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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 1

Morgan et al. present an evaluation of using ground-based measurements of gas concentrations to estimate air-sea gas fluxes in coastal waters off the northern Benguela upwelling region. This approach can provide greater resolution than ship-based sampling during upwelling events, and thus represents a valuable tool for monitoring air-sea gas flux in coastal upwelling areas, which are important sites for emitting greenhouse gases such as N2O and CH4 to the atmosphere, but may be subject to seasonal and short term variability that may be missed by infrequent ship-based sampling efforts.

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The authors validate their method by comparing ground-based flux estimates to those estimated from ship-based sampling, which is an important step. The manuscript goes on to discuss the sources of variability and error in their data, and ways in which data must be filtered to minimize error. The authors ultimately suggest that tower-based flux montoring systems should be widely adopted to measure air-sea flux in eastern boundary upwelling systems. This work thus represents an important contribution to coastal marine air-sea flux studies, and should be published with minor revisions. The presentation quality and writing is excellent, and I have only a few minor comments/questions that need to be addressed (see below).

– We would like to thank the referee for their time and constructive review of our manuscript. We include our responses to their comments below.

While I agree that this presents an important contribution and should be widely adopted, I can't help but struggle with the fact that the towers only appear to be used to estimate fluxes during upwelling events, and may be being under-utilized. Could the authors briefly discuss other potential benefits of these towers? E.g. could they be used to measure downward fluxes as well? Or perhaps measuring land-air fluxes? These towers might also naturally be compatible with eddy-covariance flux estimates.

– The reviewer brings up an important point to clarify, which we will address in the Introduction and Conclusion. We focus on upwelling events because they are detectable in the atmospheric record due to their distinct tracer-tracer relationships. The tower data can be used to quantify regional, time-varying surface fluxes for both land and ocean, but this would ultimately require a Bayesian atmospheric inversion, which is beyond the scope of our study. We will add this text 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."

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Specific Comments:

Page 2, Line 32 – can you give the approximate frequency at which measurements were taken? What standards were used, and how frequently the instruments were calibrated?

– Such details were omitted for brevity, as they were published elsewhere, but we will add the requested information.

Page 3, line 4 – perhaps include a brief explanation for presenting atmospheric oxygen relative to N2? What is this correcting for?

– We will add the clarification that we are following a standard convention here. There are several reasons to report the O_2/N_2 ratio instead of the absolute mole fraction, one being that O_2 is not a trace gas, and the mole fraction of O_2 will vary due to the addition or subtraction of other trace gases in a given parcel of air to a non-negligible degree. A common thought experiment demonstrating this: in a parcel of air containing 1e6 molecules of air, 410 of which are CO_2 and 209,392 of which are O_2, the mole fractions will be 410 ppm and 0.209392, respectively. Adding one molecule of CO_2 to the parcel changes the mole fraction by 0.999589 ppm for CO_2 but 0.2093918 ppm for O_2.

Page 3, line 7 – remove one of the 'as' (there are two in a row)

– We will remove.

Page 3 line 16 – Why not include areas further off-shelf? Perhaps a justification for selecting these boundaries would be helpful? i.e. it's upstream of your tower in the direction of prevailing winds during upwelling? Or perhaps it's based on SST anomalies during upwelling events?

– We will add the following text: "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

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was downwind of the station during upwelling events. These criteria were considered desirable because they would provide the best opportunities for relating atmospheric anomalies to upwelling events. These determinations were based on analysis of our SST dataset, and atmospheric back-trajectories simulated with the HYSPLIT model." In addition, we have added a brief description of the HYSPLIT model, for completeness.

Page 3, line 19 – how were wind-speeds and SSTs determined? Update: I see this is discussed in the subsequent section. It's nice to understand where the data come from before you explain how the data was used, so I suggest moving the discussion on Remote Sensing Data ahead of this section. But this is only a suggestion.

– We will move the section up.

Page 3 – line 19-25 – I understand that you chose the upwelling thresholds by visually inspecting the data, but perhaps you could clarify how 'extreme' the thresholds you chose were relative to the standard deviation from your smoothed SST or wind-speed curves? Or perhaps you could state how many upwelling events were flagged during this step before filtering them further based on the SST, atmospheric data, back-trajectories of winds, and CO concentration?

– We will add standard deviations of both anomaly time series to the Methods section. We will also add additional details about the number of filtered/excluded of events, and move all of that text to the Results section, since this seems a more appropriate place for it.

Page 5 section 2.4. I'm curious about the dilution factor calculation – q. This seems straightforward, but I'm sure there are complications that may be being glossed over, and I have a few questions that I feel should be addressed in the text. How many sets of data points from the ship and tower were used to derive the average q? What is meant by 'filtered to exclude for poor agreement between CO2 and CH4'? and what percentage of the potentially viable measurements were excluded from the average because of this? And finally, how does the q you derived compare with other estimates?

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– We agree this needs more discussion and clarification. The dilution factor q is probably the least constrained and well-known of all the model parameters. Since there were high-resolution atmospheric measurements of CO_2 and CH4 on the Meteor cruise, either species could be used to estimate a non-species dependent dilution factor. If the difference of q^{CO_2} and q^{CH_4} for a given comparison of was > $\pm$ 0.01, they were excluded from the average. This reduced the total n from 32 to 13. Another way to estimate q would be to look at particle dispersion rates in a Lagrangian Particle Dispersion Model (LPDM), which we can include in a revised manuscript. Either way, we will increase the uncertainty bounds on this parameter. We will also add some discussion of other published estimates, e.g. Price et al 2004 (JGR Atmos., 109, 23), who used multiple species and a model approach to arrive at a mean q of 0.010 $\pm$ 0.004 hr^{-1}, which is nearly identical to our value of 0.011 $\pm$ 0.006 hr^{-1}, although the spatial and temporal scale they considered was larger. Dillon et al 2002 (JGR Atmos., 107, D5) observed rates that were higher for a Sacremento pollution plume, 0.2 hr^{-1}. We will also broaden the discussion to include using q as a tuning parameter for the model – i.e., what value of q would best fit the shipboard flux estimates if all other model parameters were equal, and then compare this to our experimentally determined value of q, which is a conceptually different approach to the puff model. This would mean that the comparison to the shipboard estimate is approached on a different basis, i.e. as a means for improving the model rather than validating it.

Page 8 line 32 to end of para – you suggest that upwelling fluxes probably account for the majority of the mean annual flux (as reported by others). Can you support this claim by calculating how many upwelling events similar to the ones you observed (i.e. flux density x duration of UW event) would be needed to account for the annual flux assuming the fluxes the rest of the year were net-zero?

– We can provide a rough estimate of the total annual flux due to upwelling events for each species, by making some broad assumptions on the area of upwelled water, and then compare these flux totals, but we would like to restrict this to the Discussion, as

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the results are not that robust, due to the simplifying assumptions.

Page 9 line 3-6 – Good! I was wondering about how different O2/N2 vs. APO would be. Thank you for including this. . . , but where are the actual comparison between dO2/N2 and APO shown? There isn't a reference to a figure or table here, nor a slope and r2 value, or even a mean difference between the two.

– We hesitate to add another figure (to keep the total number from becoming overly high), since it only shows a strong linear correlation between the two flux estimates, but we can add some text/statistics on the comparison: "A linear regression between the two estimated flux densities yielded a slope of 0.91 and a coefficient of determination of R^2 = 0.98. The estimated APO flux density was 4.5% lower on average than the $\delta(O_2/N_2)$-inferred flux density."

Page 9 line 10 – can you include the range here as well? It would be useful to know the maximum flux density you measured to compare with the others' estimates mentioned later in the paragraph.

– This will be added to the revised manuscript.

Figure 1 – can you please show the locations of Walvis Bay and Luderitz on the maps, since you refer to them in the text?

– Yes, this will be added to the revised manuscript.

Page 9 line 25 – what were the dissolved CH4 concentrations in the upper 15m during this cruise? How do they compare with the other concentration values reported in the same paragraph?

– We will add the text, "from dissolved concentrations ranging from 6.0 to 140 nM." FYI we have identified 6 other samples from the MEMENTO database, bringing the total number to 9, though the range reported hasn't changed.

Page 10, line 1 and 2. This sentence seems to contradict the second sentence of

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the paragraph, which states you observed elevated CH4 mixing ratios. Something is unclear here.

– Thank you for pointing this out, we will clarify by adding “apart from the initial synoptic event”.

Page 10, line 18 – ‘likely a result of warming temperatures that would reduce their solubility’ are you suggesting the only reason the gases evaded to the atmosphere was warming? This implies CH4 and CO2 weren’t supersaturated before the water warmed up. . .is that true? More likely the net evasion was enhanced by warming, except for O2.

– We are not saying that the only thing influence the flux of the gases was warming, but that generally we would assume that a positive flux ratio between O_2 and CO_2 is a result of thermal processes dominating the surface flux, since net respiration/ventilation of aged waters or net productivity would produce a negative flux ratio. Given that it was strong enough to reverse the flux of O_2 relative to the rest of the filament, this interpretation seems to us to be justified. We will add the text, “The positive flux ratio of O_2:CO_2 is generally only produced when thermal processes dominate the air-sea flux.”

Conclusion – Might be worth emphasizing the fact that your tower method is capable of measuring methane flux associated with both bubble flux and diffusion, which can’t be said of ship-based measurements.

– This is an excellent point, although we are hesitant to include such a statement, since the uncertainties associated with our method are high.

As previously mentioned, I agreed that a network of these towers would be great, but it might be easier to sell the idea of a network of these towers by mentioning the other potential uses of these towers. E.g. can you not estimate air-sea fluxes during down-welling events? What about land-air fluxes when winds from the east? You already mentioned the filtering for biomass burning based on CO, so surely there is some

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value here as well?

– Thanks for pointing this out, we will adjust the concluding text, similar to our first response above, by adding the following sentences: “We have focused here on upwelling events, because they are distinguishable from other sources of intraseasonal variability in the atmospheric record. A full top-down accounting of the greenhouse gas budget of the the Benguela could be accomplished through a Bayesian atmospheric inversion of one or more coastal stations.”

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