

Authors response to Referee #1

We thank the referee for reviewing the manuscript (ms) and for his/her constructive comments. Following recommendations from the two referees, we did a substantial rewrite of the Results and Discussion sections. The discussion was strengthened, new figures were included and complementary information was added as supplementary material. Below, we respond specifically to the *Referee's Comments (RC, in italic)*.

RC1: This manuscript by Lo Monaco et al. presents results of a comprehensive field campaign studying a rather spectacular annual dynamic: the CO₂-sink induced by the spring bloom 'downstream' of Kerguelen Island. This phenomenon has received attention in earlier literature. Although this recent contribution may not provide unexpected results, I believe that it merits publication on basis of the mentioned comprehensiveness of the underlying dataset and the main result of the paper, namely that the annually integrated CO₂ drawdown by the naturally Fe-enriched water downstream of Kerguelen is some 3x higher than that of non-Fe enriched, but otherwise comparable waters.

We do not agree with the referee's feeling that the ms presents unsurprising results. Indeed, the KEOPS2 observations allowed for a better understanding of the physical and biogeochemical processes at play in the different regions of the bloom area at the start of the productive season (when very few fCO₂ observations were available before, only one cruise in Oct-2005 for surface measurements in SOCAT; www.socat.info). In this context, we found that the magnitude of the air-sea CO₂ flux was not related to the distance from the iron sources (as one could naively expect). Instead we show that fCO₂ spatio-temporal variations (and consequently air-sea CO₂ fluxes) are strongly influenced by lateral advection and vertical mixing. Due to different physical and biogeochemical settings, one could expect differences in the uptake of atmospheric CO₂ within the large bloom area, but our synthesis of observations indicates a relatively similar uptake of CO₂ over the productive season. This said, we agree that the objective of the paper is not to provide unexpected results, but rather to present a new dataset that allowed to discuss for the first time the seasonal evolution of air-sea CO₂ fluxes in Kerguelen's bloom in comparison to HNLC waters.

Main critique

RC2: Having said that, I believe that a fairly substantial rewrite of the manuscript will be required. Currently, I believe the manuscript to be too descriptive – even confusingly so. While the overall picture of the onset and evolution of the bloom-induced CO₂-sink is intriguing and (after repeated reading) coherent, that picture is lost in a rather bland and winding narrative of exacting observations of pCO₂ that is interspersed with details on nutrients, DIC and Chl.a, without tying most of these together quantitatively. Such quantitative discussion is not obligatory, but there should exist a balance in the precision with which related properties are discussed. I elaborate below.

Moreover, the paper suffers from inferences and attributions being made that do not appear fully backed by the data as presented, but rather seem to have to be taken at face value or come from earlier literature (for example, the suggested 'silicic acid depletion', 'strong lateral injection of iron'). Related criticism is that the manuscript lacks appreciable embedding in wider literature – perhaps with better referencing, the stated case would be more convincing.

If this paper is to find an audience, the authors are challenged to filter out superfluous content, to strongly shorten the descriptive sections and, ideally, to provide a means of (visually) summarizing the field results in a way that clearly illustrates the processes inferred.

Following the referee's main critique, we merged the results and discussion sections in order to avoid unnecessary descriptions, and appropriate references were included in the discussion. The mechanisms driving variations in surface fCO₂ in the different regions are now discussed in a dedicated section, and a summary of the main findings (including a synthesis of drivers) was added at the end of the ms.

RC3: In the 5+ pages of discussion, only three references are used (Qu  rou   et al. 2014, Park et al., 2014, Metzl et al 2006), all of which have been used in the text before. That is rather thin, especially in light of the large amount of hydrographical information presented (is that all from Park et al?) in the first paragraph.

More references have been added to the discussion. Regarding the hydrographical context, a lot of work was done by Park and collaborators in this region. The paper by Park et al. (2014) gives a synthesis of their work. However, we added more references in the revised ms, notably to the study by d'Ovidio et al. (2015, BGD) who investigated the small scale dynamics and the transport of iron at the surface based on altimetry.

RC4: Despite extensive invocation of iron (non-)availability to explain observed phenomena (from the summary: "Our results show that the magnitude of the CO₂ sink at the start of the productive season is closely related to chlorophyll a concentration and iron availability"), no actual concentrations of iron are mentioned anywhere in text, tables or figures. There is a reference to Qu  rou   et al., who provide the Fe measurement data for this cruise. Where relevant, however, Fe values should be explicitly mentioned in this manuscript.

Iron concentration values from Qu  rou   et al. were added in the revised ms.

RC5: Treatment of DIC and ALK measurement quality is inappropriate (although likely not relevant to conclusions of the manuscript). The statement "Accuracy estimated from the CRMs analysis was $\pm 4 \mu\text{mol kg}^{-1}$ for both TA and TCO₂." does not make much sense. Analysis of CRMs would only yield precision (i.e., "our CRM results averaged to $2150 \pm 3.8 \mu\text{mol/kg}$, $n=50$ "). CRM analysis does not give an estimate of accuracy beyond, for example, "our CRM average of 2150 ± 3.8 is about $11 \mu\text{mol/kg}$ lower than the certified value of 2161.1 ± 1.5 ". If ones corrects his measurements to CRM, he can't do much more than saying "we assured accuracy by correcting measurements to CRM results. Precision is $\pm 3.8 \mu\text{mol/kg}$." (same inappropriate use of "accuracy" for the subsequent discussion of nutrients).

CRMs are used to correct TA and TCO₂ measurements. We changed accuracy for precision in the revised ms. The quality of the measurements was confirmed by comparing the data to previous datasets (in the deep waters where concentrations remained stable during the last two decades).

RC6: The fact that pCO₂, DIC and Talk were all measured begs for a sentence comparing measured fCO₂ to fCO₂ calculated from DIC and ALK.

The comparison between calculated and measured fCO₂ does not give any information about the mechanisms that drive variations in fCO₂, and for this reason we believe it is beyond the scope of our study. Such a comparison was previously evaluated, for example by McNeil et al. (2007) because these authors then used reconstructed TA and TCO₂ to evaluate air-sea CO₂ fluxes in the Southern Ocean. Using OISO data (for both summer and winter cruises,

and at the large scale), the mean difference between measured $f\text{CO}_2$ and calculated $f\text{CO}_2$ was $3.6 (\pm 3) \mu\text{atm}$ (Fig. 4 in Mc Neil et al., 2007).

For the present study, we performed, as usual, a careful quality control on $f\text{CO}_2$, SST, SSS, TCO_2 and TA data to ensure that they are accurate. This includes comparison with previous measurements in deep waters for TCO_2 and TA, and comparison of ship-based and land-based atmospheric CO_2 , as well as inter-comparison of SST from 3 different sensors for $f\text{CO}_2$.

RC7: The paper repeatedly (and perhaps inappropriately, see below) relates Chl.a observations directly to $f\text{CO}_2$ observations. Discussing these relationships with respect to 'intermediate' property DIC would certainly benefit the conceptual accuracy of the paper. I am slightly wondered by the only very modest role of the (supposedly) high-accuracy DIC measurements in the manuscript.

We added figures showing the distribution of TCO_2 and TA, and we discuss the impact of their variations on $f\text{CO}_2$. We also show and discuss the relationship between TCO_2 and nutrients.

RC12: In the very last lines of the conclusions, the authors suddenly present an estimate of the hypothetical CO_2 uptake of a fully 'geo-engineered' or iron-enriched ice-free Southern Ocean. The manuscript would benefit if this finding is provided with some context (comparison with earlier 'naïve' estimates, Fe-C stoichiometry, relevance to geo-engineering proposals). This hypothesizing about 'iron-enrichment' of the whole sea-ice free Southern Ocean is meaningless without explicitly addressing the differences between it and the Kerguelen system – it is not just the iron concentration that differs, but conceivably also bathymetry, vertical mixing, frontal structure, etc. Be specific even when speculating. However, again, I would recommend dropping the geo-engineering angle entirely.

We extended the discussion of the seasonal evolution of air-sea CO_2 fluxes and their drivers in Kerguelen's bloom, including comparison with previous work on iron fertilization. The discussion explicitly addresses the differences between various regions of the Southern Ocean

RC13: I am generally skeptical of the occasionally sloppy oceanographic reasoning and 'explaining' at various stages of the manuscript, and would I recommend the authors to thoroughly comb the article for flawed reasoning, ambiguous statements, vague referencing and ad hoc explanations. Three examples follow.

(a) The authors repeatedly invoke "Takahishi's (1993) temperature effect" to "explain" parts of $f\text{CO}_2$ variability. While it is unequivocally true that heating a parcel of water increases its $f\text{CO}_2$, it does **not** make sense to say "two distinct parcels of water have different temperatures and because of that their $f\text{CO}_2$ s are different". However, that is exactly the reasoning evident in (for example) page 17552, line 16: "The passage of the PF was marked by a rapid change in surface temperature from 2.5 to 3.9°C between 47.5 and 47.4°S (Figs. 2 and 3). This resulted in a rapid increase in surface $f\text{CO}_2$ from near-equilibrium values south of the PF ($387 \pm 7 \mu\text{atm}$) to a small source of CO_2 north of the PF ($406 \pm 12 \mu\text{atm}$), which can be explained by the temperature effect alone (+22 μatm)". That is just page filler. For all anyone knows, the warm water to the north of the PF could have been on a southward trajectory and cooling. This is possibly the most serious example of inappropriate use of that "temperature effect". However, there are several other occasions where it seems to be invoked to 'explain' things that it can't.

(b) Page 17552, line 7: "However, since the contribution of northern waters is

expected to reduce surface TCO₂ concentrations in the Plume compared to the cold southern waters, we conclude that lateral advection has a minor impact on surface fCO₂ (due to the opposite effects of temperature and TCO₂)." This may be true but is too easily stated. Consider making this quantitative: given typical northern and southern watertypes (S,T,ALK,DIC), what is the range of pCO₂s you get for the mixing from 0% to 100% AASW? "Due to opposing effects, no bigger change than XXX is expected based on calculations of YYY".

(c) Similarly, page 17555, line 17: "According to Takahashi et al. (1993), the temperature effect [of -0.2°C cooling] explains about 25% of the sudden decrease [by 13 uatm] in surface fCO₂. The remaining 75 % is attributed to enhanced photosynthesis. Indeed, water column measurements revealed that TCO₂ concentrations were about 15 μmol kg⁻¹ lower at the surface than in the Winter Water". The question should be: "what was the decrease in DIC between the two visits to the location"? Is that decrease consistent with the "unexplained" 75% of the 13 uatm drop between the two visits?

The revised ms was corrected for unappropriate reasoning, and better referencing was included in the discussion. The relative contributions of changes in temperature and TCO₂ on surface fCO₂ was evaluated more cautiously. We were also careful to avoid shortcuts, being more specific in our reasoning.

RC14: To formalize the assessment of fCO₂ results a bit, it would not be inappropriate to provide an accurate formulation of the T/pCO₂ dependency rather than citing Takahashi's 1993 general estimate of that. The same holds for the pCO₂/DIC dependency (~2-4 uatm per umol/kg depending on pCO₂ and T), which currently is used only qualitatively.

Following the referee's suggestion, we quantified the relative contributions of changes in SST and TCO₂ (based on local conditions) to explain the changes observed in fCO₂. Values range from 12 to 18 μatm/°C and from 2 to 2.5 μatm/μmol/kg, respectively.

RC15: Page 17554, line 12: "[...] the increase to near-equilibrium observed on the 18 November was due to the combined effect of surface warming by ~0.5°C [...] and reduced chlorophyll a concentrations." Reductions in Chl.a. of course do not directly cause an increase in fCO₂. At best, one could suggest that both the Chl.a. decrease and the fCO₂ increase are both results of deep mixing, or very rapid remineralization of formerly alive organisms, or what have you, but saying one caused the other is simply not correct. The paper should be checked for the various other instances of such sloppiness (e.g., "Surface fCO₂ decreased by ~90 μatm over the 3-weeks period in response to an increase in chlorophyll a concentrations by a factor [of] 10").

We corrected for unappropriated shortcuts by being more specific in our reasoning (notably, by including TCO₂ and nutrients in the discussion).

Some minor points (again, in no particular order):

RC16: The very first sentence of the paper already contains an error (or placeholder value?) in stating that the Southern Ocean takes up "1" PgC/year. However, both cited references give values well below that (~0.4 and ~0.05, even). Please check.

We kept the value of 1 PgC/yr given by Takahashi et al. (2012) for the Southern Ocean south of 30°S (the reference was corrected in the revised ms, and the limit at 30°S was mentioned). A value of less than 0.1 PgC/yr would refer to the Antarctic Ocean, i.e. south of 50°S (Polar Front). Since the value for the CO₂ uptake strongly depends on how one defines

the Southern Ocean (see for example Figure 5 in Lenton et al., 2013), it is indeed important to indicate the latitudinal band associated to such estimate.

RC17: *Check section numbering (e.g., for wind data you refer to section 2.2, which does exist).*

Section numbering was corrected.

RC18: *17547, line 3-5. You didn't use satellite data to compute wind speeds – you downloaded that product, as detailed in section 2.1.1.*

In the revised ms, we changed the wind product for the MERRA reanalysis because we found a good agreement between the 3-hourly MERRA wind speed and the ship-based measurements in October and November 2011. MERRA winds are presented and discussed in supplementary material.

RC19: *17549, line 18. Replace "(CO2 source)" – which doesn't help – with "(i.e., the ocean is a CO2 source)". Similar for line 19.*

This was corrected.

RC20: *17561, line 8: "[...] the onset of the CO2 sink associated with Kerguelen's blooms seems to depend essentially on vertical mixing". You mean "seems to depend on the **reduction of vertical mixing in spring**" or similar. Please be specific*

This was corrected.

RC21: *I'm not sure one can say "fCO2 drawdown". Drawdown I associate with a mass flux, not a pressure change, i.e., "fCO2 decrease" or "CO2 drawdown".*

This was corrected.

RC22: *Latter half of 2.1.1. (comparison of pCO2atm with Amsterdam Island belongs to the methods section, not the flux calculation section.*

We moved the text to the previous subsection (Sampling and measurement techniques).

RC23: *First bit of "Sampling strategy and meas. tech." really belongs in introduction.*

Part of the text was moved to the Introduction section.

RC24: *17548, line 8: "All fCO2 values presented here are normalized to 1013hPa". I do not understand what that means. Did you calculate ΔpCO_2 assuming P_{atm} was 1 Atm? Something else? How does this procedure (shortcut?) affect your data and results? After reading repeatedly, it is not clear to me if you calculated pCO_{2atm} as $xCO_{2atm} * P_{atm}$ (i.e., pCO_{2atm} varies with atmospheric pressure) or as $pCO_{2atm} = xCO_2 * 1$ (i.e., pCO_2 constant despite atmospheric pressure variation). The former is correct, the latter*

likely is more appropriate given long timescale of exchange. Please be explicit about this.

fCO₂ normalized to 1013 hPa (fCO₂@1013) was calculated as follows:

$$fCO_2@1013 = xCO_2 * 1013 / P_{atm}$$

Both the oceanic and atmospheric fCO₂ were referenced to 1013 hPa, thus ΔfCO₂(sea-air) is similar with or without referencing.

fCO₂ data were normalized to 1013 hPa in order to correct for changes in atmospheric pressure, so that oceanic fCO₂ variations are only driven by SST, TCO₂ and TA.

RC25: *It is not clear to me what the relevance of ALK is for this study (except if the authors would use that to calculate pCO₂-DIC dependencies, which they currently do not).*

We added a figure showing the distribution of TA, and we discuss how it influences fCO₂. In addition, we used TA to evaluate the dependency of fCO₂ to changes in TCO₂ and SST.

RC26: *Hitaschi -> Hitachi*

This was corrected.

RC27: *Have the authors attempt to use the discrete-samples' HPLC chl.a measurements for calibration the underway fluorescence? (possibly an even worse r₂, but worth mentioning)*

The relationship is not better (nor worse) when HPLC measurements are used. The relationship does not improve either when using only night data (in order to avoid the quenching effect). Better relationships were obtained at the regional scale, notably in the PF zone: r²=0.99). We used the regional relationships to calibrate fluorescence data that are used in two occasions in Table 1 (when no Chl-a data was available).

RC28: *17549, line 6: "Here we present measurements collected at the station A3 situated over the Plateau in the core of the southern bloom investigated during the first KEOPS survey, and which was revisited twice during the KEOPS2 survey and again during the following OISO cruises". What do you mean "Here"? You present much more data than that, right? Or is the whole region referred to as "A3"?*

Measurements were collected in the water column at various stations in the bloom area. In the ms, we presented water column data from only one of those stations, A3, because observations later in the productive season were also available there. This was rephrased in the revised ms

RC29: *Upon first mention of the CARIOCA buoy, add reference to Merlivat's paper for same S.I.*

The reference for the CARIOCA buoy was corrected.

RC30: *17555, line 22. The term "biological pump" generally is used to describe the transfer of C out of the euphotic layer into the abyssal ocean. Its use here to 'biological uptake of CO₂' is not warranted. That CO₂ may well be released rather than exported.*

We agree. This was corrected in the revised ms.

References

- Lenton, A., B. Tilbrook, R. M. Law, D. Bakker, S. C. Doney, N. Gruber, M. Ishii, M. Hoppema, N. S. Lovenduski, R. J. Matear, B. I. McNeil, N. Metzl, S. E. Mikaloff Fletcher, P. M. S. Monteiro, C. Rödenbeck, C. Sweeney, and T. Takahashi (2013). Sea-air CO₂ fluxes in the Southern Ocean for the period 1990-2009. *Biogeosciences*, 10, 4037-4054.
- McNeil, B. I., N. Metzl, R. M. Key, R. J. Matear, and A. Corbiere (2007), An empirical estimate of the Southern Ocean air-sea CO₂ flux, *Global Biogeochem. Cycles*, 21, GB3011, doi:10.1029/2007GB002991.