concentrations in excess and N_2 fixation (r^2=0.86). For all the sampling stations, the PON:POP ratio was between 10.08 and 25.78 (figure not shown). The average value of the PON:POP ratio was 16.9 ± 2.4, which included the Redfield ratio (16.0). Nevertheless, a strong variability was observed between the different oceanic regions (Melanesian Archipelago *vs* gyre) and the different water masses considered. In the surface waters, the PON:POP ratio was significantly higher (p < 0.01, Table S3) in the Melanesian Archipelago than in the gyre (Table 1). In the UTW, the PON:POP ratio was not significantly different (Table S4) between the Melanesian Archipelago and the gyre. The distribution of PON *versus* POP concentrations followed a linear regression in both studied areas. In the gyre, a slope of 15.1 was observed, which was close to the Redfield ratio (blue line in Fig. 4d). In the Melanesian Archipelago, the slope observed deviates from the expected Redfield ratio, showing a value of 20.0 (red line in Fig. 5d).

5. *For example, Figures 10, 11 and 12 are very similar to each other and it is not clear what the authors are trying to convey. What is the main hypothesis that the authors are trying to address?*

Response: Figures 10, 11 and 12 of the original manuscript presented the N*, but they do not all have the same objective. In figure 10, we showed the N* observed in the water column of the Melanesian Archipelago during the cruise. Figure 11 indicated both the integrated N_2 fixation rates and the N* variations on two isopycnals (24.7 and 26.3), which are characteristics of the upper and lower thermocline waters, respectively. The aim of figure 11 is to show the exact location of the N* increase in the thermocline. We found a strong increase of N* in the thermocline waters of the Melanesian Archipelago, an area strongly subject to N_2 fixation in its surface layer. To visualize the input of N by the high N_2 fixation observed in the Melanesian Archipelago, we used the quality dataset (GLODAPv2; http://cdiac.ornl.gov/oceans/GLODAPv2/). Figure 12 presented the large-scale ocean transport of thermocline waters, obtained from the literature. The analyses of the most recent available nutrient data in the South Pacific Ocean enabled us to show a high positive N* in the thermocline waters of the whole South Pacific gyre. We consider that a major result is our hypothesis that the local high N* observed in

the Melanesian Archipelago is transported eastward *via* the circulation, and consequently that N_2 fixation in the WTSP may influence the nutrient distribution at the scale of the South Pacific basin.

General comment: In the revised manuscript, the Results sections (3.4 and 3.5) have been rewritten in a clearer and more concise way. We propose a new version in the revised manuscript.

Modified text:

3.4 Regional distribution of N* and N_2 fixation rates

At the scale of the OUTPACE cruise, N* showed positive values ranging between 0.23 and 2.15 µmol L^{-1} in the surface waters. N* was significantly higher ($p < 0.01$, Table S3) in the Melanesian Archipelago than in the gyre (Fig. 6). In the UTW and the LTW (100-500 dbar), N* showed a significant difference ($p < 0.01$, Table S4 and S5) between the Melanesian Archipelago and the gyre waters. In the UTW, N* mean was positive in the Melanesian Archipelago and negative in the gyre (Fig. 6b, Table 2). In the LTW, below 150 dbar depth, N* exhibited a marked decrease, with a minimum value of -1.55 µmol L^{-1} at 400 dbar in the gyre. In the same layer, N* was significantly higher ($p < 0.01$, Table S5) in the Melanesian Archipelago than in the gyre (Fig. 6b, Table 3), with a minimum value of -0.77 µmol L^{-1} at 500 dbar (Fig.6a). Below 500 dbar, N* variability decreased with depth and reached mostly negative values at 2000 dbar for all sampling stations (Fig. 6a).

Regarding the horizontal variability of N*, over the upper (σ24.7) and lower (σ26.3) thermocline layers (Fig. 7), N* showed higher values in the Melanesian Archipelago than in the gyre. In the Melanesian Archipelago (between SD1 and SD12), N* ranged between 1.05 ± 0.01 and 1.77 ± 0.02 µmol kg^{-1} and between -0.02 ± 0.02 and 1.21 ± 0.02 µmol kg^{-1} in the upper and lower thermocline layers, respectively. Beyond SD 12, a drastic decrease of N* was observed in both upper and lower thermocline layers (between SD12 and SD15), reaching minimum values of -0.68 ± 0.07 µmol kg^{-1} and -1.46 ± 0.02 µmol kg^{-1} in the upper and lower thermocline layers, respectively (Fig. 7).

In this study, a significant relationship ($p < 0.001$; Pearson test) was obtained between N* and N_2 fixation (r^2=0.59). Vertically integrated N_2 fixation rates in the surface waters exhibited the same trend as N*, with higher values in the Melanesian Archipelago than in the gyre. In the Melanesian Archipelago, N_2 fixation rates ranged between 100 and 840 µmol N m^{-2} d^{-1}, with an average integrated N_2 fixation rate of 443 ± 204 µmol N m^{-2} d^{-1} (red circle in Fig. 7). As for N*, a sharp decrease in integrated N_2 fixation rates was observed in the gyre, with values ranging between 5 and 73 µmol N m^{-2} d^{-1} and with an average value of 28 ± 39 µmol N m^{-2} d^{-1} (blue circle in Fig. 7).

3.5 Large scale distribution of N* in the South Pacific

At the basin scale of the South Pacific, N* was minimum in the upper thermocline (σ24.7) between 5° S and 20° S, and between 80° W and 100° W (P21, P18, P19 sections in Fig. 8a). Between 5° S and 15° S, N* showed a westward increase along the northern branch of the SEC. In the WTSP, N* increased and exhibited maximum values at 155° E, with an average value of -0.27 ± 0.97 µmol kg^{-1} (P11S section in Fig. 8a). South of 15° S, N* increased more rapidly than north of 15° S. In the WTSP, between 175° E and 150 °E, N* increased sharply during the westward transport of waters by the NCJ (Fig. 1b), reaching a value of 0.39 ± 0.58 µmol kg^{-1} (P21 section in Fig. 8a). The maximum of N* was observed in the Coral and Tasman seas, between 23° S and 30° S, reaching a value of 1.86 ± 0.19 µmol kg^{-1} (P15 section in Fig. 8a).

As observed in the upper thermocline, N* showed minimum values in the lower thermocline around the OMZ region, between 80° W and 90° W and at 18° S (P21, GT13 sections in Fig. 8b, Fig. S2), and increased westward. The maximum values of N* were observed in the Melanesian Archipelago waters, in the southern Coral and Tasman seas, exhibiting an average value of 0.52 ± 0.12 µmol kg^{-1} (P14C section in Fig. 8b, Fig. S2). In the Tasman Sea and north of New Zealand, at 30° S, and between 150 °E and 180 °E, N* showed high values, with an average of 0.66 ± 0.46 µmol kg^{-1} (P06 section in Fig. 8b, Fig. S2). These high N* values persist in the central part of the South Pacific, between 23° S and 32° S and 175° E and 135° W, with an average value of 0.42 ± 0.31 µmol kg^{-1}. In the eastern part of the South Pacific, at 110° W and between 85° W and 90° W (P18, P19 sections in Fig. 8b, Fig. S2), a drastic decrease of N* was observed. At 32° S and close to the South America coast, N* dropped sharply to reach an average value of -17.33 ± 2.63 µmol kg^{-1} (P06 section in Fig. 8b, Fig. S2).

6. *Reviewer: Line 21 suggests positive N* while line 24 suggests N* to be zero. I presume these must be from different regions. Please specify.*

Original text (lines 21-22): A positive N* anomaly was observed in the productive layer reflecting the combined effect of phosphate uptake by diazotrophic organisms and remineralization of excess particulate organic nitrogen. The South Pacific Gyre waters between 170° W and 160° W were depleted in nitrate but rich in phosphate. Surface waters exhibited very low dinitrogen fixation rates, an absence of excess particulate organic nitrogen and a N* signal close to zero.

Response: The positive N* anomaly observed in the productive layer described in this sentence refers to the Melanesian Archipelago (160° E- 170° W) (red marker on Figure 2b of the original manuscript for the positions of station; red markers on Figure10b of the original manuscript for the values of positive N* anomaly). The N* signal close to zero refers to the South Pacific Gyre surface waters (blue markers,

Figure 10b for the values of N* signal close to zero). To avoid any confusion, the sentence has been rephrased in the revised version of the manuscript, and we propose a new version of the abstract in the revised version. All mistakes have been corrected, and the paragraph was rephrased as follows:

Modified text:

Abstract. As part of the Oligotrophy to UlTra-oligotrophy PACific Experiment (OUTPACE), which took place in the Western Tropical South Pacific (WTSP) during the austral summer (March-April 2015) between the Melanesian Archipelago and the South Pacific gyre, we present the effects of intense dinitrogen (N_2) fixation on the dissolved inorganic nitrogen (NO_3^-) and particulate organic nitrogen (PON) pools. The stoichiometric nitrogen-to-phosphorus (N:P) ratio of the inorganic and particulate organic material, and the distribution of the geochemical tracer N* are described. N* has been previously used to trace changes in the proportion of nitrate (NO_3^-) relative to phosphate (PO_4^{3-}) associated with denitrification and/or diazotrophy in the open ocean. Our results show that the surface waters of the Melanesian Archipelago between 160° E and 170° W are characterized by significant N_2 fixation rates and relatively high PON concentrations (0.32-1.18 µmol kg^{-1}). In the surface and upper thermocline waters PON was present in excess (PON excess) relative to that expected from Redfield stoichiometry. In agreement with the high diazotrophic activity observed in the region, positive N* values (N excess) were observed in the Melanesian Archipelago, probably reflecting the combined effect of PO_4^{3-} uptake by diazotrophic organisms and the remineralization of organic matter enriched in nitrogen (N). PON excess and N* were significantly correlated to N_2 fixation rates, probably reflecting the supply of N to the inorganic and organic N pools *via* N_2 fixation. The excess N observed in the thermocline waters of the WTSP is probably related to the sedimentation of N-rich matter produced at the surface by intense N_2 fixation activity. Following an analysis of the South Pacific nutrients database (Global Ocean Data Analysis Project version 2), we find a strong positive N* signature in the thermocline waters of the WTSP, the Coral/Tasman Seas, and the southern part of the subtropical gyre between 23 and 32° S. Together with a basin-scale analysis of water mass circulation, we hypothesize that the outstandingly high N_2 fixation rates observed at the surface influence the N status of thermocline waters across the whole South Pacific Ocean.

7. *Reviewer: Line 53: But nothing can be better than in situ measurements! Criticism of in situ measurements should be with caution and proper justification.*

Original text (lines 53): The spatial and temporal variability of the ocean nitrogen budget remains difficult to quantify from in situ measurements.

Response: We entirely agree with Reviewer#1. Our sentence is not a criticism of *in situ* measurements, but rather refers to the lack of *in situ* measurements in the WTSP. Only a few oceanographic cruises have been undertaken in this specific area (P21-1994, P21-2009 and OUTPACE cruise, Figure 3 of the original manuscript). Nevertheless, we recognize that the sentence may cause confusion, and it has been deleted in the revised manuscript.

8. **Reviewer: Line 63: Let the equation come first before it is to be referred.**

Original text (lines 63): Despite the apparent simplicity of the proposed formula (Eq. 3), there is nonetheless a certain complexity, especially in the deconvolution of the different processes influencing the intensity of this tracer.

Response: Our thanks to Reviewer#2 for this comment. The equation has been added inline in the text.

Modified text: Michaels et al. (1996), Gruber and Sarmiento (1997), and Deutsch et al. (2001) proposed the N^* ($N^* = NO_3^- - 16 \times PO_4^{3-} + 2.90$) as a tracer to determine changes in the stock of dissolved inorganic N as deviations from the Redfield ratio resulting from the combined effect of denitrification and/or of diazotrophy.

9. **Reviewer: Line 83: area should be replaced with areal**

Original text (lines 83): with an average aera rates of 570 µmol m^{-2} d^{-1} (Bonnet et al., 2017)

Response: We agree with Reviewer#2. We have corrected it in the revised manuscript.

10. **Reviewer: Line 114: Addition of HgCl2 might degrade Organic matter and one my overestimate nutrients. Convention is just to freeze the water for nutrient analysis. Why was HgCl2 added despite that samples were frozen?**

Response: The samples used for inorganic nutrient measurements were not frozen but poisoned with HgCl$_2$. This is a choice we have opted for in the laboratory for many years whenever samples must be transported over thousands of miles and for several months. Reviewer#1 is right in saying that current convention is just to freeze, but it has not always been the case, and alternative sampling preservation methods is still a subject of research (Wong et al., 2017). We have ourselves also undertaken a lot of tests in order to check our preservation procedure. Degradation of organic matter was never shown to significantly influence our measurements in our relatively low organic matter (oligotrophic) areas.

Please note that we used pre-filtered samples to measure nutrients and that we get relatively high quantification limits for nutrients (0.05 µM).

11. Reviewer: Line 140: Provide lot no for the 15N2 gas as these have been subject to contamination recently.

Response: We are aware that Dabundo et al. (2014) reported potential contamination of some commercial $^{15}N_2$ gas stocks with ^{15}N- enriched NH_4^+, NO_3^- and/or NO_2^-, and nitrous oxide (N_2O). Nonetheless, that does not significantly affect our results, as explained by Caffin et al. (2018) in another paper from the OUTPACE special issue. In their study, Dabundo et al. (2014) analyzed various brands of $^{15}N_2$ (Sigma, Cambridge Isotopes, Campro Scientific), and found that the Cambridge Isotopes brand (i.e., the one used in this study) contained low concentrations of ^{15}N contaminants. By using this contamination level, the potential overestimated N_2 fixation rates modeled would range from undetectable to 0.02 nmol N L^{-1} d^{-1}. The N_2 fixation rates measured in our study were on average ~10 nmol N $L^{-1}d^{-1}$, suggesting that the stock contamination would be too low to affect the results reported here. To verify this, the Cambridge Isotopes batches that are routinely used by our team have been analyzed for potential contamination in Julie Granger and Richard Dabundo's lab. The results confirm that the contamination of the $^{15}N_2$ gas stock was low: 1.4 x 10^{-8} mol of $^{15}NO_3^-$ per mol of $^{15}N_2$, and $1.1x10^{-8}$ mol NH_4^+ per mol of $^{15}N_2$. The application of this contamination level to our samples using the model described in Dabundo et al. (2014) indicates that our rates could only be overestimated by 0.01 to 0.12 %. We thus confirm that the stock contamination issue did not affect the results reported here.

12. Reviewer: Why 2.90 was not added in equation 2 but in 3. Is PONexcess zero without 2.90.

Response: "PON excess" was calculated in order to show whether a N excess exists in the organic matter when compared to the classical composition of the organic matter, defined by the Redfield ratio (PON:POP = 16:1). There is no reason to add a 2.90 in this equation.

13. Reasons should be provided why is it necessary to make it zero. I believe the equation was originally given by (Gruber and Sarmiento, 1997; Michaels et al., 1996) and not by Deutsch et al (2001) as authors seem to suggest (lines 150 and 191)

Original text (lines 59-61) : Michaels et al. (1996) and Gruber and Sarmiento (1997) proposed the N* as a tracer to determine the proportion of changes in the fixed nitrogen stock (all nitrogenous forms except atmospheric N_2) resulting from the combined effect of denitrification and/or diazotrophy.

Response: We agree with Reviewer#1; the N* definition was indeed introduced first by Michaels et al. (1996). These authors first defined the N* parameter as the concentration of nitrogen in excess (or deficit) compared to that expected from remineralization of phosphate at Redfield stoichiometry. The first equation was:

$$N^* = NO_3^- - 16 \times PO_4^{3-} + 2.72$$

Thereafter, Gruber and Sarmiento (1997) have used the N* and modified it to investigate the distribution of nitrogen fixation and denitrification in the world ocean. The equation became:

$$N^* = (NO_3^- - 16 \times PO_4^{3-} + 2.90) \times 0.87$$

In our manuscript, we emphasized the fact that Michaels et al. (1996) and Gruber and Sarmiento (1997) were the first to define the N*. Nevertheless, in the South Pacific, the N* was described in detail by Deutsch et al. (2001). These authors used the equation:

$$N^* = NO_3^- - 16 \times PO_4^{3-} + 2.90$$

In our study, we chose the same definition, and we added the constant 2.90 only to compare the observations with the literature data (Deutsch et al., 2001).

14. Reviewer: If inorganic and particulate values of N and P were considered for "excess" analysis here (equations 2 and 3) then why not dissolved organic values (DON and DOP) as well? That is considered to be an equally significant pool of N and P?

Response: Reviewer#1 is right and it would have been a good idea to also show the N excess in the dissolved organic matter, but it was beyond the scope of this paper. The elemental ratio of dissolved organic matter does indeed deviate widely from the canonical Redfield ratio (i.e. Hopkinson and Vallino, 2015; Letscher and Moore, 2015). Neverthless, the turnover of the dissolved organic pool which is a gret deal longer than that of the particulate organic pool, making the N excess interpretation for the dissolved pool more difficult. Considering only the particulate matter (biomass) and inorganic matter (nutrient) is the simplest scheme. However, since this simplest scheme enables us to understand the main trends, it does not seem necessary to add more complexity and detail to the story. To clarify our manuscript, we have removed the part related to the dissolved organic matter.

15. Reviewer: Provide the reproducibility (precision) and detection limit of all the measured parameters in the method section. Can PON and POP be measured in three decimal digits of mico-mol/L, which is almost like nmol/L in 3 decimal digits?

Response: The precision (repeatability) and quantification limit (above detection limit) were established for all parameters and indicated table R4. Please note that a large volume was filtrated for particulate organic matter measurement, allowing, for example, a quantification limit of 1 nmol L^{-1} for POP measurements.

Table R4. Accuracy (repeatability) and quantification limits of inorganic nutrients and particular organic N and P. CV for coefficient of variation.

	Repeatability (CV in %)	Quantification limit (μmol L^{-1})
NO_3^-	0.2	0.05
PO_4^{3-}	0.7	0.05
DON	3.0	0.51
DOP	3.7	0.05
PON	5.3	0.02
POP	3.2	0.001

16. Reviewer: Line 383: there is a typo in "surface"

Response: Yes, we apologize for this mistake. In fact, the sentence has been deleted in the revised manuscript.

17. Reviewer: Lines 424-425: sentence "the . . . .zooplankton" is not clear

Original text (lines 424-425): The occurrence of diazotrophs and the associated nitrogen supply would allow the concomitant development of non-diazotrophic phytoplankton and zooplankton (Mulholland et al., 2007).

Response: Although we measured strong N_2 fixation rates, our PON:POP ratio was significantly lower than that observed by Karl et al., 1992 and Letelier and Karl (1998). In this part, we discuss the possible causes of the observed differences, and in particular, the fate of the nitrogen fixed by diazotrophs and the impact on the PON:POP ratio observed during the cruise. Sheridan et al. (2002), and then Mulholland (2007), showed that 85% of Trichodesmium colonies were inhabited by other organisms. Therefore, the occurrence of diazotrophs would allow the concomitant development of non-diazotrophic organisms (Caffin et al., 2018; Leblanc et al., 2018; Mulholland, 2007). The concomitant development

of non-diazotrophs organisms could explain the difference in the ratio of particulate organic matter observed between our observations and those of Karl et al. (1992) and Letelier and Karl (1998).

General comment: We propose a new version of the whole paragraph in the revised manuscript.

Modified text:

POM measurements showed a significant accumulation of particles in the surface waters and at the top of the UTW, followed by an attenuation in the central and lower part of the UTW (Fig. 5a, 5b). Despite the extremely low surface nutrient concentrations observed, a significant amount of POM was measured in the Melanesian Archipelago sampling stations, with PON and POP concentrations reaching 1.175 and 0.057 µmol L^{-1}, respectively. PON concentrations in oligotrophic regions are typically <0.6 µmol L^{-1} (Hebel and Karl, 2001; Moutin and Raimbault, 2002; Martini et al., 2013). The observed values, and particularly at station LD B, were in the same order of magnitude as those measured in subpolar waters of northern latitudes (Sarmiento and Grüber, 2006), which are usually >1.2 µmol L^{-1} in PON (Martini et al., 2013; Gruber 2008). The PON:POP ratios averaged over the entire water column were very close to the Redfield ratio, except for the most productive zones located in the Melanesian Archipelago, which had an excess of PON relative to that expected from Redfield stoichiometry, particularly close to the surface (Fig. 5d). This excess of PON was as high as 0.37 µmol L^{-1}, suggesting that the POM was enriched in N, despite the undetectable NO_3^- concentrations.

Most PON:POP ratio data available in the literature corresponds to cruises in the North Pacific (Karl et al., 1992; 2001) and North Atlantic (Hansell et al., 2004), while the WTSP has so far remained an undersampled region. These studies showed that the chemical composition of the suspended particulate matter collected during *Trichodesmium* blooms was enriched in N relative to P with PON:POP ratios ranging between 40 (Letelier and Karl, 1998) and 125 (Karl et al., 1992). The stoichiometric PON:POP ratios observed during the OUTPACE cruise were much lower, with a maximum of 25.3 observed at station LD B. The differences observed in the PON:POP ratios may be explained by differences in nutrient availability, as previously suggested by Karl et al. (2002). Oligotrophic gyres in the northern hemisphere have lower concentrations of PO_4^{3-} than those measured in the southern hemisphere (Moore et al., 2013), theoretically leading to higher PON:POP ratios in the North than in the South Pacific. Adding to in situ nutrient availability, new N derived from diazotrophic activity would allow the successive development of non-diazotrophic microorganisms (Caffin et al., 2018; Leblanc et al., 2018; Mulholland, 2007) with lower N:P content than diazotrophs, leading to a relatively lower PON:POP ratio.

18. **Reviewer: Line 428-431: "On an annual. . .. . .N* signal". See (Mills and Arrigo, 2010; Singh et al.,2017) for an alternative hypothesis.**

19. **Reviewer: Lines 433-436: "Nevertheless. . .. . .atmospheric nitrogen input". See (Jickells et al., 2017; Singh et al., 2013) for more details.**

Original text (lines 428-431): On an annual time scale, the strong anomalies of surface N* can be explained, on the one hand by the formation of diazotrophic organic matter, which together with the fixation of N_2 reduces the stock of PO_4^{3-} and positively influences the N* signal. On the other hand, part of the excess of PON which goes after remineralization towards the stock of fixed nitrogen may therefore also positively influence the N* signal.

Response: We are aware that there are alternative hypotheses to possibly explain the N* variation, and it is the subject of the following paragraph. We understand the necessity of discussing all the hypotheses that have been proposed to explain the important variations in N* against the relatively low N_2 fixation rates measured. But it should be borne in mind that the N_2 fixation rates measured in the WTSP are exceptionally high (Bonnet et al., 2017), and that strong N* variations are probably mainly explained by N_2 fixation. Furthermore, it is beyond the scope of this (already long) paper to deal with community structure, a subject that is discussed in several of the 26 other papers from the OUTPACE special issue [Interactions between planktonic organisms and biogeochemical cycles across trophic and N_2 fixation gradients in the western tropical South Pacific Ocean: a multidisciplinary approach (OUTPACE experiment)], available on the Biogeosciences website.

In the revised manuscript, alternative hypotheses have been added (Mills and Arrigo, 2010; Zamora et al., 2010; Singh et al., 2015;2017), as suggested by Reviewer#1 (see modified text below) and the sentence has been rephrased.

Modified text: However, N* could be influenced by processes other than N_2 fixation, such as the non-Redfield nutrient assimilation by non-diazotrophic phytoplankton (Mills and Arrigo, 2010; Singh et al., 2017), and/or remineralization of non-diazotrophic organisms (Zamora et al., 2010; Singh et al., 2013). Furthermore, N* could also be influenced by lateral advection of DOM with a high DON:DOP ratio (Letscher et al., 2013), and atmospheric deposition of low N:P ratio aerosols (Jickells et al., 2017). The transport is probably another process which affects N* distribution in the ocean interior (Gruber and Sarmiento, 1997).

20. Reviewer: Line 521: how can "0" rates be measured.

Original text (lines 521): Dekaezemaker et al. (2013) measured N_2 fixation rates between 0 and 148 μmol N m^{-2} d^{-1},

Response: Yes, we agree with Reviewer#1, the sentence was confusing. N* could be influenced by processes other than N_2 fixation. This sentence has been removed in the revised manuscript.

21. Reviewer: Line 555: "deficit of NO3 and PO4". Mathematically (from the N* concept used here), both nutrients cannot be deficit.

Original text (lines 555): the waters located in the Melanesian Archipelago (160° E-170° W) have a deficit of NO_3^- and PO_4^{3-} in the productive layer (0-100 dbar),

Response: Thank you for these comments, it was indeed a mistake in the formulation of the sentence. This sentence has been deleted in the revised manuscript.

22. Reviewer: Line 572: "occurred" should be replaced by "been undertaken"

Original text (lines 572): Only few cruises have occurred in the South Pacific Ocean.

Response: This sentence has been deleted in the revised manuscript (see the modified text below answer to comment 23).

23. Reviewer: Line 680: Letscher is misspelled (Letscher et al., 2013)

Response: We apologize for the mistake. The reference has been corrected in the revised manuscript.

Modified text: Letscher, O., T., Hansell, D. A., Carlson, C. A., Lumpkin, R., and Knapp, A. N.: Dissolved organic nitrogen in the global surface ocean: distribution and fate Global Biogeochemical Cycles, 27, 141–153, 2013.

General comment: In the modified text, all the corrections referred to above have been taken into account, and we propose a new version of the Conclusion.

Modified text:

As part of the OUTPACE cruise in the western tropical South Pacific (160° E-160° W, 18°-20° S), we have analyzed the effect of the local N_2 fixation process on the dissolved and particulate organic N pools. We have revealed the presence of two distinct oceanic areas. The waters located in the Melanesian Archipelago (160° E-170° W) were characterized by significant N_2 fixation rates, high PON concentrations and a PON in excess relative to that expected from Redfield stoichiometry in the surface waters and in the top of the upper thermocline waters. In this area, we showed a significant relationship between N_2 fixation and PON in excess. Positive N* in the surface waters and in the upper and lower thermocline waters of the Melanesian Archipelago was observed in parallel with record-high rates of N_2 fixation. In contrast, the surface waters and the top of the upper thermocline waters of the South Pacific Gyre (170° W-160° W) are characterized by low N_2 fixation rates and low PON concentrations. In agreement with the low diazotrophic activity observed, negative or close to zero N* values were observed in the South Pacific Gyre.

The intense N_2 fixation observed in the WTSP significantly influences the dissolved inorganic (N excess) and particulate organic (PON excess) N pools of the surface and thermocline waters in the WTSP. The analyses of the most recent available nutrient data in the South Pacific Ocean enabled us to show a high positive N* in the thermocline waters of the whole South gyre. We therefore hypothesized that the local high N* observed in the Melanesian Archipelago is transported eastward *via* the circulation, and consequently that N_2 fixation in the WTSP may influence the nutrient distribution at the scale of the South Pacific basin.