

Dear editor,

Thanks very much for your and reviewers' professional suggestions and comments. The authors have carefully revised the manuscript entitled as "*Effect of land-sea air masses transport on spatiotemporal distributions of atmospheric CO₂ and CH₄ mixing ratios over the Yellow Sea*". All the suggestions and comments were replied point by point as below.

Reply to Reviewer 1

Comment 1:

Page 1, line 17-18: change to "An improved data filtering method was established ..."

Response 1:

Accept.

The authors have revised the sentence as "An improved data filtering method was optimized and established to ..." in line 18, page 1 in the revised manuscript.

Comment 2:

Page 1, line 19: "Fe found that, ..." ??

Response 2:

Accept.

The authors have changed the sentence as “We found that...” in line 19, page 1 in the revised manuscript.

Comment 3:

Page 3, lines 28 ff: "... to optimize an improved data filter approach for shipborne underway continuous observed atmospheric CO₂ and CH₄ mixing ..."; either "continuously observed" or "... for continuous shipborne ... observations ..."

Response 3:

Accept.

The authors have revised the sentence as “...to optimize an improved data filter approach for shipborne continuous mobile observation of atmospheric CO₂ and CH₄ mixing ratios, ...” in line 1-3, page 4 in the revised manuscript.

Comment 4:

Page 4, line 17: references to the three land-based stations is missing. No data source is given, no mentioning of the data providers in the acknowledgements; this is a poor scientific standard.

Response 4:

Accept.

In caption of Figure 1, the authors have supplemented a website: https://www.esrl.noaa.gov/gmd/dv/site/site_table.html, from which the observation data at three regional atmospheric stations was downloaded and authorized to use in this work. Of course, a specific THANKS has been supplemented in Acknowledgement.

Comment 5:

Page 5, line 20: why does it need a mass flow controller in the inlet system? The CRDS is controlling its inlet flow according to the requirements to keep the pressure in the cavity stable. I wonder how to avoid conflicts between the external MFC and the instrument-internal control.

Response 5:

Thanks very much.

Generally, the CRDS analyzer needs a flow rate of approximate 110 mL/min, and can maintain a low pressure of 140 Torr in its cavity. We introduced a mass flow controller (MFC) in the self-assembled measurement system for keeping the same flow rate for ambient air and standard gases, which were pressed in cylinders with high pressure. Although the outlet pressure could be regulated by valves, the MFC could keep the same flow rate of all standard gases better.

In fact, we have observed atmospheric greenhouse gases for more than

ten years in this way, as well as most labs around the world. No abnormal phenomenon has been found. Data that observed at Waliguan station, Linan station, Shangdianzi Station and Longfengshan station have been shared to World Meteorological Organization and National Ocean and Atmosphere Administration for assessment of global climate change, such as IPCC report.

Comment 6:

Page 5, calibration: There was no calibration after the campaign? How do you ensure stability in the instrument response? The calibration system, as laid out in Fig. 2. Should be capable of calibrating automatically. Why there was no calibration during the cruise? Was there any other (target) tank to keep track of the variability of the instrument performance?

Response 6:

The authors had revised the manuscript and supplemented more essential and detailed information in page 5 and page 6 in the revised manuscript. Because the homeport of the *RV Dongfanghong //* is Qingdao City, thus all instruments and materials had to be transported from Dalian city to Qingdao city (approximate 600 km, by truck and ferry). In order to obtain data along the cruise track, the CRDS analyzer would be calibrated and checked both before and after each campaign for several times, including

electricity supply, status of the analyzer, absorption of water vapor and air tightness, etc. During the cruise, three standard gases were automatically introduced into the CRDS analyzer, one by one for 5 min, regulated by the CRDS analyzer, with flow rate of 200 mL/min. Standard gases were measured one round every day.

One of the standard gases was regarded as the target gas for monitoring status of the CRDS analyzer, mainly according to the compatibility goal of the WMO/GAW.

Comment 7:

Fig. 3 (left panels): reduce y-axis range to at least 350 to 1000 ppm.

Wind information shown in Fig. 5 (right panels) could be added to Fig. 3 and Fig. 4 (right panels).

Response 7:

Partial accept.

According to the field observation data, the authors have modified the range of Y-axis to 350 to 1000 ppm for atmospheric CO₂ in Figure 3a and Figure 3c in the revised manuscript.

However, the authors thought it would be better to introduce atmospheric CO₂ and CH₄ mixing ratios, and wind data separately, for showing spatial and temporal distribution of them more clearly.

Comment 8:

Chapters 3.1 and 3.2 (presentation of mean, highly aggregated numbers) do not provide much information.

Response 8:

Partial accept.

Both chapters 3.1 and 3.2 were basic parts of “3 Results”, in which the authors showed and introduced the original observed data, such as variation characteristics of atmospheric CO₂ and CH₄ mole fractions, and wind along the cruise track.

In the revised manuscript, chapters 3.1 and 3.2 were merged. In addition, more detailed contents were introduced in chapter 4.1, such as preliminary comparison between our observed results and the simulated MBL references, and data observed at adjacent atmospheric stations.

Comment 9:

Page 8, line 4: the JGS and TAP numbers: are these mean numbers over the duration of your campaigns? Are these monthly means? Again, this (very crude) comparison does not provide much information. Nothing is said about the characteristics of the three land-based stations. Are they considered to be background stations? Why do you compare with these stations and the marine boundary layer reference? How do you explain that the CO₂ mixing ratio at LAN is below the MBL range? There is no

need to give numbers with two decimal places. Maybe it could be useful to also show time series of the land-based stations, to put the observed variability of your shipborne measurements in a better perspective.

Response 9:

Partial accept.

The used values were weekly means, and were observed at three adjacent atmospheric stations during the same periods of our field surveys in November 2012 and June 2013.

The key purpose of comparison was to show comparability of our observed results and data observed at adjacent atmospheric stations during the same periods.

The JGS, TAP and LAN are three atmospheric regional background stations. However, data observed at these stations could represent regional background level, unless they were filtered according to specific approaches and processes, such as method described by Fang (2015) for the LAN station (Figure 1).

- Fang S X, Tans P P, Steinbacher M, et al. 2015. Comparison of the regional CO₂ mole fraction filtering approaches at WMO/GAW regional stations in China. *Atmospheric Measurement Techniques*. 8: 5301-5313

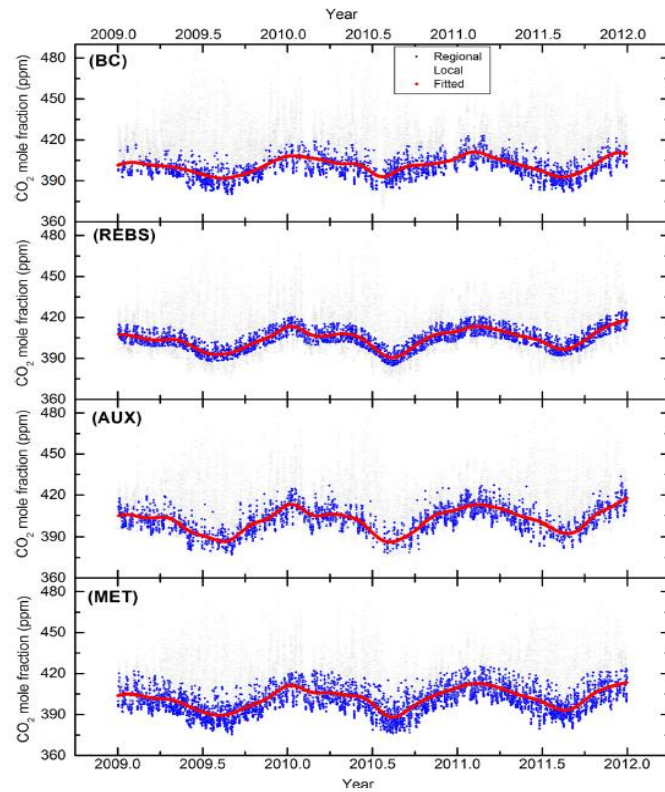


Figure 1. Filtered CO₂ mole fractions in the four approaches (BC: black carbon as tracer; REBS: Robust Extraction of Baseline Signal; AUX: CH₄ as auxiliary tracer; MET: meteorological filter). The closed blue circles represent the filtered regional events. The open gray circles represent local events which are influenced by very local sources or sinks. The red lines are results fitted to the filtered regional events using the curve-fitting method by Thoning et al. (1989).

As showed in Figure 2, atmospheric CO₂ mole fraction ranged from 377.06 to 438.46 ppm during 22nd to 30th June 2013, with a mean value of 396.43 ppm. Simulated MBL-CO₂ references, represented mean spatiotemporal distribution in marine boundary layer, ranged from 397.38 to 397.92 ppm in latitude zone of 30 °N to 37 °N during the same period.

Observed mean atmospheric CO₂ mole fraction at LAN station was approximate 1.49 ppm lower than that simulated MBL-CO₂ reference during the same period. This phenomenon was mainly influenced by land ecosystem, which could uptake amount of atmospheric CO₂, especially in June. In fact, seasonal variation of atmospheric CO₂ mole fraction observed at LAN station were larger than those observed at island-based stations, such as Mauna Loa station (Fang et al., 2015).

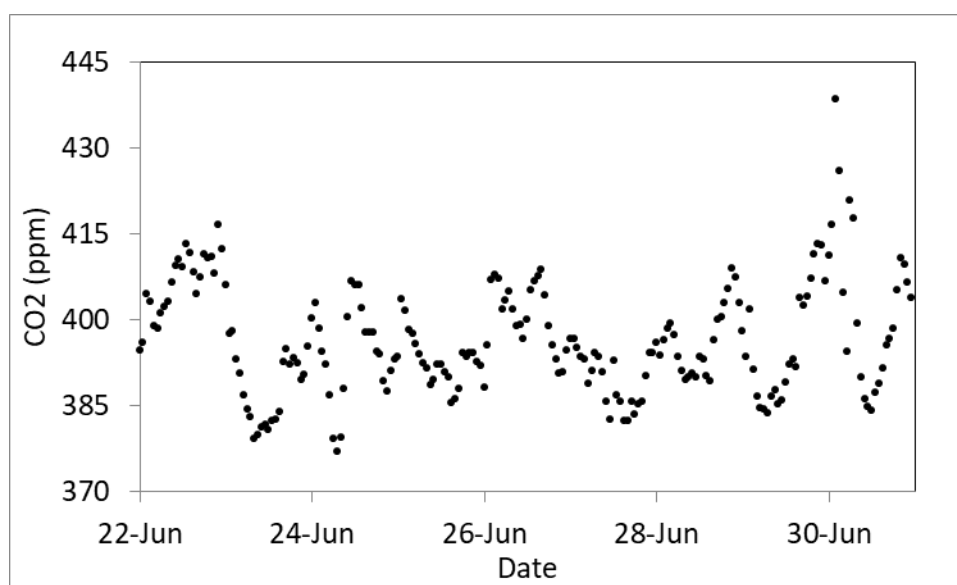


Figure 2. Atmospheric CO₂ mole fraction at LAN station IN 22nd to 30th June 2013.

For numbers with two decimal places in this manuscript, it is not a mandatory style. The authors have shown atmospheric CO₂ and CH₄ mole fraction with two and one decimal places, respectively, in publications for more than ten years. In fact, the CRDS could obtain high resolution and precision data. Thus, we thought the expression was reasonable and acceptable.

Comment 10:

Page 8, line 6: url does not exist. Elaborate. What does MBL mean? How does NOAA calculate the MBL numbers? How sensitive is the MBL product to regional signals, like the discussed changes in the regional patterns due to monsoon? This is of importance when comparing your observations with the MBL product (e.g. in Fig. 12).

Response 10:

Accept.

The authors have replace URL by <https://gml.noaa.gov/ccgg/mbl/data.php> in line 15 page 13 in the revised manuscript, due to previous link was no longer used.

MBL was abbreviation of marine boundary layer (atmosphere). MBL references of CO₂ and CH₄ are simulated based on measurements from a subset of sites from the NOAA Cooperative Global Air Sampling Network. Only sites where samples are predominantly of well-mixed marine boundary layer (MBL) air representative of a large volume of the atmosphere are considered. The exact calculation method is described in detail on the website (<https://gml.noaa.gov/ccgg/mbl/mbl.html>).

Comment 11:

Page 8, lines 12 -15: incomplete sentence, please revise.

Response 11:

Accept.

The sentence has been revised as “Observed mean CH₄ mixing ratios were 1924.8 (27.8) ppb and 1918.0 (25.7) ppb in November 2012 and June 2013, respectively, which were slightly higher than historical data of 1915.5 ppb in the SYS in March 2013 (Zang et al., 2017), and higher than the MBL-CH₄ references of November 2012 (1869.5 to 1880.3 ppb) and June 2013 (1835.3 to 1846.6 ppb).” in line 19-22 page 13 in the revised manuscript.

Comment 12:

Page 11, lines 4 ff.: I do not understand the statement "... observed atmospheric CO₂ and CH₄ mixing ratios... were corrected by a linear function ...". What does that mean? Did you interpolate in between different calibrations? Above (page 5), you mention "Before each campaign, three standard gases were used to calibrate ...". Thus, I understand from there that there was only one calibration performed.

Response 12:

For express more clearly, the authors have revised these sentences as “Firstly, observed atmospheric CO₂ and CH₄ mixing ratios along the cruise tracks in November 2012 and June 2013 were calibrated by a linear function, averaged every one minute, and named as Raw Data for the

subsequently process.”

As introduced in chapter 2.2 (line 5-11) in the revised manuscript, three standard gases were measured one round each day. Based on measured results and standard values, a linear equation was established, according to the least square method. Then, observed atmospheric CO₂ and CH₄ values every day could be calibrated by the linear equation.

Comment 13:

Page 11, lines 19 – 21: I am not aware of any WMO/GAW recommendation making statements on the maximum standard deviations. Add reference (if there is any). Sentence is also (grammatically) incomplete.

Response 13:

Accept.

The author has added the reference associated quality control which recommended by WMO/GAW:

- WMO, 12th WMO/IAEA Meeting of experts on carbon dioxide concentration and related tracers measurement techniques. NO. 161[R]. Toronto, Canada, 2005.

In addition, sentence was revised as “According to the quality control criteria of CO₂ (\pm 0.10 ppm), which recommended by the World Meteorological Organization Global Atmospheric Watch (WMO/GAW)

(WMO 2005), 3 knots was optimized as the threshold. Results showed that, 15.5% and 21.9% of total observed data in November 2012 and June 2013, respectively, were flagged in this step” in line 22-26 in page 11 in the revised manuscript.

Comment 14:

Fig. 6 (left panel) shows two black data streams. Why?

Response 14:

One black data stream represented ship speed. The other one represented wind direction, which should not be plotted in Figure 6a. The authors have removed it in page 12 the revised manuscript.

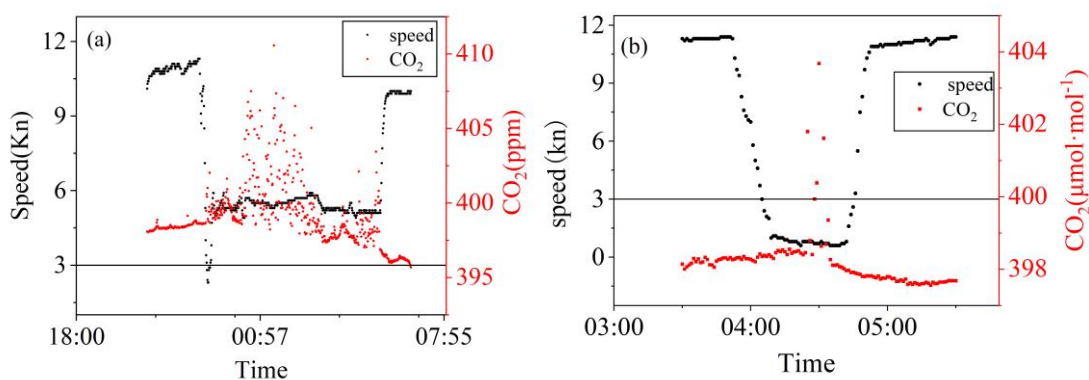


Fig. 6. Variations of observed CO₂ mixing ratios and ship speed from 20:40 on 28th to 6:40 on 29th June 2013 (a) and 3:30 to 5:30 on 3th November 2012(b).

Comment 15:

Page 12, lines 9-10: "This procedure was repeated until no outliers were identified ...". Is it really possible to repeat till there are no outliers

identified anymore. As a rule of thumb, +/- 3 sigma covers about 99% of the data, thus, there will be always about 1% identified as outliers, right?

Response 15:

The Pauta criterion (“ 3σ ” method) is a classical and widely used data filter approach, which has been used for many years. Theoretically, ± 3 sigma could cover 99.7% of a group values. When the procedure was repeated, all differences between observed values and mean value will less than ± 3 sigma. Specially, this method could not work if data include less than 10 values.

Comment 16:

Fig. 7: how do you explain manually flagged CO₂ data (top left) while the corresponding CH₄ aren't flagged (bottom left)?

Response 16:

Manual in Figure 7 mean flagged observed values manually, because these values were observed when the dry tube was replaced and the indoor air was pumped into the CRDS analyzer. Both abnormal CO₂ and CH₄ were flagged in Figure 7 (top left and bottom left).

In addition, when the dry tube was replaced, influenced by human breathing, fluctuation of CO₂ values were more intensive than that of CH₄ values.

Comment 17:

Page 16, line 3: "... almost all the transport track" must read "... almost all the transport tracks ..."

Response 17:

Accept.

The author has revised the sentence as “Furthermore, back trajectory analysis showed that almost all the transport tracks were originated from the Asian Continent in November 2012,...” in line 4-5, page 16 in the revised manuscript.

Comment 18:

Chapter 4.3: I mainly see one striking difference when looking at Fig. 8 and 9. And this is the reversal of the CH₄ W-E gradient in section 4 in July 2013. All other transects do show the (expected) gradients with lower mole fractions the more go away from the continent.

Response 18:

Striking differences in section 2 were plotted in Figure 8 and Figure 9, because only data observed in sections were showed in the two figures. However, real cruise track was not continuous in section 2, as showed in Figure 1a. In fact, atmospheric CO₂ and CH₄ were observed in two different date in section 2 in Figure 8. In addition, wind direction varied

remarkably during field survey of June 2013, as showed in Figure 5c, caused atmospheric CO₂ and CH₄ varied dramatically, as showed in Figure 9, because of land-to-sea air masses transport from surrounding continent. But, to study influence of land-to-sea air mass transport on distributions of atmospheric CO₂ and CH₄, data observed in November 2012 was better than that of June 2013, which was interfered by complex land-sea interaction by variable wind direction.

Comment 19:

Page 16, lines 5-6: "... which resulted in higher atmospheric CO₂ and CH₄ mixing ratios in November 2012 ... than that in July 2013 ...". I disagree, see my comment just above. It is also pretty difficult to compare Figs. 8 and 9 due to the different color codes. Could you use the same colors for the same mixing ratio ranges?

Response 19:

Partial accept.

Atmospheric CO₂ mixing ratios ranged from 392.94 to 432.70 ppm with mean value of 404.95 ppm in November 2012, and from 384.95 to 418.90 ppm with mean value of 392.49 ppm in June 2013.

Atmospheric CH₄ mixing ratios ranged from 1880.2 to 1981.0 ppb with mean value of 1930.7 ppb in November 2012, and from 1820.8 to 2185.0 ppb with mean value of 1923.4 ppb in June 2013.

Thus, we concluded that more intensive land-to-sea air mass transportation caused higher atmospheric CO₂ and CH₄ mixing ratios in November 2012 than those in July 2013.

Initially, the authors have tried to plot Figure 8 and Figure 9 by using the same range and color codes, but gradient distributions could not be displayed well.

Comment 20:

Figs. 8 and 9: how can you interpret some other, smaller scale, features in the transects. E.g. increases in CO₂ in sections 2 in Nov '12 and in section 3 in July '13 (when looking from W to E).

Response 20:

We have found that distributions of atmospheric CO₂ and CH₄ mixing ratios were mainly regulated by air mass transportation. Take section 2 in Figure 2 as an example, and as show in Figure 3, during field survey, wind flowed from east to west for almost half a day. Thus, air mass was transported from Korean Peninsula to survey area with high content of CO₂ and CH₄, induced decreasing distributions of atmospheric CO₂ and CH₄ mixing ratios from east to west.

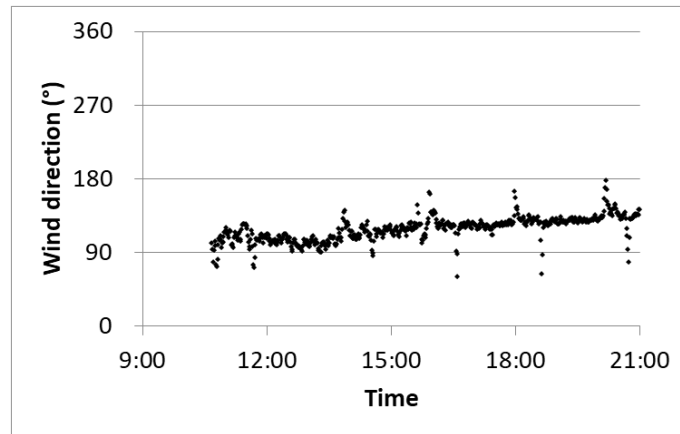


Figure 3. Wind direction in section 2 in November 2012. Prevailing wind direction was E-SE during this period.

Comment 21:

Page 18, lines 8 -10: "... the inflection points of gradient could be set at 123.30 °E, 123.50 °E and 123.40 °E for section 3, 4 and 5, respectively ..." What the interpretation of this finding?

Response 21:

Previous studies have reported atmospheric greenhouse gases and their distributions in marine boundary layer were influenced by land-to-sea air mass transportation. However, in this work, the authors tried to establish a method to determine the distance away from coastal line, that influenced by land-to-sea air mass transportation.

According to the precision of the CRDS analyzer and compatibility of measurement that recommended by WMO/GAW, ± 0.1 ppm and ± 2 ppb were recognized as criterion of determination.

Then, based on field observed results, as showed in Figure 10 in the

manuscript, the authors found 123.30 °E, 123.50 °E and 123.40 °E were inflection points for section 3, 4 and 5, respectively. ΔCO_2 and ΔCH_4 were greater than 0.1 ppm and 2 ppb on west part of these inflection points, suggested influenced remarkably by land-to-sea air mass transportation.

Comment 22:

Page 18, lines 10-12: "... the offshore distances away from continent could be calculated as approximate 27.0, 26.3 and 11.7 km, respectively, with a mean value of 21.7 km ...". Interpretation? Once you are 20-30km of the coast, the influence of the land-based emissions is negligible? Is this what you want to say?

Response 22:

The authors have replied it in comment 21. Moreover, for the two typical surveys in November 2012 and June 2013 in this work, the influence of land-to-sea air mass transportation on distributions of atmospheric CO_2 and CH_4 mixing ratios in marine boundary layer were considered remarkable and directly measurable in distance of approximate 21.7 km away from coastal line.

When distance away from coastal line was greater than 21.7 km, influences of land-to-sea air mass transportation on distributions of atmospheric CO_2 and CH_4 mixing ratios was existent but negligible in

this work.

Comment 23:

Fig. 11: does it show data for both campaigns?

Response 23:

No.

Chapter 4.4 was discussion about the influence of land-to-sea air mass transport, thus only data observed in November 2012 was used, when the air mass was transported from Asian continent to Pacific Ocean driven by the East Asian Winter Monsoon. The authors have revised the sentence as “As shown in Fig. 11, atmospheric CO₂ and CH₄ mixing ratios observed in November 2012 showed the same fluctuating feature versus wind direction, ...” in line 5-6 page 17 in the revised manuscript, for expressing more clearly.

Comment 24:

Fig. 12: does it show data for both campaigns?

If so, I do not understand that the CH₄ increase when going east in section 4 in July 2013) is not reflected here.

Response 24:

No.

As the authors replied in comment 23, only data observed in November

2012 was used, when the air mass was transported from Asian continent to Pacific Ocean driven by the East Asian Winter Monsoon. The observed data in November 2012 was an ideal case for studying the influence of land-to-sea air mass transport.

Reply to Reviewer 2

Comment 1:

1. The manuscript could use some homogenization and restructure. I suggest moving the procedure of data cleaning to the methods section. The data cleaning procedure can still be discussed or its effect on the reported results.

Response 1:

Unaccepted.

There have been several data filtering approaches based on the concentration of atmospheric black carbon (BC), on a statistical approach (robust extraction of baseline signal, REBS), and on meteorological parameters (MET), for atmospheric greenhouse gas that observed at land-based stations around the world. Thus, in this study, the authors tried to establish a data filtering approach for ship-based underway observation, which was an essential part of this work. We believed this work would be a contribution to science community. Based on this initial approach, we could discriminate emission sources and insight mechanisms of

spatiotemporal distributions of observed atmospheric CO₂ and CH₄.

Comment 2:

2. There is no clear distinction between the results and discussion section. Either merge them into one section "Results and discussion" or move many of the plots and reported results from the discussion to the results section.

Response 2:

Accept.

The authors wanted to firstly introduce observation data in section Results, then analysis mechanisms in section Discussion.

We have tried our best to clearly distinct between the Results and Discussion, via moving some contents from the Discussion to the Results.

We hope the revised manuscript could express well.

Comment 3:

3. Reported means and standard deviations should be calculated after data cleaning and quality checks. Otherwise, comparisons and interpretations are biased.

Response 3:

Accept.

The authors have calculated means and standard deviations of CO₂ and CH₄ mixing ratios after data filtering and quality checks, which were

reedited in line 9-22 page 13 in the revised manuscript.

Comment 4:

4. The presented analysis in general could benefit from better structure by defining the main questions and discussing whether the measured data can provide enough evidence to answer these questions.

Response 4:

Accept.

Thanks for your professional suggestion.

The authors have revised both the section Results and Discussion followed the reviewer's comment, to clearly demonstrate the causal relationship between observed data and scientific questions in this study.

Specific comments

Comment 5:

Page 1.

Ln 16. Please state early on what you mean by the numbers after the \pm sign. If this is SD, you can write .. the ratios were (mean \pm sd) as follows,

Response 5:

Accept.

The authors have revised the expression from 403.94 ± 13.77 ppm to

403.94 (13.77) ppm throughout the manuscript, as well as data related to CH₄.

Comment 6:

18. I believe you mean "different" sources.

Response 6:

Accept.

The authors have changed “diverse sources” to “different sources” in line page in the revised manuscript.

Comment 7:

19. Typo: We found that ..

Response 7:

Accept.

The authors have revised the sentence as “We found that...”in line 19 page 1 in the revised manuscript.

Comment 8:

19. The expression "compared to .. " is not obvious.

Better rephrase to clarify what quantity of quality is being compared.

Averages and SD reported in the abstract seem to be before data cleaning and filtering, which would result in a biased estimates and invalidate all the later comparisons.

Response 8:

Accept.

The authors have revised the expression as “We found that, the spatiotemporal distributions of atmospheric CO₂ and CH₄ mixing ratios over the south Yellow Sea were dominated by land-sea air masses transport, which was mainly driven by seasonal monsoon, while the influence of air-sea exchange was negligible”.

Values of atmospheric CO₂ and CH₄ mixing ratios in the Abstract were replaced by means and standard deviations that calculated from filtered observed data.

In addition, mean CO₂ and CH₄ mixing ratios were calculated after data filtering, which were introduced in chapter 4.1 in the revised manuscript.

Comment 9:

Page 2.

5 Here the \pm is used as measurement uncertainty which is different from the rest of the paper, I think this is confusing for the reader. It is important to stick to one use of the same sign, I would suggest you keep the use here as the measurement uncertainty and change the rest of the paper to express as mean (sd) to report your results e.g. 403.50 (13.70) ppm.

Response 9:

Accept.

Thanks for your professional comment.

The authors keep the expression here (line page in the revised manuscript) as the measurement uncertainty, and modified the rest of manuscript to express as mean value (sd). This expression could tell the measurement uncertainty and standard deviations of atmospheric CO₂ and CH₄ mixing ratios more clearly.

Comment 10:

11 Not clear here what's meant by "According to the observation platforms or methods" I suggest rephrasing

Response 10:

Accept.

Previous publications reported that, measurements of atmospheric greenhouse and related gases could be classified into six observation categories, according to the observation platforms or methods used:

1. Air sampling observation at a ground-based station
2. Air sampling observation for a vertical profile (e.g., multiple heights using a tower)
3. Ice core observation
4. Surface seawater and overlying atmosphere observation
5. Satellite-based observations
6. Air sampling observation by mobile platforms (e.g., aircraft, ships, etc.)

In order to expression concisely, the authors delete “According to the

observation platforms or methods”, because it was not essential for this study. The sentence has been revised as “Shipborne observation was considered as one of six common and important methods for observing greenhouse gases” in line 12-13 page 2 in the revised manuscript.

Comment 11:

15 "shape drop"?

Response 11:

Accept.

The authors have rewritten this sentence as “... sharp drop in the area of 20 °N...” in line 15-17 page 2 in the revised manuscript.

Comment 12:

Page 3.

23 "regulation mechanisms"

Response 12:

Accept.

The authors have rephrased “regulate mechanism” to “regulation mechanism” in line 24 page 3 in the revised manuscript.

Comment 13:

28 "objectives"

Response 13:

Accept.

The authors have rephrased “The major objects ...” to “The major objectives ...” in line 1 page 4 in the revised manuscript.

Comment 14:

29 "underway"?

Response 14:

This word mean parameters were measured by instruments when ship (or other mobile platforms) was cruising. It was frequently-used in oceanography. In order to avoid confusing readers, we replaced this word by “mobile” in line 2 page 4 in the revised manuscript.

Comment 15:

Page 4.

- It would be helpful to add a picture of the research ship indicating where the inlet port was.

Response 15:

Accept.

The authors have added a picture of the famous “*RV Dongfanghong 2*” with sign of air inlet in figure 2 in page 6 in the revised manuscript.

Comment 16:

20 Fig 1. I suggest you use different shading for sea/land Additionally Fig (1) It seems that the researchers has mistakenly used the June cruise track for both plots (i.e. same track on both plots)

Response 16:

Accept.

In order to distinct continent and sea area more obviously, the authors have replaced map with different shading in Figure 1 and other Figures in the revised manuscript.

In addition, we replotted ship's cruise track in June 2013 in subplot Figure 1b in page in the revised manuscript.

Comment 17:

Page 5.

24 .. Since you have calibrated against WMO primary standards it would be interesting to report the uncertainty of your measurements as propagated from the primary standards.

Response 17:

Accept.

All we used standard gases were propagated from the WMO primary standards by the Chinese Academy of Meteorological Sciences, Chinese Meteorological Administration. The authors have supplemented the uncertainty of standard gases in line 4-5 in page 6 in the revised manuscript.

Comment 18:

Page 6.

16 .. is as follows

Response 18:

Accept.

According to the comment, the authors have rewritten the sentence as “The principle of simulating the air mass transportation path is as follows: ...” in line 3-4 in page 7 in the revised manuscript.

Comment 19:

Page 7.

6 .. 7 are the reported standard deviations calculated for all raw data points? in this case they are not very useful in understanding the variability as the outliers will have disproportional weight.

I suggest data cleaning/quality checks should be conducted before reporting any statistics. In particular any interpretation that follows would be highly biased by outliers.

Response 19:

Accept.

The standard deviations in line 6-7 in page 7 in previous manuscript were calculated for raw observation data. The authors have deleted these values and related content in the revised manuscript, and supplemented mean and standard deviations that calculated by using the filtered observed data.

Comment 20:

9..10 what are the seasonal variations considered here, please elaborate

Response 20:

Accept.

Since the expression of “seasonal variations” would confuse readers, the authors have rewritten this sentence as “Atmospheric CO₂ and CH₄ mixing ratios were comparable with the historical observation results of the north hemisphere (Matsueda et al., 1996; Zang et al., 2017; Liu et al., 2018). Abnormal high observation values were attributed to exhaust gases of ship or anthropogenic interference of analyzer” in line 8-11 in page 9 in the revised manuscript.

Comment 21:

Fig. 3. Again I would appreciate to shade land/sea differently, the outliers on figures a and c limits the usability of this figure.

Response 21:

Accept.

The authors have replotted maps with different shading of land and sea. In addition, the Y axes in subplot Figure 3a and Figure 3c have been optimized, as showed in page 8 and page 9 in the revised manuscript.

Comment 22:

- The figures in general should be 300 ppi or vector graphics to aid in

readability.

Response 22:

Accept.

In order to improve the readability of figures in this manuscript, the authors have checked all figures, and replotted some of them. The original figures were also submitted as appendixes.

Comment 23:

Page 8.

1 The comparison of CO₂ mixing ratio difference of 2 ppm with a reported SD 13.7 is not meaningful as the variability of the signal is much higher than the observed difference.

Response 23:

Accept.

The authors have rephrased this sentence as “Observed mean CO₂ mixing ratios were 403.94 (13.77) ppm and 395.90 (3.53) ppm in November 2012 and June 2013, respectively, which were slightly lower than previous studies’ mean values of 405 ppm and 410 ppm in the YS and ECS in March 2013 and March 2017, respectively (Zang et al., 2017; Liu et al., 2018).” in line 9-12 in page 13 in the revised manuscript.

Comment 24:

Fig. 7. "Manual" legend is barely visible, I suggest to use higher

resolution plots.

Response 24:

Accept.

The authors have replotted Figure 7 to improve visible, especially replaced Manual legend “×” by blue points, in page 12 in the revised manuscript.

Comment 25:

17 Is the provided uncertainty here a sampling uncertainty of the fluxes or is it the observed variability over the course of the day?

Response 25:

The provided data and uncertainty in line 17-19 were calculated by observed in field surveys, which covered the same sea area in seven to ten whole days for each survey.

Comment 26:

Page 10

7.. no need to specify programming language in the figure caption.

Response 26:

Accept.

The authors have deleted “and drawn by python 3.7.0” in caption of Figure 5 in page 10 in the revised manuscript.

Comment 27:

12.. change the verb "read" Page 11.

Response 27:

Accept.

The authors have replaced the verb “read” by “suggested” in line 17 in page 7 in the revised manuscript.

Comment 28:

5.. better use "calibrated" to indicate this linear correction if that is what is meant here.

Response 28:

Accept.

The authors have replaced “corrected” by “suggested” in line 9 in page 11 in the revised manuscript.

Comment 29:

15.. what is meant here by "without statistical analysis"

Response 29:

3 knots of ship speed have been empirically used as a criterion of raw observed data in previous publications, with no rigorous demonstrate. However, in this study, we studied relationship between ship speed and standard deviations during field surveys, and explain why 3 knots of ship speed was a reasonable criterion for data filtering.

Comment 30:

19. Not clear what is meant by this quality control criteria that is recommended by WMO/GAW and how relevant it is for the observed variability Probably better to plot ship speed vs. measured concentration to see if the determined threshold is robust, Fig.6 is difficult to interpret as time is a confounding variable for both speed and concentration.

Was the wind direction considered in this quality scheme?

Response 30:

WMO/GAW has established a global system, including more than 200 stations, for observation of atmospheric components. In order to ensure the comparability of observed data among stations, a quality control criterion was recommended. In this study, on the basis of this quality control criteria, 3 knots was demonstrated as threshold and one step of filtering approach.

In Figure 6, two typical cases were showed that ship's speed and atmospheric CO₂ mixing ratio varied with time. During the period of each case, when ship' speed slowed down, the observed atmospheric CO₂ mixing ratio fluctuated more and more intensively, indicated the influence of ship's emission on measurement results.

Wind speed was not always considered in this step. Specially, for the R V Dongfanghong 2, when wind direction was the same with ship' heading, and wind speed was higher than ship' speed, observed data would influenced by stack gas of ship.

Comment 31:

Page 17

5.. same "phase"?

Response 31:

Accept.

The authors have revised the sentence as “As shown in Fig. 11, atmospheric CO₂ and CH₄ mixing ratios observed in November 2012 showed the same fluctuating feature versus wind direction,...”in line 5-8 in page 17 in the revised manuscript.

Comment 32:

Page 18.

9. What is the "inflection points of gradient"

Response 32:

In order to express more clearly and accurately, the authors have rephrase the sentence as “According to the calculated slope values, gradient would be gradual at 123.30 °E, 123.50 °E and 123.40 °E for section 3, 4 and 5, respectively.” in line 9-10 in page 18 in the revised manuscript.