Dear reviewers,

We appreciate your constructive comments. In the following, we provide a detailed response to each of your comments with bold fonts.

**Reviewer #1:**

This paper combines $pCO_2$ measurements made on a cruise in the eastern equatorial Indian Ocean in May 2012 and compares them to $pCO_2$ observations made in the same general area over the last 50 years. While I did not find any fatal flaws in the manuscript, I was rather unimpressed with the level of effort and scientific work that went into this paper. It basically describes changes in surface $pCO_2$ that are fully expected and predictable.

**Re: We will highlight the implications of this manuscript in the revised version, especially in the Introduction section. In fact, this manuscript has implications in better understanding global carbon cycle and in promoting studies on variations in carbon sink and ocean acidification (OA):**

**(1) Probably due to geopolitical factors, observation-based studies on ocean $pCO_2$ change and its mechanism as well as it influence on ocean carbon sink and OA are less in the Indian Ocean compared to the Pacific and Atlantic (Lenton et al., 2012; Fay and McKinley, 2013; Takahashi et al., 2009), except for the sparse study e.g., by Metzl (2009) in the south-western Indian Ocean. In addition, the Indian Ocean, which is surrounded by land in the north and lacks steady equatorial easterlies in the equatorial belt, is different from the other oceans (Valsala et al., 2012), and no study is found on $CO_2$ change in the equatorial Indian Ocean. Thus, this manuscript, which reported the ocean $pCO_2$ increase based on observed data, explored the possible factors responsible for $pCO_2$ increase and discussed the influence of $pCO_2$ increase on carbon flux and OA in the eastern equatorial Indian Ocean for the first time, will provide a better understanding of the carbon cycle in the Indian Ocean and may get more attractions for the Indian Ocean in the future.**

**(2) Although ocean $pCO_2$ increase due to quickly rising atmospheric $CO_2$ levels is expected, its increase rate varies geographically due to the modulation by local**

**oceanographic processes (e.g. lateral mixing, upwelling and biological activities) (Takahashi et al., 2006). An extreme example is that sea surface $pCO_2$ in the vicinity of the Bering and Okhotsk Seas decreased during 1970-2004 (Takahashi et al., 2006), as atmospheric $CO_2$ level quickly rose. Thus, investigating the long-term trend of ocean $pCO_2$ in this region is of much importance in precisely determining global ocean carbon sink (Takahashi et al., 2006).**

**(3) Surface pH in the Indian Ocean is relatively low compared to other oceans, and the Indian Ocean is more susceptible to OA (Takahashi and Sutherland, 2013). However, by now little information on OA is available in the Indian Ocean, although the Bay of Bengal Ocean Acidification (BOBOA) moored buoy was deployed on 23 November 2013 (http://www.pmel.noaa.gov/co2/story/BOBOA). Furthermore, coral has wide distributions in the Indian Ocean (Allen and Adrim, 2003) and is sensitive to OA (Orr et al., 2005). Thus, discussion on OA in this manuscript is scientifically full of significance.**

**(4) This manuscript attempted to investigate the trend of ocean $pCO_2$ change by using data that can be available, and suggested big change in ocean $pCO_2$, $CO_2$ flux and carbon species (OA), which are new findings for the equatorial Indian Ocean. Although the limited data is available, which does not allow us to discuss the interannual variability and the influence of climate events, it will not affect the long-term trend of $pCO_2$ change given that climate signals will fade away when timescales lengthen (Fay and McKinley, 2013). More important, this manuscript will attract more attentions for the Indian Ocean in the future.**

For the most part, the attempt to attribute the changes to different mechanisms was qualitative and the authors readily acknowledge the limitations based on the paucity of data.

**Re: In the revised version, we first identify quantitatively the contribution of temperature, salinity, dissolved inorganic carbon (DIC) and total alkalinity (TA) to sea surface $pCO_2$ increase and then find out several candidates for DIC increase (we find DIC has the biggest contribution), which provides in-depth discussion on the mechanism of $pCO_2$ increase.**

**Yes, the limited data indeed did not allow us to discuss the interannual variability and the influence of climate events on ocean $pCO_2$ and we have stated this point clearly in the manuscript and stressed that more $CO_2$ study is need in the future in this region. However, it is robust to use these data to determine the mean trend of $pCO_2$ change in this region.**

I saw no discussion of the calibration or any attempt to verify the accuracy of the 2012 data. Were there any discrete carbon samples collected to validate the accuracy? Did the methods of analysis follow the best practices handbook (Dickson et al., 2007; http://cdiac.ornl.gov/oceans/Handbook_2007.html)?

**Re: We will add the discussion of the calibration and the accuracy of the 2012 data, especially the in-situ calibration of the data, although this work was presented in detail by Fietzek et al. (2013). Fietzek et al. (2013) asserted that "the average difference between sensor and reference $pCO_2$ was -0.6 ±3.0 µatm with an RMSE of 3.7 µatm". And our in-situ discrete carbon samples indicate the accuracy better than 5 µatm.**

The discussion of the drop in pH was very elementary and added very little to the paper. I would have liked to have seen a much more in depth analysis of ocean acidification in this area and how it is affecting the local marine ecosystem.

**Re: Yes, that is the point. In the revised manuscript, we will conduct an in-depth analysis of OA, e.g., quantify the contribution of temperature, salinity, TA and DIC to pH drop and discuss the influence of OA on local marine ecosystems. However, specific study of OA on marine ecosystem physiologically will be not discussed in this manuscript, considering that this is beyond the scope of this manuscript.**

I also note that there were a number of grammatical errors in the manuscript that should be corrected. Perhaps the authors can find a native English speaker to help proofread the text.

**Re: Thanks for your suggestions. In the revised manuscript, we will correct the grammatical errors and polish the language of the manuscript with the help of a**

**native English speaker.**

In summary, I have no major concerns if the manuscript is published after the grammar is corrected, but I do not think that this work significantly advances the science. I would prefer if the authors went back and thought a little harder about what interesting science could come from this work than to just say that rising atmospheric $CO_2$ and increasing temperature are causing the surface water $CO_2$ to go up and the pH to go down. I recommend major revisions.

**Re: Thanks for your comments. Major revisions have been made in the revised manuscript based on your suggestions, e.g., (1) clarify and highlight the implications of this manuscript in science in the Introduction section, (2) identify quantitatively the contribution of temperature, salinity, DIC carbon and TA on $pCO_2$ increase, and (3) conducting in-depth analysis of the influence of $pCO_2$ increase on air-sea $CO_2$ flux and OA. We hope that you will find this manuscript will be more impressive and do some contributions scientifically after major revision, in combination with the background of carbon cycle in the Indian Ocean.**

**Reviewer #2:**

In the manuscript Xue et al uses recent measurements of oceanic $pCO_2$ collected in the east equatorial Indian Ocean during 2012 and the database of Takahashi et al to explore the interannual variability in oceanic $pCO_2$ and quantify the changes in ocean acidification (OA). Overall I found this paper quite disappointing, it is inconsistent in sections and poorly written. Scientifically I am not convinced that the results are robust, not do I feel that the paper mounts a strong scientific argument to support the statements presented in the paper. Overall I think that the scientific question is essentially solid however more work needs to be done to convince me that there are new and novel results being presented here. At this stage I have no choice but to recommend major revisions, I would also suggest that if the authors are intended to submit a revised version that it be edited by a native English speaker.

**Re: Thanks for your comments. On the basis of your suggestions, major revisions have been being made:**

**(1) Clarify and highlight the implications of this manuscript in science in the Introduction section, including several points:**

- **a. Background of carbon cycle in the study region. Probably due to geopolitical factors, observation-based studies on ocean $pCO_2$ change and its mechanism as well as it influence on ocean carbon sink and OA are less in the Indian Ocean compared to the Pacific and Atlantic (Lenton et al., 2012; Fay and McKinley, 2013; Takahashi et al., 2009), except for the sparse study e.g., by Metzl (2009) in the south-western Indian Ocean. In addition, the Indian Ocean, which is surrounded by land in the north and lacks steady equatorial easterlies in the equatorial belt, is different from the other oceans (Valsala et al., 2012), and no study is found on $CO_2$ change in the equatorial Indian Ocean. Thus, this manuscript will provide a better understanding of the carbon cycle in the Indian Ocean and may get more attractions for the Indian Ocean in the future.**
- **b. Necessarity for ocean $pCO_2$ change study. Although ocean $pCO_2$ increase due to quickly rising atmospheric $CO_2$ levels is expected, its increase rate varies geographically due to the modulation by local oceanographic**

**processes (e.g. lateral mixing, upwelling and biological activities) (Takahashi et al., 2006). An extreme example is that sea surface $pCO_2$ in the vicinity of the Bering and Okhotsk Seas decreased during 1970-2004 (Takahashi et al., 2006), as atmospheric $CO_2$ level quickly rose. Thus, investigating the long-term trend of ocean $pCO_2$ in this region is of much importance in precisely determining global ocean carbon sink (Takahashi et al., 2006).**

**c. Necessarity for OA study. Surface pH in the Indian Ocean is relatively low compared to other oceans, and the Indian Ocean is more susceptible to OA (Takahashi and Sutherland, 2013). However, by now little information on OA is available in the Indian Ocean, although the Bay of Bengal Ocean Acidification (BOBOA) moored buoy was deployed on 23 November 2013 (http://www.pmel.noaa.gov/co2/story/BOBOA). Furthermore, coral has wide distributions in the Indian Ocean (Allen and Adrim, 2003) and is sensitive to OA (Orr et al., 2005). Thus, discussion on OA in this manuscript is scientifically full of significance.**

**(2) a lot of work on data processing and corrections (deseasonalize) has been added:**
**a. adding the description of the accuracy of Hydro $CO_2$ sensor,**
**B.considering the $pCO_2$ seasonality as well as data heterogeneity spatially.**
**These will make our results more convincible.**

**(3) We will refine some statements in the revised version. For example, it is impossible to discuss interannual variability based on the limited data. However, it will be OK to discuss the long-term trend (mean trend) of $pCO_2$ change.**

**(4) We identify quantitatively the contribution of temperature, salinity, DIC and TA on $pCO_2$ increase, and conduct in-depth analysis of the influence of $pCO_2$ increase on air-sea $CO_2$ flux and OA.**

**(5) We will improve and polish the language of this manuscript.**

**We hope that you will find the results in this manuscript will be more convincible and this manuscript will do some contributions scientifically after major revision.**

Major Comments

Overall the authors discount some potential mechanisms of interannual variability (not very well) but then to go onto present only a hand waving argument (at best) as to what mechanism maybe controlling variability.

**Re: In the revised version, we first identify quantitatively the contribution of temperature, salinity, DIC and TA to sea surface $pCO_2$ increase and then find out several candidates for DIC increase (we find DIC has the biggest contribution), which provides in-depth discussions on the mechanism of $pCO_2$ increase.**

As the major result of the paper is comparing one cruise with the historical data, this is clearly not enough. That the paper lacks serious background e.g. what is the seasonality? doesn't help. I remain unconvinced that the interannual changes are not in part due to this.

**Re: Yes, it is not enough to discuss the interannual variability by comparing one cruise with the historical data. However, it will be OK to use these data to obtain the long-term mean trend of $pCO_2$ change after deseasonality, especially in the region with fewer data. For example, in the early period Oudot et al. (1995) compared two cruises in 1993 and 1984 (published in *Tellus B*). More recently, Byrne et al (2010) reported an average change rate of -0.0017 $yr^{-1}$ for pH in the North Pacific Ocean based on data in 1991 and 2006 (published in *Geophysical Research Letters*). In addition, in the manuscript we will clearly point out the flaws of limited data and the necessarity for more $CO_2$ observations, which will favorably promote the carbon cycle studies in this region.**

**In the revised version, we will add serious background including the $pCO_2$ seasonality. We find there is indeed a large seasonal variation for $pCO_2$ in this region, which will be shown. Thus the seasonality may influence the interannual variation or long-term trend. So in the revised manuscript, we first deseasonalize and then determine the trend of $pCO_2$ change.**

Equally the statement is made that this region acts as a strong source, but this is always inferred and never shown.

**Re: In fact, Fig. 1 provides the climatological annual $CO_2$ flux by Takahashi et al**

**(2009). And we also cited the papers of Bates et al. (2006) and Takahashi et al. (2009) to verify this in line 16-17 on page 529. In the revised version, we will discuss $CO_2$ flux in-depth. So we will calculate the air-sea $CO_2$ flux using the $pCO_2$ data in the seawater and in the atmosphere as well as the NECP wind data and show this figure.**

The authors also assume that there is linear response in oceanic $pCO_2$, while over this period the response is clearly not linear in the atmosphere (see Fig 4) – therefore I question the results that the strength of the $CO_2$ source is decreasing over the study period, and also how sensitivity are these results to the 1962 values. That said, clearly large changes have occurred in this region over the last 4 decades, without understanding how it may have changed assuming that TA has not changed over this period seems a erroneous assumption make – clearly using CO2sys or other carbonate chemistry with only changes in oceanic pCO2 is not enough.

**Re: In fact, we asserted in the manuscript that the strength of the $CO_2$ source was increasing in line 9-10 on page 533. In the revised version, we will calculate the air-sea $CO_2$ flux and check the strength of the $CO_2$ source. And the trend of $\Delta pCO_2$ ($\Delta pCO_2 = pCO_{2\ air} - pCO_{2\ sea}$) is not necessarily consistent with that of $CO_2$ flux, since $CO_2$ flux is also influenced by wind speed (gas transfer velocity). In addition, in the revised version, we will estimate surface TA using sea surface temperature and salinity data based on the formula of Lee et al. (2006), which has been widely used, and thus we will not assume that TA does not change over the period.**

I am also concerned that the authors discount MLD based on their limited data – certainly changes in other seasons can have a profound impact on mixed layer dynamics. The authors are worried about salinity changes on $pCO_2$ as these can only be minor (< 1%) and the salinity changes are likely a tracer of water mass changes, which is a line of evidence that such be pursued. Overall I find the presentation of the figures challenging, as they do nothing to help the arguments presented here.

**Re: The variation of MLD will reflect the extents of vertical mixing, which will influence the variation of surface DIC content. In the manuscript, we used the MLD**

**data during 1969-2009 from http://www.ifremer.fr/cerweb/deboyer/mld/Surface_Mixed_Layer_Depth.php. More details can be found in Keerthi et al. (2013). In the revised version, we will add the MLD data in the year of 2012. We will refer to related literature to check whether there is a change in water mass. And we will check the figures carefully and remove some that are not necessary.**

Minor Comments

There are numerous grammatical errors through the text that need to be addressed.

**Re: In the revised manuscript, we will correct the grammatical errors and polish the language of the manuscript with the help of a native English speaker.**

Some of the statements made in the text are redundant e.g. Generally, seawater $pCO_2$ increases with temperature.

**Re: We will check these statements and remove the redundant ones.**

The introduction of OA is confusing and aragonite is not introduced at all – nor is its relationship to temperature.

**Re: In the revised manuscript, we will add the detailed introduction of OA including aragonite and its relationship to temperature.**

What does +/ - 1% of the upper range values of $pCO_2$ equate to (line 23 , p526)

**Re: "±1% of the upper range value" refers to the maximum $pCO_2$ value the sensor is calibrated for. If the HydroC is e.g. calibrated from (200-500) µatm the uncertainty is ±5 µatm. We will add the discussion of the calibration and the accuracy of the 2012 data, especially the in-situ calibration of the data, although this work was presented in detail by Fietzek et al.( 2013). Fietzek et al. (2013) asserted that "the average difference between sensor and reference pCO2 was -0.6 ±3.0 µatm with an RMSE of 3.7 µatm". And our in-situ discrete carbon samples indicate the accuracy better than 5 µatm.**

There are lots of inconsistences such as in the abstract - the EIO being a source of atmospheric $CO_2$ but then state it's a sink.

**Re: We will check these inconsistences carefully and correct them. I guess this situation is to a large extent due to the poor use of language. For example, this sentence "The $pCO_2$ increase in the equatorial waters ($CO_2$ source to the atmosphere) was probably due to the transport of carbon accumulated in the $CO_2$ sink region (to the atmosphere) towards the $CO_2$ source region on a basin scale via ocean circulation." can be changed to "The $pCO_2$ increase in the equatorial waters ($CO_2$ source to the atmosphere) was probably due to the uptake of $CO_2$ in the sink region e.g. in the subtropical ocean, where carbon accumulated can be transported to the equatorial belt on a basin scale via ocean circulation.". We will polish the language of this manuscript.**

A discussion of what $pCO_2$ rates faster or slower than the atmosphere means does need to be included somewhere in the text.

**Re: A simple discussion on this has been made on 5-14 on page 533. In the revised version, a thorough and in-depth discussion will be conducted.**

How is MLD calculated?

**Re: This mixed layer depth is in fact a temperature-mixed layer depth, or isothermal layer depth. Mixed layer depths are computed as the depth with a 0.2 °C absolute temperature difference from 10 m temperature (Keerthi et al., 2013). This can also be found on page 542.**

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