# Scaling and intermittent properties of oceanic and atmospheric  $pCO<sub>2</sub>$ time series and their difference

## Reply to Referees' Comments

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We would first like to thank Referee 1  $(R1)$  and 2  $(R2)$  for their comments and for their involvement in this review process. In the sections below, we have made some enhancements to better delineate between Referees' comments and our manuscript corrections. In this iteration, comments from Referees are highlighted in blue text, while our modifications within the manuscript are denoted in green text.

This letter of response is structured into three parts: Section [1](#page-0-0) addresses R1's comments, Section [2](#page-5-0) covers R2's comments, and finally, Section [3](#page-7-0) describes the other changes made in the text.

## <span id="page-0-0"></span>1 Referee 1 comments (RC1)

#### 1.1 Major comments

1. The authors firstly present results based on Fourier PSD for spectral exponents, while later they use EMD/HSA to provide more insights on high-order statistics, as well as, to also reduce effects of periodicity in time series that could destroy scaling behavior. Why not to directly use EMD/HSA for also investigating spectral slopes by using the second-order moment of the generalized Hilbert spectra? This would directly overcome limitations provided by Fourier PSD.

We used Fourier spectral analysis in a first step because it is the most precise method to consider the scaling properties of the series, especially because the peaks are very localized in the Fourier spectral space. The effect of the periodicity on the scaling of the spectrum is reduced. Yet, this method allows to consider the spectral dynamics only for the second-order moment and not for the other statistical moments q. This is why we then used EMD-HSA to study intermittency: it is a method allowing to consider the spectral dynamics for different statistical moments  $q$  while suppressing the effect of the periodicity [\(Huang et al., 2011\)](#page-9-0).

Concerning the second-order moment it is expected to observe an agreement between the spectral slope evaluated via Fourier PSD and those evaluated via EMD/HSA. This agreement seems to be missed if one looks at the second-order exponents for SSS in Figure 8 for Gulf of Maine and especially for pCO<sup>2</sup> air time series.

It is right that in the first version there was a poor agreement between the Fourier spectrum and the second moment order exponent extracted with EMD-HSA. We corrected this by using the same bounds for the regression. See one example below for BOBOA SST time series (Fig. [A\)](#page-1-0).

<span id="page-1-0"></span>

Fourier spectrum O Hilbert spectrum  $\circlearrowright$ 

Figure A: Fourier spectrum  $E(f)$  and second order moment Hilbert marginal spectrum  $L_2(\omega)$  for BOBOA SST time series. The associated slope values for Fourier  $(\beta)$  and Hilbert  $(\zeta(2))$  spectra are also provided.

<span id="page-1-1"></span>The following graph (Fig. [B\)](#page-1-1) represents the second order moment slope value extracted with Fourier  $(\beta)$  and EMD-HSA  $(\zeta(2)+1)$ . There is now an overall very good agreement between values estimated by both methods. The correspondence for the Gulf of Maine case in now good as can be seen in the new figure Fig. 8 in the modified manuscript.



Figure B: Comparison between the second order moment slopes found using the Fourier spectral analysis ( $\beta$ ) and the EMD-HSA method ( $\zeta(2)+1$ ). The black dashed line represents the first bisector  $y = x$ .

2. Concerning the scaling exponents of  $pCO<sub>2 air</sub>$  time series it seems that, especially for Gulf of Maine, a plateau is reached for high-order statistics that could be related to missing statistics for proper eval-

uation or to the choice of the range of scales where scaling exponents are evaluated. This needs to be fixed or explained.

The plateau mentioned by the referee for the the  $pCO<sub>2</sub>$  air time series of Gulf of Maine buoy is not present anymore in the revised version, due to the modified range of scales for the extraction of the moment function using the EMD-HSA method. In few cases, we still find this saturation (see Gulf of Maine SSS example in Fig. 8 of the revised version). This kind of saturation phenomenon has already been observed for passive scalar turbulence [\(Watanabe and Gotoh, 2006;](#page-9-1) [Schmitt and Huang,](#page-9-2) [2016;](#page-9-2) [Iyer et al., 2018\)](#page-9-3).

3. One of the main result is that multifractal intermittency is shown for the first time for oceanic and atmospheric  $pCO_2$  time series. However, by looking at Table 5 I would say that intermittency is almost zero, considering average values of the  $\mu$  parameter and its error range. Could this be related to the choice of the multifractal model, i.e., the log-normal one?

The multifractal intermittency is visible since all the moment functions are non-linear: the more it is non-linear, the more it is intermittent. We have proposed to quantify this level of intermittency using the parameter  $\mu$  defined by the values extracted for statistical moments order  $q = 1$  and 2:  $\mu = 2\zeta(1) - \zeta(2)$ . With this definition, the intermittency for the velocity field and for the passive scalar fields are also small (respectively 0.04 and 0.12; [Schmitt et al., 1996;](#page-9-4) [Schmitt, 2006\)](#page-9-5). It is right that there is a relatively large variability for this parameter estimated from our time series. In fact by considering boxplots below (Fig. [C\)](#page-2-0), we can better understand these values. The fact that the standard deviation values are close to the mean values is due to the asymmetry of the distribution. Moreover, with the new estimation of the intermittency parameter we have done here, the  $\mu$  values are slightly larger than in the first version of the manuscript.

<span id="page-2-0"></span>

Figure C: Boxplot for  $\mu$  values obtained with the EMD-HSA analysis.

4. A general comment on Figs. 4, 5, 7, 8: it would be desirable to add error bars on the estimated quantities (exponents) since this would allow directly to see if they are really different or can be comparable in the range of uncertainty. Furthermore, I would suggest to add in Fig. 4 both errors on  $\beta$  as well as the indication of the range of frequencies where the spectral law is evaluated. The latter can be really useful to estimate the goodness of the fit, especially if looking at lower-middle panel when peaks appear at high frequencies.

The required changes have been made for Fig. 4, 5, 7 and 8. A modification of the caption of Fig. 4 has been made: "The horizontal dashed lines represent the range used for the estimation of the slope".

5. Figure 7. More than a comment it is a suggestion. It is an interesting result that seems to suggest that something occurs at northern mid latitudes. What about to see if there are variations in the slope of the low frequency regime that could be related to some large-scale forcing affecting that region?

These latitudes are dominated by coral reefs sites for which the  $\beta$  values are lower, this could be the reason of these lower values. A generalization for latitudinal effect at global scale would need much more measurements from buoys at different places. The text has also been modified to mention this point: "This latitudinal gradient can also be explained by the fact that there are more series belonging to the coral reefs ecosystem in the database for latitudes between  $20^{\circ}N$  and  $30^{\circ}N"$  (line 176).

What about similar analysis for SST and SSS?

<span id="page-3-0"></span>Below we show the same analysis for SST and SSS. We didn't added it in text because the trend is less clear for these scalars (Fig. [D\)](#page-3-0).



Figure D: Average Fourier spectral slope  $\bar{\beta}$  in function of the latitude for SST and SSS time series. The red dotted line represent the value 5/3. The ranges of values used for each point are represented by the black dotted lines:  $[46.8^{\circ}\text{S}$ ; 0°[ (3 stations),  $[0^{\circ}; 20^{\circ}\text{N}]$  (9 stations),  $[20^{\circ}\text{N}; 30^{\circ}\text{N}]$  (7 stations),  $[30^\circ N; 50^\circ N]$  (13 stations) and  $[50^\circ N; 69^\circ N]$  (6 stations).

#### 1.2 Minor comments

– Line 16: "mitigated" seems to not be appropriate.

The term "mitigated" have been replaced by "partially counterbalanced" (line 18).

– Line 26: please clearly state which temporal and spatial scales are referring to.

Two reference scales have been added in the text: "annual" for the temporal scale and "planetary" (line 28) for the spatial scale.

 $-$  Line 29: "flux"  $\rightarrow$  "mixing"?

The term "flux" seems to us more appropriate here as the Eq. 1 is used to calculate the flux of  $CO<sub>2</sub>$  at the interface between the lower atmosphere and the surface ocean.

 $-Eq.$  (3): does it hold for current measurements? Is β a function of the depth and of the frequency? Please clarify.

We are not sure to properly understand the question here. This equation can be satisfied for any scalar transported by a homogeneous and isotropic turbulence. In this equation,  $\beta$  is independent of the frequency: it is a scale invariant exponent. However, it could depend on the depth but here we have fixed depth measurements.

 $-$  Lines 124 and 125: "Pikes"  $\rightarrow$  "Peaks"?

Fixed.

– Figure 6: what are the dots?

This is a classical notation in the boxplot representation: the dots mean outliers, indicating values that fall below or above the first or third quartile (respectively) by more than 1.5 times the interquartile range (the distance between the first and third quartiles). This have been added in the caption of the Fig. 6: "The dots signify outliers, indicating values that fall below or above the first or third quartile (respectively) by more than 1.5 times the interquartile range (distance between the first and third quartiles)".

– Line 199: linear interpolation could introduce some kinds of spurious high-frequency intermittent-like bursts where it is performed. Is it the case? If yes, these time intervals are removed for the evaluation of slopes?

It was a mistake in the text. This has been corrected: "This transformation needs regular time steps as it done using fast Fourier transform (Huang and Schmitt, 2014). The missing data were so replaced by the value 0. [...] When we have the instantaneous frequency information, missing data parts are then excluded in the following steps." (lines 202 to 204 and 205 to 206).

 $-Eq.$  (6): there is a missing "Principal Value".

The "PV" notation was added into the equation (6) which is now followed by the sentence "where PV indicates the Cauchy principal value" (line 200).

– Line 234: I do not see this agreement for  $pCO_{2}$  air time series (see Major Comments 1 and 2).

This has been corrected, please see the major comments section and our answers.

## <span id="page-5-0"></span>2 Referee 2 comments (RC2)

 $- I$  suggest to include 'in a turbulence framework' at the end of the title, especially since it is the first time this is done in this study.

Done.

– The abstract should include some important results. At the moment the abstract mostly lists what was done with very little information on what was found as a result.

We changed the abstract to introduce more results at lines 2, 10, 11 and 12.

– Line 5: Remove brackets: '...ecosystems such as coastal shelf,..'.

Done.

– Line 18: In context of the anthropogenic  $CO<sub>2</sub>$  uptake by the ocean, the authors should also cite Sabine et al.  $2004$ : The Oceanic Sink for Anthropogenic  $CO_2$ . Science 305, 367-371(2004). DOI: 10.1126/science.1097403.

Done.

– Line 29: influence of small scale turbulence.

We have added "multiscale" (line 31), because as we shown in the paper through the scaling properties analysis, the turbulence effect is not limited to small scales.

– Line 38: partial pressures.

Fixed.

 $-$  Line  $43$ : Here it is unclear whether the authors refer to the small scale or large scale turbulence, given that it is shown to evolve over a large period up to 3 months. Authors should clarify which kind of turbulence they refer to here, and also at other instances in the manuscript where turbulence appears.

We have added a new sentence to indicate that turbulence is not limited to small or large scales, but it is in reality a multiscale process: "This means that in agreement with the Richardson cascade theory, there is an inertial range where turbulence influence is present over a rather large range of scales from the largest (e.g. months) to the smallest (e.g. seconds)" (lines 45 to 47).

 $-$  Line  $44$ : '...their scaling properties'. Whose scaling properties? Clarify.

Done: " $\ldots pCO_2$  scaling properties" (line 48).

 $-$  Line 59:  $\ldots$  by the authors'. Which authors does this phrase correspond to: the present authors or the authors of a different manuscript? Please specify.

Done: "...by Sutton et al. (2019)..." (line 63).

 $-$  Line 62: 'The data paper presenting the database...'. Please cite the publication instead of such a phrase.

Done.

– Line 86: '...proportion of time in negative or positive values.' This is unclear, please clarify the sentence.

In order to clarify this, we have modified the sentence as: "The sink or source of  $CO<sub>2</sub>$  of the different ecosystems depends on the proportion of time spent in the negative or positive values: the global mean can be written as  $\langle \delta \rangle = p^+ \langle \delta | \delta > 0 \rangle + p^- \langle \delta | \delta < 0 \rangle$ " (lines 89 and 90).

– Line 86: 'It indicates..'. It refers to what?

This has been modified, please refer to the previous comment.

– Line 88: ...sinks than sources...

Fixed.

– Line 89: what is reversed? Clarify.

This has been clarified: "For the coral reefs sites, it is the opposite: they are more often sources than sinks  $(70\%$  versus  $30\%$ " (lines 93 and 94).

 $-$  Line 95: ...a proportion that is reversed... Fix the typo.

This part of the sentence was modified, please see previous comment.

 $-$  Line 95:  $\ldots$  a proportion that is reversed...'. Is the proportion reversed exactly also in percentages? Please clarify.

This was clarified in the text.

– Line 132: Please rewrite this sentence to make it clearer.

This sentence has been removed following RC1 corrections.

– Lines 175 and 244: Here and elsewhere: please be consistent with the user of either  $CO<sub>2</sub>$  or carbon dioxide throughout the manuscript.

Fixed. The "CO<sub>2</sub>" notation have been chosen. The expression "carbon dioxide" have been used only at the first " $CO_2$ " appearance in the text (line 15).

 $-$  Line  $245: ...$  these quantities...

Fixed.

 $-$  Line  $246: ...$  on multiscales...

This part of the sentence has been modified.

– Section 5: The main results should be mentioned in this section more clearly which are currently missing or somehow unclear to the reader.

We have modified this section in order to provide more details on the results (lines 249 and 267 to 271)

– Table A1: Is there another way of depicting the information in this table? Or if it is already published, perhaps it can be simply referred to the published dataset as a citation?

The original datasets have been slightly modified to harmonize the 3-hours time step. The proportions of missing data and the size of the datasets are therefore no longer exactly the same as in Sutton et al. (2018, 2019). Moreover, the proportion of missing data is not given in the original references (it was computed by us). This prevents us from simply referring the reader to this source to get the dataset information.

Furthermore, the table format, although cumbersome, seems to us to be the only option for containing information on these datasets.

## <span id="page-7-0"></span>3 Other changes

In addition to the changes following the referee's comments, we have made some changes listed below, to improve the manuscript.

#### 3.1 Minor changes

The Fourier scaling exponents of some time series possessing a large proportion of missing data have been re-estimated taking into account only a portion (the longer one) of these time series. Indeed, some suspicious values were found (close to  $\beta = 1$ ) and we realized that these slopes were due the large proportion of missing values in the series. This was the case for the following time series:

- SST: Coastal MS, Kaneohe, M2
- SSS: CCE2, Coastal MS, Kaneohe, M2, TAO155W, TAO170W
- $pCO<sub>2 air</sub>: M2, TAO155W$
- $pCO<sub>2 sw</sub>: M2, TAO155W, TAO165E$
- $\Delta p \text{CO}_2$ : M2, TAO155W, TAO165E

For theses time series, a separation into different smaller series (of at least 1,000 observations) with fewer missing values was made and the Fourier analysis was performed on the longer portion after removing the missing values.

Because of these new results, the following sentences were removed: "Only in few cases the SST spectral slopes is much smaller than 5/3: for M2, Coastal MS and Kaneohe. Different explanations can be given for this. First, M2 is an Alaskan buoy where ice is seasonal (Jin et al., 2007; Stabeno et al., 2010). This phenomenon could be the origin of a peculiar scaling. Second, Coastal MS is a buoy located in the Mississippi Delta. Rivers can influence the physicochemical parameters of coastal waters, such as temperature, salinity (Crossland et al., 2005; Walker et al., 2005) and also  $pCO<sub>2</sub>$ (Kealoha et al., 2020). Thirdly, Kaneohe is situated in Kaneohe Bay (Hawaii). In this region, the Kaneohe stream and other streams have an impact on the marine environment (Drupp et al., 2011), which could influence the spectral slope of the temperature field."

#### 3.2 Technical changes

– The expression "in-situ" has been replaced by "in situ" in the text (line 29).

– The caligraphy of some " $p$ " has been changed in the text in order to maintain homogeneity.

– The expression "In each case" has been replaced by " $In most cases$ " in the text as the scaling is not found at smallest scales for some spectra (line 127).

– The word "*potentially*" has been added in the text as it is not sure that the 12-hours peak is linked to tidal cycle. Indeed, it can also be an artefact of the daily periodicity due to the used method (line 129).

– The minimum and maximum values found for  $\beta$  have been changed in the text. Indeed, the former values were not the right ones for the totality of the dataset but only for the time series observable in Fig.4 (line 131).

– A reference (Anderson and Verma, 1985) has been added in the text (line 141).

 $-$  "β" has been replaced by " $\bar{\beta}$ " in Fig. 7 caption.

– The caption of Table 5 has been modified to be more precise: "For homogeneous and isotropic turbulence, the expected value for H is 1/3 with an experimental intermittency parameter  $\mu \simeq 0.04$ ".

– The word "buy" has been removed (line 175).

– The sentence "sea surface salinity, sea surface temperature" has been replaced by "sea surface temperature, sea surface salinity" in the text to maintain homogeneity of their order of apparition (line 243).

– The name "Corrsing" (instead of "Corrsin") has been corrected (line 254).

– The reference Sutton et al., 2018 have been added (lines 53, 277 and in Fig. 1 caption). This is the reference of the dataset used in the paper, later presented in the paper of Sutton et al., 2019. The map reproduced in Fig. 1 come from Sutton et al., 2018 and not directly from Sutton et al., 2019. This have been corrected.

– Some missing DOI have been add for the following references:



## References

- <span id="page-9-0"></span>Huang, Y. X., Schmitt, F. G., Hermand, J.-P., Gagne, Y., Lu, Z. M. and Liu, Y. L. (2011), 'Arbitraryorder Hilbert spectral analysis for time series possessing scaling statistics: Comparison study with detrended fluctuation analysis and wavelet leaders', Physical Review E 84(1), 016208.
- <span id="page-9-3"></span>Iyer, K. P., Schumacher, J., Sreenivasan, K. R. and Yeung, P. K. (2018), 'Steep Cliffs and Saturated Exponents in Three-Dimensional Scalar Turbulence', Physical Review Letters 121(26), 264501.
- <span id="page-9-5"></span>Schmitt, F. G. (2006), 'Linking Eulerian and Lagrangian structure functions' scaling exponents in turbulence', Physica A: Statistical Mechanics and its Applications  $368(2)$ , 377–386.
- <span id="page-9-2"></span>Schmitt, F. G. and Huang, Y. (2016), Stochastic Analysis of Scaling Time Series: From Turbulence Theory to Applications, Cambridge University Press.
- <span id="page-9-4"></span>Schmitt, F., Schertzer, D., Lovejoy, S. and Brunet, Y. (1996), 'Multifractal temperature and flux of temperature variance in fully developed turbulence', Europhysics Letters 34(3), 195.
- <span id="page-9-1"></span>Watanabe, T. and Gotoh, T. (2006), 'Intermittency in passive scalar turbulence under the uniform mean scalar gradient', Physics of Fluids 18(5), 058105.