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Interactive comment on "Air–Sea Fluxes of Greenhouse Gases and Oxygen in the Northern Benguela Current Region During Upwelling Events" *by* Eric J. Morgan et al.

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Anonymous Referee 2

The manuscript by Morgan et al., discusses a new dataset of land-based observations of greenhouse gases (CO2, CH4, N2O) and oxygen (O2) from the Benguela upwelling region. These observations are used to estimate air-sea fluxes of these gases during upwelling events, which are then compared to traditional ship-based estimates, suggesting an overall agreement between the different methods. The flux reconstructions are also used to discuss the processes underlying the cycles of these gases in the upwelling system, and to argue that the region hosts a significant source of CH4, pre-

sumably related to sedimentary production.

The main finding of the paper is that during periods of upwelling, identified from satellite-based SST observations, land-based measurements of greenhouse gases show positive excursions of CO2, CH4, N2O (and possibly CO), and negative excursions in O2. These atmospheric concentrations are then translated with a simple transport model into air-sea fluxes of these gases, resulting into fluxes from the ocean to the atmosphere for greenhouse gases, and from the atmosphere to the ocean for O2. This picture is broadly consistent with our understanding of upwelling systems, and is supported by analysis of the stoichiometry of these fluxes, e.g. N2O:O2. The air-sea fluxes estimated by this top-down method are further compared with ship-based estimates for a particular upwelling event during which in-situ observations were collected, suggesting consistency between the (very different) approaches. This broad consistency is used to advocate for continues monitoring of greenhouse gas air-sea fluxes by land-based stations in this and other upwelling systems.

The Authors present a novel dataset of atmospheric measurements and a fairly through analysis that connect them to air-sea fluxes from the region. The focus of the paper are upwelling events, when the signature of air-sea gas exchange is particularly recognizable in the nearby atmosphere. Overall, the methods and type of data discussed are not completely new, but the application to the Benguela upwelling system is, and the Authors present an argument for the usefulness of this type of measurements. Upwelling systems are regions of enhanced exchange between the ocean interior and the atmosphere, and important components of global greenhouse gas budgets (e.g. N2O and CH4). Thus, the study connects to a topic of global relevance that should be of interest to a broader readership in oceanic/atmospheric biogeochemistry. The paper is overall well written, and the figures informative. The interpretation of the data is fairly clear and overall sound, and the paper in principle suitable for publication in Biogeosciences. That said, I have a several comments that I think should be addressed before the paper is ready for publication.

– We thank the reviewer for their thorough and thoughtful review of our manuscript.

The model used to estimate the air-sea flux densities (Section 2.4 and equation 3) is essential for the top-down estimates presented in the paper. While the model is fairly simple, it is not particularly well described and critically assessed (see specific comment below). In particular, no uncertainty estimate arising from the model itself is discussed or quantified. The only air-sea flux estimate uncertainties appear to be standard deviations from all estimates, but the model presumably introduces inherent uncertainty in each estimate, which should be quantified. In particular, assumptions on the mixing constant "q" may be particularly impactful.

– We agree with the reviewer's assessment. For the original paper, uncertainties for each term were calculated or estimated and propagated in quadrature sums; this is the source of the dotted lines on the top-down estimate in Figure 8. We will revisit these uncertainties and present them in a table with relevant statistics.

The Authors claim that the resulting top-down fluxes agree well with the in situ, ship-based estimates, even though in practice there's only one event for which the comparison is possible. However, the top-down estimates seem to be systematically higher than the ship-based estimates (e.g. in Fig. 8). While indicating "order-of magnitude" agreement at the peak outgassing (ingassing for O2), the figure points to a potential overestimate of the top-down method for all gases, especially if one integrates the fluxes over time (e.g. over the period indicated with the horizontal bar in Fig. 8). This is somewhat surprising, because the top-down method should integrate over broader regions with compensations between high and low fluxes. This discrepancy should be reconciled or more thoroughly discussed, but it feels minimized by the Authors (e.g. "good agreement" in the abstract and in few parts of the main text), which is a disservice to the readers. The Authors could be more nuanced with the discussion of this comparison, and more forward with the limitations of the approach, and discuss ways to address them, if the objective is a credible extension and application in future studies e.g. for monitoring and quantitative estimates.

– This is a good point; the method tends to overestimate fluxes as compared to the shipboard measurements, and because we can only identify upwelling evens with anomalies that are above the baseline, smaller events are not included in our average flux densities. We will broaden our discussion of the limitations of our approach.

Related to the point above, while the Authors are clear that the comparison is not a calibration of the top-down method, at some point such calibration will be needed to make the estimates quantitative and reliable, thus it would be useful if the Authors could add a discussion of the possible work and steps needed to turn this comparison exercise into a credible approach that could be used for monitoring. This type of discussion would strengthen the conclusions (page 13, lines 3-6), which right now feel somewhat superficial.

– This is a good suggestion. We will add some text that discusses necessary steps for continuous top-down monitoring.

The rationale for focusing the study on upwelling events could be better explained earlier in the paper. I assume it is related to the strength of the signal to be detected, stronger during upwelling, which allows a first demonstration of the method, but I may be wrong.

– The motivation was that we were interested in upwelling as a potential source of GHGs, and their episodic nature and distinctive signatures made it possible to estimate the flux without performing a full inversion of 3+ species from a single station. We have added this sentence to the introduction: "We focus on individual upwelling events as we expect them to be distinguishable from other sources of intraseasonal variability based their apparent stoichiometry in the atmosphere, and because there are relatively few observation-based studies from this region, relative to other EBUS."

In the same way, the study falls a bit short of fully connecting regional results to the big picture of greenhouse air-sea fluxes in upwelling systems. While it is interesting and valuable to provide air-sea flux estimates for all upwelling events in the region (Table 1), it would be even more relevant to couch these estimates into the big picture of air-sea fluxes for the region. For example, is outgassing of greenhouse gases during upwelling region important for total gas budgets? Could upwelling events be responsible for most of the outgassing, or is outgassing during non-upwelling periods also important? Of course, this would require comparison with other large-scale estimates for the region, and some degree of extrapolation/speculation, but it could add breath to the paper.

– We can provide rough estimates of the annual flux of each species from the study region, but this will require assuming a constant flux density and knowing the total area of upwelled water, simplifying assumptions which we can bracket with appropriate uncertainties, but will not be terribly robust. So, we will add this to the Discussion section, rather than present them in Results.

Page 2, line 9: why "yearly"? It seems that this approach could be applied to any timescale long enough to encompass the observations utilized. Please clarify or remove.

– We will remove.

Page 3, line 6 and following: I see the point of utilizing APO, however its introduction is somewhat abrupt and not every reader may be familiar with the concept and scope of it. I suggest a sentence or two to clarify and explain the usefulness of this tracer in the context of the paper (it is only affected by air-sea gas flux differences between O2 and CO2). A justification is presented later (e.g. page 9) but it could be more useful early on.

– We will revise the text introducing APO to read as follows: "In order to isolate the influence of air-sea exchanges on O_2/N_2, we have employed the use of a data-derived tracer, known as atmospheric potential oxygen (APO), which masks variations of O_2/N_2 that are due to terrestrial biosphere exchange (Stephens et al, 1998). Variations in APO are thus primarily due to fossil fuel burning and air-sea gas exchange of O_2. APO is defined as:"

Page 3, lines 19-25. I wonder if any consideration was given to including the direction of wind in the upwelling detection algorithm, since Ekman theory implies that only favorable wind directions (here equatorward and parallel to the coast) would induce upwelling. This could be clarified.

– We did not explicitly consider wind direction in our detection algorithm, but consideration was given to wind direction at the station (selection for sea breeze) and air mass origin (back trajectory filtering). These two steps together effectively filtered out all events which were not upwelling-favorable. Only upwelling-favorable winds would bring air to the station from these two upwelling cells, which is part of the reason for their selection. This is specified in the text now: "We selected this domain because it represented an area of the coast where strong upwelling occurs regularly (Demarcq et al, 2007), where this upwelling was spatially distinct from other upwelling cells reported in the literature (Lutjeharms and Meeuwis, 1987; Veitch et al, 2009), and where upwelling was downwind of the station during upwelling events."

Section 2.4. The model rationale, variables, uncertainty, and limitation should be discussed in better detail, as it form the basis for the top-down air-sea flux estimates. First of all, it is unclear what is solved for (I assume F) and how, e.g. based on what other quantities. Second, it is not clear how the back-trajectories were determined (there is no discussion of it that I could find) and how they are used in the model â ËŸAĔĞT I presume the variable "x" is the distance along these trajectories.

– We can add some text on the HYSPLIT model. That is correct, x is the distance along the trajectory. We will clarify this in the text.

The atmospheric boundary layer is assumed to be constant with a thickness "h", but this possibly varies substantially on a variety of temporal scales, e.g. going from the ocean to the land, and over the course of a day. Maybe variations in h can be folded into variations in the mixing rate q, but this rate is assumed to be constant, which is a big assumption. More critically, q is determined from equation (4), which presumably is a derivation of equation (3), although my sense is that it can be only derived if one assumes F=0 for the two gases, which is inaccurate. In equation (4) it is not clear what "t" represents and how it was determined (I suppose from x/U). The determination of "q" seems critical for the method, and it should be discussed in more detail, and results shown, for example of the determination of q for CO2, CH4, etc. Uncertainty in q could then be propagated into the model, or at least its effect on the flux estimates discussed.

– We will broaden this discussion in both the Methods and Results. One does need to assume F=0 for both gases, which is not accurate for the marine environment, but an acceptable assumption for the terrestrial component, as the terrain between the station and coast is devoid of vegetation or human settlement. In light of both reviewers' comments, we will compare our estimate of q to published values, and also conduct some simulations with a particle dispersion model to estimate q. We can then tune the q parameter to best match the shipboard measurements, and compare this with our estimates of q.

Page 6, line 12. The method by Lee et al. is somewhat outdated, and has been superseded by more recent approaches, e.g. the "LIAR" method, Carter et al., 2016, Limn. Ocean. Methods, although I suspect the alkalinity approximation is not a major source of error in the CO2 flux calculation.

– Thank you for bringing this to our attention. We can recalculate with the LIAR method.

Page 7, lines 6-15. The choice of piston velocity formulation seems hazardous, and needs some justification and perhaps clarifications. The Wanninkhof 1992 formulation has been superseded by a more recent one in Wanninkhof 2014, Limn. Ocean. Methods. The old formulation is biased too high by approximately 20% and should not be used. The Nightingale 2000 formulation is an odd choice because it was developed for the North Sea, and its range of validity is 3 to 14 m/s (wind speeds used in this paper can be smaller than that). The paper by Roobaert et al., 2017, Biogeosciences, provides useful guidelines for the choice of piston velocity that could be considered in

the study.

– We included the W92 because it is still widely in use, but are happy to replace it with W14. We thank the reviewer for bringing the Roobaert publication to our attention. We favored N00 because it was empirically derived in a coastal region, but we can include other k_w parameterization(s) that better encompasses the wind speeds of our flux event (2 - 15 m sec^{-1}). For the direct comparison of peak observed shipboard flux, winds were in the ca. 12-15 m sec^{-1} range.

Page 7, line 13, "kw and U10 must be in the same units, e.g. m sec-1": this is incorrect. With the coefficient reported, U10 must be expressed in m/s, and kw in cm/hour. Effectively, the coefficients have units, e.g. cm/h/(m2/s2) for the quadratic coefficient, etc.

– Thank you for bringing this to our attention, this was some text that got confused with k_w being in m sec^{-1} for Eq 7. We will change the text to read: "The Schmidt number is dimensionless, and U10 is in units of m sec^{-1}, producing k_w in units of cm hr^{-1}."

Page 8, lines 3-4: clarify the difference between "identifying" and "detecting" an upwelling event, otherwise the sentence is not clear.

– We will change the text to: "detection of an anomaly in the atmospheric time series", the distinction being that we identify upwelling events based on SST, wind speed, etc, but can only detect them in the atmospheric time series when atmospheric transport allowed.

Page 8, line 14: this sentence begins discussing an upwelling event, but then two are mentioned. Please clarify the time for the second event (presumably Dec. 4th)

– Text now reads: "4th of December, when SST dropped again. During these two low SST pulses, chl-a values were higher."

Page 9, line 24: please provide a reference or some context for the GENUS cruise.

– Will do.

Page 9, line 32: is this “synoptic event” also corresponding to an upwelling event?

– No, the upwelling event began after the synoptic event. We will clarify this in the text.

Page 9, line 34: “This coincides with . . .”. Please clarify this sentence; it is not clear what “this” refers to.

– We will clarify, "this" referred to the sporadic enhancements.

Page 13, line 3-6: “Based on our results . . .”. Measuring programs of this type are already in place in few regions, e.g. as part of the Advanced Global Atmospheric Gases Experiment (AGAGE). For example there are two land-based monitoring stations in the California upwelling region. It may be useful to expand this part of the conclusions to acknowledge existing observational programs and previous work, discussing what has been learned from them, and what is still missing (e.g. spatial coverage over other upwelling systems?). It would also be useful if the Authors could speculate on how far this type of measurements can go in order to provide truly quantitative estimates of air sea fluxes from coastal upwelling regions, since the paper only provides a proof of concept that still suffers from very large uncertainties.

– We can add discussion of existing stations, and deepen our discussion of top-down quantification of coastal marine fluxes using atmospheric data.

Page 5, line 26: “deviated” from what?

– The cavity pressure setpoint of 140 torr, we will clarify.

Figure 1, and Fig. 8: it would be useful to add to the maps a few geographic reference points, e.g. the town of Luderitz, which is mentioned multiple times in the manuscript, for the readers who are not familiar with the region.

– Will do.

Figure 1, left panels, and Fig. 4: please highlight the upwelling events as detected by the algorithm used, e.g. with vertical bars or shadings.

– Will do.

Figure 3: please highlight, the periods corresponding to the M99 cruise, and the upwelling events shown in Fig. 1, e.g. either at the top/bottom of the figure, or using bar shadings of a different color. Why is chl-a shown as dots instead of as a continuous line?

– Will do. We can change the chl-a to a line, I think this plotting choice was made because there were more outliers than the other time series, and it made the plot harder to read with the shading.

Figure 5: please clarify the duration of the back trajectory periods.

– Will do (5 days).

Figure 8, right panel: it would be useful to mark the days on the cruise track, to allow a comparison with the left panels.

– We can add these as points.

Figure 9, left panels: the gray symbols are very hard to see on the purple background, please use a different color (e.g. darker). At the end of the caption: “correspond to” instead of “correspond with”.

– Will do.

Please clarify early in the paper what “bottom-up” and “top-down” estimates refer to. This terminology in the specific context of the paper may not be clear to every reader. E.g. in Fig. 8, the Authors could add a clarification on “bottom-up” (ship-based) and “top-down” (land-based).

– We can clarify this in the text.

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