Author Comment with regard to:

"Insights from year-long measurements of air-water CH4 and CO2 exchange in a coastal environment"

by Yang et al.

Many thanks for the detailed *comments and suggestions from Anonymous Referee #2*. Below are our replies to the referee's comments, which are in *italic*.

Anonymous Referee #2

General Comments

Yang et al. present an annual monitoring of CH4 and CO2 fluxes from a coastal location in the southwest UK, using the eddy covariance technique. The paper is generally well-written and easy to follow. The manuscript shows important findings related to the investigation of carbon dynamics in coastal zones, which remains largely unknown due to its intrinsic high spatio-temporal variability. The authors showed high differences between fluxes measured at daytime and nighttime, implying that results covering only daytime may be biased due to the influence of biological activities (called diurnal) and tidal processes (called semi-diurnal). The methodology of in situ measurements and data processing are consistent. However, the authors must to address the important appointments from Erik Nilsson and the other anonymous referee. The authors also must improve the description of state of art of CO2 and CH4 dynamics in coastal zones and estuaries (introduction). This section is poorly described. The same appointment is also true for the discussion section related to the dynamics of these GHGs. This part of the manuscript is also unsatisfactorily constructed. The manuscript is well described/structured in terms of technical aspects, but the data interpretation is not sufficient discussed in terms of ecological/biogeochemical processes. A more detailed bibliographical survey is strongly recommended to support your findings and to better contextualize this study.

Specific Comments

Abstract What it means the semi-diurnal timescales (tidal processes)? Please, describe what is the pCO2 (partial pressure of CO2 in water/air).

We will specify that 'semi-diurnal' refers to tide-driven variability. We will define pCO2.

Introduction

See the general comments.

Lines 31-32: ". . .have been increasing over the last few hundred years primarily due to human activities (Hartmann et al. 2013)." I agree. However, the fastest increase is related to the last 50 years. . .

We will mention that the fastest increases have occurred over the last 50 years.

Lines 38-43: This paragraph is a simplistic exposition of the CH4 dynamics/cycling. You should go deep in this part, especially in the studies covering the coastal oceans/estuaries. I recommend a better literature research.

We will add the following:

CH4 concentrations in estuaries can be influenced by processes including biological productivity, organic carbon input, benthic and particle-derived CH4 production, oxygen content, as well as the hydrodynamics (e.g. Upstill-Goddard et al. 2000; 2016). In regions of intense benthic methanogenesis, gas bubbles supersaturated with CH4 episodically rise through the water column to the surface (e.g. Dimitrov, 2002; Kitidis et al., 2007). This process of ebullition will result in CH4 emissions that are not quantified using air-sea flux calculations based on seawater CH4 concentration. In coastal seas, CH4 saturation tends to be lower than in estuaries, but is still much greater than 100% (e.g. mean >200% for European shelf waters; Bange et al. 2006).

Lines 45-47: "Globally averaged, the open ocean is modelled to absorb about a quarter of the anthropogenic CO2 emission (Le Quéré et al. 2015)." You can give numbers, and update this reference (from the global carbon budget 2018; Le Quéré et al. 2018). https://www.earth-syst-sci-data.net/10/2141/2018/

Suggestion accepted.

Lines 50-56: "Estuaries, on the other hand, are generally net sources of CO2 for the atmosphere (e.g. Frankignoulle et al. 1998). . ." How much? Frankinoulle et al. 1998 is a classical reference. However, there are more recent references that you should include when is describing the global emissions of CO2 by estuaries. In addition, you should shortly describe processes that can affects the dynamics and fluxes at the air- water interface.

Processes that affect gas exchange at the air-water interface (in terms of delta C and K) are already described between lines 61 and 74. However, we will add the following text:
"Inner estuaries are estimated to emit about 0.3 Pg C yr-1 of CO2 globally (Laruelle et al. 2010; Cai 2011). Most of this CO2 emission is due to the degradation of allochthonous organic matter rather than a direct input of dissolved inorganic carbon (Borges et al. 2006)."

Lines 55-56: "The shallow seas are predicted to become a greater net sink of CO2 in the future due to rising atmospheric CO2 and increasing inorganic nutrients (e.g. Andersson and Mackenzie, 2004)." However, other studies showed that the estuaries could emit more CO2 due to the enhancing of organic matter respiration.

We will rephrase to "The coastal seas may have been heterotrophic during preindustrial conditions and thus a net source of CO2 due to organic carbon degradation (e.g. Smith and Hollibaugh, 1993). Some studies (e.g. Andersson and Mackenzie, 2004; Cai, 2011) predict that shallow seas will become a net sink (or a reduced source) of CO2 in the future due to rising atmospheric CO2 levels and increased inorganic nutrient inputs."

Lines 73-74: "Thus a wind speed-only dependent representation of K, incomplete for the open ocean (Wanninkhof et al. 2009)." Please rephrase.

We will rephrase to: "Thus a wind speed-only dependent representation of K is probably less appropriate for coastal environments than for the open ocean."

Experimental Lines 105-107: "For this paper, wind data from the Windmaster Pro sonic anemometer were used between September 2015 and March 2016. Since March 2016, wind data from the R3 sonic anemometer (not operational for the first 6 months of this annual study) were preferred because of its higher precision and better performance during heavy rain events. Did you compare the wind velocity from the 2 different anemometers used in this study?

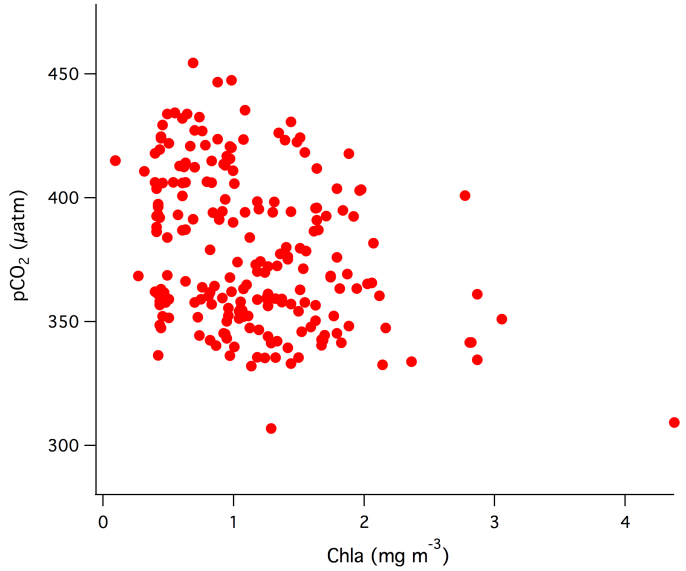
Yes, the wind velocities from two sonic anemometers agree to within 2%. We will add this information to the supplementary.

Results Fig. 1. Could you better explain the causes of the negative fluxes of CH4? This means that the water was sub-saturated with respect to atmospheric CH4 concentrations? What are the main causes of this under saturation?

As shown in Fig. 2, in January 2016 the mean CH4 flux was about zero and the 25th percentile of CH4 flux was about -0.025 mmole/m2/d. The precision of the CH4 flux was at best 0.01 mmole/m2/d, and tends to worsen at lower wind speeds. Thus most of what appears to be 'negative' CH4 flux is within the precision of the eddy covariance flux measurement and is not statistically different from zero.

Could you add a plot combining the results of chl a concentrations and the pCO2 fluxes/values (scatter plot)?

The plot below shows that pCO2 broadly decreases at higher Chla concentrations. We will add this to the supplementary. There isn't any significant relationship between CO2 flux and Chla, since the CO2 flux is primarily determined by the air-sea pCO2 difference as well as near surface turbulence.



Lines 210-213: "If the PPAO open water footprint is representative of the nearest 1.4 km (i.e. X90 of our fluxes, see Section 2.2) of the UK coast, our measurements extrapolate to a total CH4 flux of 4.8 Gg yr-1 in UK coastal seas." The extrapolation of the results to other areas is a good exercise. However, CH4 is a gas that present very special conditions of production and consumption. I am not convinced about this calculation.

We will state that this is only an order-of-magnitude calculation.

Lines 219-221: "Ambient variability (in e.g. dissolved concentrations) largely drives the rapid temporal fluctuations in CO2 flux, which is unlikely to be fully captured by weekly or monthly seawater sampling." This passage is confuse. Please rewrite.

We will rephrase to "The rapid temporal fluctuations in CO2 flux are likely to be driven by variability in winds as well as variability in seawater pCO2. The latter is unlikely to be fully captured by weekly or monthly seawater sampling."

Fig. 4. CO2 flux from the Plymouth Sound sector appeared to be more positive than from the open water sector in some months. I would expect larger differences, but this is not the case. Could you explain this?

As shown in Fig. 10, pCO2 in the Plymouth Sound sector wasn't very different from pCO2 at L4. It seems that the direct influence of the river Tamar on pCO2 during this period was rather limited.

Lines 239-242: "Wind speed was generally higher at night during these few days and the measured fluxes imply that ΔpCO2 (see next sections on this calculation) changed from about -40 µatm during the day to about 15 µatm at night." Then, during this period, the system was a net sink of CO2?

The mean CO2 flux during these three days was small (about -2 mmol/m2/d).

Lines 283-285: "The greatest undersaturation in CO2 is observed in late spring and early summer, coinciding with an increase in chlorophyll a concentration at the nearby L4 station (Figure S6)." As I said before, I would like to see graph showing the chl a and the CO2 concentrations.

Please see above.

Lines 295-298: "The diurnal variability we observed is important in the context of estuarine CO2 (and carbonate system) observations that are predominantly carried out during daytime when sampling and navigation are easier." Many studies were published recently covering the diurnal (biological effect) and the semi-diurnal (tidal effect) variability on pCO2 changes, which are poorly described and constrained in the present manuscript.

We will rephrase this sentence to "The diurnal variability we observed is important in the context of the estuarine CO2 (and carbonate system) observations that are only made during the daytime." We will also add references of some recent studies that look at the diurnal and tidal-driven variability in pCO2 where appropriate.

Line 304: Spatial variability in seawater concentrations. Please see the general comments. This could be because the influence of Tamar estuary on the PPAO flux footprint is less in terms of pCO2 (e.g. due to the already large burden of carbonate and bicarbonate in seawater), and more on physical and biogeochemical processes. I am not sure about this statement.

We will rephrase this sentence to "pCO_2 measurements within both flux footprints were very similar to pCO_2 at the L4 station.", and remove the latter phrase.

Line 306: "dissolved pCO2." This is unusual. You should refer "pCO2" or "dissolved CO2".

We will rephrase to seawater pCO2.

Lines 329-331: "It could be that the large burden of carbonate and bicarbonate in seawater partially buffered the impact of the freshwater input on pCO2 within the flux footprints." This is poorly discussed.

We will remove this sentence as a similar point is already made at the end of section 3.4.

Lines 373- 377 "The implied pCO2 from EC fluxes in monthly bins and in situ measured pCO2 agree quite well over the annual cycle for the open water sector (Figure 7), suggesting that the use of the wind speed dependent transfer velocity parameterization of Nightingale et al. (2000) is largely reasonable." I am not sure that this type o graph is the best to show a comparison between estimated and measured pCO2.

We think this is a reasonable way to show both the seasonality in pCO2 and qualitatively compare the implied (=flux/K_Nightingale) against the measured pCO2. A more quantitative comparison between the implied and measured pCO2 is in essence made in the calculation of kCO2,660 (which is then compared against the Nightingale et al. 2000 K parameterization).

Figure 7. Y-axis : seawater pCO2 ?

Agree changing to seawater pCO2.