

Re: "Biogeochemical variations at the PAP-SO in the northeast Atlantic Ocean from weekly to inter-annual time scales"

Dear Prof. Kaiser,

Many thanks for your comments on our manuscript following initial publication and discussions. Thank you for agreeing to publish it after the small revisions you suggested have been completed. We have addressed each minor revision and technical comment in turn for your consideration, as detailed in the following document,

Sue Hartman

> 1) Please add page numbers to the next version.

This has been done.

> 2) Where have the SOO data been deposited? Are they publicly accessible?

A line has been added to the manuscript: Data are available from the Surface ocean CO₂ atlas (SOCAT) <http://www.socat.info/>.

> 3) The author affiliations should use the same format in each case - at the minimum, include the city and post code; the street address can be omitted.

This has been corrected.

4) Figures 2, 3 and 5: The presentation of the data as two separate time series is confusing because it suggests that the data refer to simultaneous or comparable time periods. Please use a single time-axis for each panel, starting in May 2003 and ending in June 2012, with a break from April 2005 to April 2010. Please also add an x-axis and x-axis ticks for the tick labels. The December data point in the NAO time series does not line up with the "Dec" label. Please indicate whether the x-axis ticks (to be added) refer to beginning of the corresponding month or the middle of the month. Please also check the location of the vertical blue lines - I guess they should represent the beginning of the year?

We have followed the editor's suggestion to "use a single time-axis for each panel starting in May 2003 and ending in June 2012". However we find that it is difficult to now make a direct comparison between the 2003-2005 and 2010-2012 data sets. The x-axis now clearly shows monthly tick marks and the vertical line corresponds to the start of each year so this has been clarified.

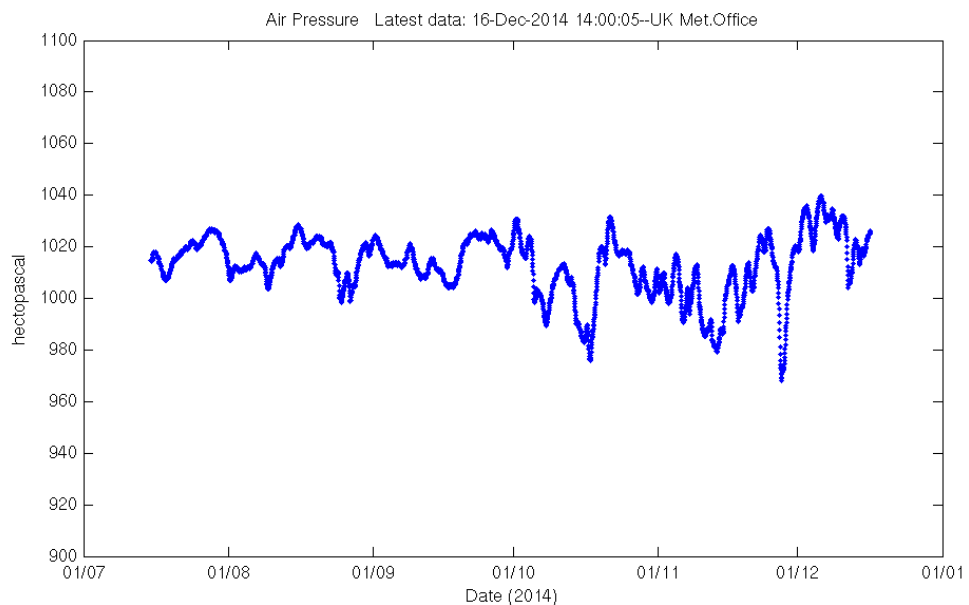
5) The "correction" of DIC concentrations for the CO₂ air-sea flux has not been explained. Please add a corresponding paragraph. Do you use a constant or varying Revelle factor? How is the CO₂ air-sea flux time series "mapped" onto the DIC concentration time series, e.g. to correct the DIC concentration change between time t₁ and t₂, do you use the average flux at t₁ and t₂, multiplied by (t₂-t₁)? If so, how is this correction partitioned between the concentrations at t₁ and t₂? And how were MLD variations over the period between t₁ and t₂ taken into account? Also, when the MLD was less than 30 m, presumably no correction was applied to the DIC concentration?

"The interrelation between DIC and nitrate concentrations can be considered by comparing the C : N ratios to the Redfield ratio (Redfield, 1958). The 2003-2005 time period has already been considered in Körtzinger et al. (2008) so is not reproduced here. In a similar way, the monthly gas exchanged-corrected DIC concentrations (the DIC changes driven by air-sea exchange was estimated as $\Delta \text{DIC}_{\text{gas}} = F_{\text{CO}_2} / \text{MLD}$ following Körtzinger et al. (2008)) were plotted against the *in situ* nitrate concentrations in different seasons for 2010-2012 (Fig. 4). "

Response to the Reviewer: We followed Körtzinger et al. (2008)'s method to correct the DIC changes driven by air-sea exchange: $\Delta \text{DIC}_{\text{gas}} = F_{\text{CO}_2} / \text{MLD}$. Therefore, no Revelle factor was used. The correction was applied to the monthly averaged DIC using monthly averaged F_{CO_2} and MLD. The result was similar to that of Körtzinger et al. (2008): the gas exchange played a negligible role in winter while the influence increased in summer when the MLD was shallow.

6) The $p(\text{CO}_2, \text{air})$ calculations assumes a surface pressure of 1013.25 hPa. Is this reasonable? The global mean sea level pressure is 984 hPa, nearly 3 % lower. As CO_2 is undersaturated, this would reduce the corresponding air-sea flux, but ideally the actual pressure should be used in the calculation and can be obtained, for example, from global reanalysis products (ECMWF, NCEP) if not available from the surface buoy observations.

The buoy data shows a higher annual average air pressure than you suggest (closer to the 1013 hPa used). See figure below.



> 7) Please define the 2003-2005 and 2010-2012 time periods. Neither of them span 3 complete years, so it's important to mention the start and end months.

The time periods were defined at the end of section 2.2. 'Data presented here cover the period when $p(\text{CO}_2)$ measurements are available, July 2003 to the end of June 2005 (with deployments in July 2003, November 2003, June 2004) and the period from May 2010 to June 2012 (with sensor deployment in May 2010, September 2010, July 2011, May 2012).' We feel that this is the appropriate place for it as the dates displayed are driven by deployment dates and by when the sensors are functioning so is entirely relevant to the in situ data section. For clarity we have added the months of data coverage to the end of the introduction section too: 'In this paper, we present recent year round time-series data of temperature, salinity,

nitrate concentration, chlorophyll a-fluorescence and $p(\text{CO}_2)$ collected at 30 m depth from May 2010 to June 2012. The data are compared with an earlier published dataset (from July 2003 to July 2005) and additional $p(\text{CO}_2)$ measurements made from a ship of opportunity.'

8) p. 2, l. 9 (2/9): It is unclear why you don't mention the secular CO₂ increase due to anthropogenic CO₂ emissions here and instead refer to the similar temperatures etc. for both time periods.

We have corrected this: The 2010–2012 period shows an overall increase in $p(\text{CO}_2)$ values when compared to the 2003–2005 period as would be expected from increases due to anthropogenic CO₂ emissions.

The following comments on the text have all been corrected:

> 9) 2/24: Delete "However" - there is no contradicting or contrasting statement.

> 10) 2/27: Please add "for the role of the ocean" after "now and".

> 11) 3/29: Change "from 30 m" to "at 30 m".

> 12) 4/18: "within deep chlorophyll maxima" or "within the deep chlorophyll maximum".

> 13) 5/1: "Note that a measurement error of an early version"

> 14) 5/2: "fluctuation"

> 15) 5/5: "accelerate the equilibration" or "accelerate attaining equilibrium"

> 16) 5/6: Please add a brief explanation why the time series was interrupted.

This has been added in the appropriate place. CO₂ data were not collected from 2005-2010 due to funding issues (although the 2011 data were missing due to problems with the instrument logger and that was already disclosed).

The following comments on the text have all been corrected:

> 17) 5/11: "checks"

- > 18) 5/16: "chlorophyll a concentrations"
- > 19) 5/17: "precision for fluorescence measured by these fluorometers is"
- > 20) 5/19: "approximation of the chlorophyll a concentration. The fluorescence/chlorophyll a concentration ratio"

- > 21) 6/16: Please add the microcat data to the revised time series (item 4). This is an interesting and useful comparison.

We have added the microcat data to the plots, where available.

The following have all been corrected:

- > 22) 6/28: "value" or "average of the p(CO₂) values"
- > 23) 7/8: Please format this equation as follows:
> $F(\text{CO}_2) = k K_0 [p(\text{CO}_2, \text{sea}) - p(\text{CO}_2, \text{air})]$ Only F , k , K_0 and p should
> be in italics.
- > 24) 7/12: $p(\text{CO}_2, \text{sea})$ and $p(\text{CO}_2, \text{air})$
- > 25) 7/14: "averaged CO₂ dry mole fractions measured" - please also see item 6 above.
- > 26) 7/18: "Total alkalinity was calculated ..."

- > 27) 7/19: Does this uncertainty include the 1 % temperature and 2 % salinity uncertainties due to the use of the Argo data (p. 6)?
In the revised manuscript, we have taken these factors into account in estimating the overall uncertainty (see response to comment 29).

- > 28) 7/23: The "K"s should all be in italics because they are physical quantities. We have corrected this.

- 29) 7/24: "calculate DIC concentrations introduces an uncertainty of $\pm 6.4 \mu\text{mol kg}^{-1}$ " [or more? - see item 27]

Using TA and $p(\text{CO}_2)$ to calculate DIC, taking the various uncertainties in the calculation into account, introduces an error in the order of $7.0 \mu\text{mol kg}^{-1}$.

We have corrected the following:

> 30) 7/26: "monthly mean MLD"

> 31) 9/1: "The increase in chlorophyll a fluorescence during the 2011 spring bloom was also larger"

> 32) 9/13: Replace "an opposite trend" with "opposite seasonal variations"

> 33) 9/31: See item 5 above.

> 34) 10/4-15: These paragraphs need to come before the use of air-sea flux corrected DIC concentrations.

We do not agree in this case and have left the paragraphs as they are as the discussion of DIC & nitrate then come before CO_2 flux and wind measurements.

> 35) 10/14: Please enclose (5.7 ± 2.8) and (5.0 ± 2.2) in brackets (so that the unit applies to the mean and the uncertainty).

We have done this although we have seen this written both ways in the literature.

> 36) 10/14: The start (2003: April, 2010: May) and end months (2005: Mar, 2012: June) of both periods differ. Please discuss the bias caused by these differences. Do the flux calculations include the uncertainties due to a) $p(\text{CO}_2)$ calibration, b) the temperature and salinity-substitution from Argo floats, c) uncertainty in wind speed (due to the use of the $1^\circ \times 1^\circ$ FNMOC data), d) uncertainty in sea level pressure, e) uncertainties in the gas exchange coefficient. At least the first four uncertainties (a to d) should be quantifiable.

These uncertainties are the same for both periods of time. The ranges that we see are based on eth standard deviation of the results seen rather than a consideration of every possible error (some of which we cannot quantify). We have added a line to this effect to the text.

The following comments have been acted on and small changes made to the text.

> 37) 10/26: "inversely" suggests a "1/temperature" relationship - I guess this should be "seasonal variations opposite to temperature"?

> 38) 10/28: Delete "the" before "convective".

> 39) 11/22: Delete "the" before "p(CO2)".

> 40) 11/24: The reference refers to tropical cyclones and has no immediate relevance to the PAP site. Please use a more suitable reference.

We agree and have added a different reference. 'In the North Atlantic the strength and frequency of wintertime storms is significantly increasing (Donat et al., 2011).'

Reference added:

Donat, M. G., Renggli, D., Wild, S., Alexander, L. V., Leckebusch, G. C., and Ulbrich, U.: Reanalysis suggests long-term upward trends in European storminess since 1871. *Geophys. Res. Lett.*, 38, L14703, 2011.

> 41) 12/4: The missing data have been substituted by SOO data in Figure 2, so I suggest you use these to fill in the gap.

We have now tried to fill in all gaps in the PAP-SO data where SOO data are available. This is for pCO₂, CO₂ flux, nitrate and calculated DIC.

> 42) 12/8: This suggests an average over 3 complete years in each case, but see item 36 regarding start and end months.

As commented above we have added a line about this. 'The start and end months of the two periods of time differs, which will contribute to the errors in the flux measurements. However the errors are comparable for the two periods of time considered and overall the average for the two time periods is similar.'

These small changes have been made:

- > 43) 12/21: 2010-2012
- > 44) 13/5: Please add brackets around (mean±uncertainty).
- > 45) 13/7: "p(CO₂)
- > 46) 13/8: 2010-2012
- > 47) 13/13: "at PAP-SO"
- > 48) 13/22: "require" and "At PAP-SO"
- > 49) 14/3: "effect of storms"

> 50) Figure 1: Legend (colour bar) and axis labels are missing. Please add white contour lines and contour labels to the bathymetry. Please use a white symbol for the PAP site. The quality of the figure is rather poor (jpeg?) - please use PDF, EPS or another vector graphic format instead.

This figure has been updated to include a colour bar for the bathymetry. A PDF version is available. We tried adding white contour lines but it was clearer to just present the colour bar & bathymetry.

> 51) Figure 2: y axis labels should be "p(CO₂)", "chlorophyll a concentration" and "nitrate concentration" (or "NO₃⁻ concentration"). Does weekly nitrate concentration refer to "weekly average" or "measured once on a weekly basis"? Please clarify.

We have updated all figures and the figure legend refers to weekly averaged nitrate concentrations. We do not agree to add the word 'concentration' to the y-axis label in line with other papers (although this is specified in the legend text).'
Figure 2. *In situ* 30 m PAP-SO data (blue circles) from 2003–2005 and 2010–2012 and 5 m SOO data (red squares) with vertical lines to represent the start of each year showing: (a) p(CO₂); (b) chlorophyll a concentration; (c) weekly averaged nitrate concentration.'

> 52) Figure 3: Please cross-refer to the corresponding section in the text where you explain how DIC was calculated (based on $p(\text{CO}_2)$ and salinity-based TA parameterisations).

The legend has been modified to include the word 'concentration' also to refer to the text in the paper for methods as follows: 'Figure 3. Data from 2003–2005 and 2010–2012 (blue circles) with vertical lines to represent the start of each year showing: (a) Argo temperature data from 30 m depth around the PAP-SO and *in situ* MicroCAT temperature data at 30 m (red circles); (b) calculated mixed layer depth (MLD) data; (c) calculations of weekly dissolved inorganic carbon (DIC) concentrations based on *in situ* PAP-SO $p(\text{CO}_2)$ and salinity-based TA parameterisations (see text for details) with additional DIC calculations based on SOO data (red circles)

> 53) Figure 4: Please add "concentration" to x and y axis labels. "Jan" in the legend should have a capital "J".

As stated before we do not agree to add the word 'concentration' to the y-axis label in line with other papers (although this is specified in the legend text). We have amended the figure legend.