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Interactive comment on "Methane distribution, flux, and budget in the East China Sea and Yellow Sea" *by* M.-S. Sun et al.

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We are very grateful to the anonymous referee #2 for his/her constructive suggestions on our manuscript. They are really very useful to make our paper more organized and persuasive. We considered every comment seriously and gave the responses as follows. We also attached the revised manuscript with this reply for reference.

Q1. Fig. 3-5 need to be redrawn with uniform scales for the shown parameters on all individual panels.

Reply: Figures 3-5 have been redrawn with uniform scales to make a better comparison of parameters during the five cruises.

Q2. The mentioning of the history and development of the atm. growth rate over the

last 2 decades is irrelevant for the paper and should be cut (Page 3, line 5 middle to line 9 end).

Reply: We introduced the atmospheric CH4 growth in order to highlight the imbalance of CH4 sources and sinks. However, we might show too much about the history of atmospheric CH4. So here we rewrote this part as follows: The global atmospheric CH4 has increased significantly since the industrial revolution, and was reported as 1803±2 ppb in 2011, which is about 2.5-fold higher than that (722±25 ppb) in 1750 (IPCC, 2013).

Q3. The statement in line 13, page 3 is wrong.

Reply: Here we just want to express that although a huge amount of CH4 in the seawater is oxidized by microbes under the anaerobic or aerobic environment, ocean is still one of the natural CH4 sources. It indicates that the total CH4 production is significant but difficult to quantify. Oceanic CH4 production is always ignored because of the relative small net emission. Here we made a big confusion due to the poor expression. So we corrected this sentence as "Although most of oceanic CH4 are oxidized by O2 and sulfate in both aerobic and anaerobic environment before escaping into air, ocean still releases about 11-18 Tg CH4 yr-1 (Bange et al., 1994) into the air".

Q4. The 2nd paragraph should be extended. After all, there are several studies in the area, the majority of them involving one or several of the authors themselves, and this work should be briefly summarized and incorporated in the considerations later on. The work of Rehder and Suess (Mar. Chemistry, 2001, 75, 89-108) already using the equilibration technique at an early stage is completely neglected, though providing a considerable data set for the Kuroshio-influenced surface waters in the ECS. The same is true for the work on the hydrothermal activity in the Okinawa Trough, which would be surely be worth consulting in the context of Fig.5.

Reply: It is a good suggestion that we should briefly summarize the previous work in this area. We follow the reviewer's advice and add some concise introduction about

the progress of dissolved CH4 in ECS. We also cited the work by Rehder and Suess (2001) and discussed the influence of hydrothermal activity on the CH4 distribution in shelf slope area in part 3.4.

Q5. Also, the authors might consider to widen the context by briefly addressing some similar systems (marginal seas mainly fed by oceanic waters but with riverine imprints) under investigation (e.g the North Sea)

Reply: The North Sea is a similar oceanic system with the ECS, and there are also other marginal seas influenced by the rivers (i.e. the Baltic Sea, the Arabian Sea, and the Gulf of Mexico). However, considering the length and the emphasis of this paper, we do not introduce them in the introduction.

Q6. Page 4 Line 6, "The production. . .. . ."(Karl and Tilbrook, 1994). I do not understand this sentence at all, nor the context to the reference (which deals with relation of the methane subsurface methane maximum to other variables.

Reply: We agree that this sentence cause confusion due to poor expression. Hence, we removed the relevant statements from the manuscript, because it seems to contribute little to the introduction of ECS.

Q7. Page 4, line 12-13: Researchers. . .. . . CUT. This is more of a political statement, and it is also absolutely not clear what is meant by "beneficial effects of its biogeochemical cycles . . .. . .).

Reply: Here we deleted the improper statements.

Q8. 2.1 Page 5, line 19: check concentrations given for the standards: there are the digits behind 2 and 4 missing, I believe. Please quote accuracy of standard concentrations given by the RI-CNSM.

Reply: Two digits were missing from the numbers of CH4 standard concentrations and we revised them accordingly.

Q9. 2.2 Page 6, line 9, what is meant by "at ambient temperature" Ambient in the lab or ambient in situ? Following: What was the difference between in situ and ambient. Q10. Please explicitly write down the equation used for T-adjustment ". . .. . .calibrated by the Arrhenius empirical equation"?????

Reply: In this study "the ambient temperature" means the ambient room temperature, and the in situ temperature means the ambient bottom water temperature. During the incubation, sediment cores were kept at ambient room temperature and incubated in the dark for 24$\sim$48 h. The effect of temperature discrepancy (usually 0.8-10.5°C) on the CH4 emission rate from sediments was corrected by the Arrhenius empirical equation (Aller et al., 1985; Song et al., 2015). When temperature increases by 10 °C, the chemical reaction (here referring to CH4 production and consumption) rate will increase by 2$\sim$4 times. We took 3 times for calibrating the calculation of sediment-water CH4 fluxes accordingly. Use of the Arrhenius equation for temperature correction is usually reasonable and acceptable when an incubation experiment is not conducted at the in situ temperature (e.g. Aller et al., 1985; Song et al., 2015). We have described the calibration method in detail in the revised paper.

Q11. Methods and Figure S1: you refer to "water column control" in the text, "control" in the figure caption of S1 and "blank" in the legend of S1. Please be consistent.

Reply: We named them "blank" consistently in the revised paper.

Q12. 2.3 Page 7, line 3-4 the concentration patterns of these stations, is quite unusual, with TAP and SDZ showing highest concentrations in summer and a clear land-influenced signal. I would expect the airborne mole fractions over the sea quite different. Though not really essential for the paper, the authors might want to add a sentence on this, as on the use of an annual mean in general of a gas which is known to show a seasonal cycle.

Reply: We agree with the reviewer's comment. It is true that atmospheric CH4 concentrations in these stations have a seasonal variation due to terrestrial influence, espe-

cially the high CH4 concentration in summer, but they make a minor difference in the results of sea-air fluxes. So we still use their annual average CH4 concentration for calculation, but we added the above statements in the text.

Q13. Page 7, last sentence of 2.3 “more representative and trustworthy” – than what? Please provide a scientific rationale (very easy for the most recent paper by W2014).

Reply: Nightingale et al. (2000) reported the median sea-air flux among the extensive methods and models. It is also a representative and widely-used method for sea-air fluxes calculation. The W2014 equation is an update result by Wanninkhof (2014) using the modified global ocean 14C inventories and improved wind speed products. Considering the W92 has a strong impact on the calculation of sea-air fluxes, we also gave the estimation results by W2014 to compare with Nightingale 2000 and get a rough range of sea-air fluxes. However, the results obtained by these two methods are close to each other, so we take W2014 to further report the annual CH4 emission in this area. Here we didn′t mean to make a comparison, but to explain why we choose these two methods for calculation. Hence we deleted the last sentence of 2.3 to avoid ambiguity and described the scientific rational of W2014 in the revised paper according to the reviewer′s advice.

Q14. Results: 3.1 Line 17 “succession”? , => confluence

Reply: ‘succession’ indicates the dynamic of Kuroshio and Changjiang freshwater during a whole year. In summer, the Changjiang outflow is 2 times higher than that in winter, while it is on the contrary as for the Kuroshio. In winter, the water charge of Kuroshio is much larger than that in summer. Since the word ‘succession’ seems to cause confusion, we revised this part to avoid it.

Q15. Fig 2: Figure Caption, last line, pls. extend: “..... CH4 sampling points, with concentration indicated by color code.

Reply: We added it to the Fig. 2′s caption as suggested: Color dots: CH4 sampling

points, with concentrations indicated by color scale.

Q16. Page 7, Lines 22ff: from here on, you are also referring to Fig. 3 already, which has the spatial information, so insert a (Fig. 3) here.

Reply: We accepted the reviewer′s suggestion and inserted a “Fig.3” there.

Q17. Page 8. Line15: It is odd to infer the mixing of the water masses from the methane pattern, with a.) lots of hydrographic parameters at hand and b.) a conclusion later on which suggests that most of the methane is produced in the water column. Skip or make the point from the physical parameters.

Reply: We realized that it was improper to discuss the mixing of the water masses from the point of CH4 concentration. So we skipped this point here and deleted the last sentence in part 3.1 in the revised paper.

Q18. 3.2, 3.3, 3.5 and Tables 2 and 3 there are major issues with the (statistical) handling and interpretation of the data in these sections. Table 2, to my understanding, basically gives the ranges of data measured (so far so good), and the average and SD, where it should be stated that the SD gives the average difference between the average value and the individual values (and not any kind of uncertainty of the average, as suggestive from the sign). But what kind of average was chosen, given the fact that the measurement points were not evenly distributed. I assume just the linear) average. Moreover, there are strong variations in the areal coverage of the different surveys over the year. Later in the result and discussion, the differences in the mean values are then used to address seasonal changes. This is simply not scientifically sound. For instance, one of the findings of the paper is that the concentrations and fluxes were highest in summer. However, this cruise had a more limited spatial coverage, with a strong bias towards the mouth of the Chang Jiang river, which is –correctly – identified as the major hot spot of creating enhanced methane concentrations. At the other hand, the waters in the axis of the Kuroshio current were not sampled in summer at all. So a part of the summer maximum of surface and bottom methane concentrations is

definitely a result of a sampling bias, and it is not intelligible how large this bias actually is. On the counterside, the lowest average concentrations were found in December, when the grid was heavily reduced towards the river mouth, but had lots of sampling stations in the Yellow Sea and the area south of 32 N, producing a bias towards a lower CH4 concentration field. To be clear: I do not mean that the authors are not right in their conclusion about the annual behaviour; from the approach, I just cannot tell -at least not quantitatively- and the authors can't either, I fear. One possible approach to overcome this problem would be to use integrate over the gridded surface and bottom concentration fields (getting rid of the spatial inhomogeneity) and to compare results just from areas which are covered by all surveys. In 3.6, the authors estimate the surface ASE fluxes. Here again, several things are unclear or potentially wrong. First of all, the boundary between the YS and ECS is not clearly defined, nor the total areas of the two seas indicated for which the approach is then followed. As the ASE flux is, under the assumption of common winds over the entire area, is proportional to the sea surface area, here at least a spatial weighting would be needed. Here again there is a problem with the spatial bias.

Reply: We agree with the reviewer that there was potential deviation due to the large sampling bias and improper data processing method. So we divided the sampling region into four subareas, i.e. the Yellow Sea, the Changjiang estuary (S<30), the ECS shelf (S >30, depth <200 m) and the ECS slope area (S >30, depth >200 m). To eliminate the influence of strong bias in the areal coverage of the different surveys, we only discussed CH4 seasonal variation in the ECS shelf, which was covered by all survey (Fig. 4). The corresponding discussion was updated in part 3.2. It indicated that surface and bottom CH4 concentration had an obvious seasonal variation in the ECS shelf area, with the highest level in summer and the lowest level in early spring (March). Surface CH4 concentrations in summer were slightly higher than the other seasons, while bottom CH4 concentration had an obvious seasonal variation with an extremely high value in August. Similarly, we also reported the range and mean sea-air flux density in each area during the four cruises, respectively (Table 3). An average

area weighed sea-air fluxes in ECS and YS was recalculated to avoid the potential spatial bias. Data in Table 3 were also classified and updated in the latest version. Further, we also recalculated the annual emission in ECS and YS combined with the averaged area weighed sea-air flux density and their areas. It was estimated that the total CH4 emission from the ECS and YS was 4.09$\times$109 mol (about 0.065 Tg) during 2011, and we have updated this result in part 3.6.

Table 2 gave us the background information, just like temperature, salinity and CH4 level in ECS during all the cruises. We think it is necessary and important, so we still keep it. However, we stated that the SD gives the average difference between the mean value and the individual values in the table title as suggested by the reviewer.

Besides, the boundary between the Yellow Sea and the East China Sea was the straight line from the northern tip of the mouth of the Changjiang (Yangtze River) toward the Jeju Island, which has been mentioned in the introduction and also shown in Fig. 1 with the blue dashed line.

Q19. On top of this, there is again a huge problem in the understanding and use of statistics obvious in connection to Table 3. What is meant here be the values given behind the averages? Here, uncommented, a sign would suggest the uncertainty range of the average. But there are basically no data supporting an undersaturation, and in particular, the (unclear) error propagation gives raise to ASE flux estimates with an error margin allowing for considerable fluxes from air to sea.

Reply: The value behind the average is the standard deviation, which means that the dispersion degree of the individual measurement value relative to the average of all the values in this cruise, rather than the uncertainty of the average. Maybe we made confusion when we use the average and standard deviation to represent the data. So here we only give the range and average of sea-air flux density. We have already updated Table 3 and reorganized data according to salinity and depth, which was mentioned in detail in the reply for the Q18. According to our observation, almost

all CH4 is oversaturated except several stations in spring.

Q20. Page 7027, Line 11: is section P not the same than section CJ?, you refer to this section as section P also for May in the Fig caption at some other places of the mscrpt.

Reply: These sections are all along the PN line. But the stations along this section have different names in different cruises. To avoid causing any confusion, we named the section using a consistent name “section PN” in the text and figures.

Q21. Page 2027, Line 13. It is not true that the water is well mixed in the upper 100 m along the entire March Section. This is not true for the plume-affected west, but also for the very north.

Reply: The water column develops a perennial stratification in the estuary, so it is not true that the water is well mixed in upper 100 m along the whole section PN in March. At least, from station P01 to P03 in March (Fig. 4) showed that the temperature decreased and the salinity increased gradually from surface to bottom. So we revised the statement to “the water column in the middle of ECS shelf was almost well-mixed in the top 100 m, and temperature and salinity along section PN were nearly uniform from the surface to the bottom” in part 3.4, paragraph 2.

Q22. Page 7027, Line 20: Correspondingly: be clear here. There are two processes involved: the annual development stratification, and the gradient (and run off induced permanent stratification in the methane-rich coastal near part.

Reply: It is really a good remind for us. The permanent stratification exists in the CH4-rich coastal area, while in the middle of the ECS shelf, there is an annual stratification development ongoing with the seasonal variation. Thus, the discussion in part 3.4 was revised as “Perennial stratification in the water column occurred in the Changjiang estuary, while water column stratification in the middle shelf began to occur during late spring, faded during the fall and disappeared completely during December.”.

Q23. Page 7027, Line 26: “Surprisingly” is not really scientific. It would be rather

interesting to argue (in the discussion) whether this points to sedimentary sources feeding into the bottom layer.

Reply: We deleted “surprisingly” in the revised paper and added some discussions about the sedimentary CH4 source in part 4.1.

Q24. Fig 5: I would again suggest using at least the same scales for depth and T, S; CH4 would maybe not work here. Also indicate these positions in Figure 1 (by arrows or surrounding circles. Else, one has to search quite a while.

Reply: Figures 1 and 6 were redrawn according to the reviewer′s advice and the positions of these stations were labelled with red circles in Fig.1 to find them easily.

Q25. As for the interpretation of the data shown in Fig. 5, it appears that the occurrence of the hydrothermal activity in the Okinawa Trough and its potential impact on the methane distribution of the deep water is completely neglected (and might be the reason for some of the observations (this should however be in Discussion).

Reply: There were two peaks in the CH4 depth profiles during spring and autumn, and the second peak occurred at a depth of 600 m during May and at 800 m during October (Fig. 5). CH4 concentrations further increased below 800 m, with high CH4 levels occurring in the bottom waters during May and December. Previous studies also showed that many submarine mud volcanoes and hydrothermal vents occur along the continental slope of the ECS (Zhao et al., 2006; Kawagucci et al., 2011). Methane-containing fluid was episodically vented, then transported to the water column. The hydrothermal end-member CH4 has been considered as a thermogenic origin and the CH4 level are 104-107 times higher than those in the ambient ocean water. When released from the vent, it forms buoyant plumes rapidly (Tsunogai et al., 2000). During the dilution, CH4 can be oxidized quickly and quantitatively in the plume in association with the microbial oxidation (De Angelis et al., 1993). The residual CH4 spreads into the upper seawater. We added the discussion about its potential impact on CH4 distribution in the deep water in Part 4.1.

Q26. Page 7028, Line: 9 Citations – make clear that the citations are on subsurface maxima at different places, but not at the pycnocline of the ECS and YS.

Reply: Methane maxima occurred in the upper pycnocline along with the suspended matter maxima in the northwestern Gulf of Mexico (Brooks et al., 1981). The similar observations were also found by Burke et al. (1983) in the eastern tropical North Pacific, but they reported that CH4 distribution had a weak correlation with the particulate and biological parameters, and mainly controlled by the physical processes in the ocean. Outdot et al. (2002) found a sharp subsurface maximum at the bottom of the mixed layer in the equatorial Atlantic, with a chlorophyll a maximum. We described these research results in detail in part 3.4.

Q27. 3.5 Sediment fluxes. While the compiled sediment flux data are a nice data set, the spatial and temporal inhomogeneity is not really supporting a basin-wide extrapolation. Way more critical, however, is that in this case, the seasonal bias of sampling is too strong to allow the conclusion that fluxes are strongest in summer (though this might well. . .. . .). The summer sites are all on the western rim, in shallow waters and relatively near to the outlet of the Chang Jiang, other than for any other sampling campaign. Is there a possibility to look at trends with respect to e.g. bottom water temperature and/or Oxygen, water depth, organic fraction of the sediment). Without thorough scientific thought, there is only very limited use of these data, and in particular the annual pattern of fluxes might be a mere sampling bias.

Reply: We agree with the reviewer that the scarce sampling location might cause a spatial and temporal bias, but we all know that it is difficult to get sediment cores during tough weathers and the sediment incubation experiments involve a great deal of lab work and resources. Due to the limitation of weather and lab resources, our sediment collection in August 2011 mainly covered the coastal areas. To avoid spatial bias, we added the sediment-water CH4 fluxes obtained from a cruise in August 2013 in the manuscript which provides data for the shelf and slope regions. We re-estimated that average sediment-water CH4 flux from the ECS and YS was about 1.06 and

0.73 μmolâĂćm-2âĂćd-1 in 2011, respectively. Based on their surface areas (about 7.7$\times$105 and 3.8$\times$105 km2), the annual CH4 emission from sediments of the ECS and YS in 2011 was about 2.98$\times$108and 1.01$\times$108 mol, respectively. We also updated the related value in the abstract and part 4.2 in the revised paper, accordingly.

Q28. From the first paragraph of the discussion, one might argue that fluxes are even stronger correlated with driving parameters than concentrations. So what about checking on a correlation of CH4 sedimentary fluxes with T (and other parameters, see above).

Reply: We checked the correlation of the average CH4 sedimentary fluxes and average T, which had a weak correlation with T (FCH4=0.06 T+0.03, R2=0.3).

Q29. Page 15, Line 18-19: There is little to no indication for methane production in the deep water column. Even in anoxic waters (e.g. Black Sea, Baltic Sea) methanogenesis is mostly related to the sediments. If the authors want to make this point (which is a requirement for the reasoning on the box model results later on) they should reference this hypothesis well. There is no direct indication of methane production in the water column here, thought this is the main hypothesis based on the model approach.

Reply: During the incubation of bottom waters (the control group in the sediment incubation experiment), CH4 concentration increased linearly with time, which indicated CH4 can be produced in bottom seawater. Moreover, this process can be enhanced by the low oxygen and high organic carbon level (Naqvi et al., 2010; Ye et al., 2015). Extremely high bottom CH4 values occurred near the Changjiang Estuary and outside Hangzhou Bay, such as station D1 (26.22 nM) and C0 (19.92 nM) during August 2011. Apart from sediment emission, in situ production might be another CH4 source, especially in the environment with low oxygen level (2.00-4.00 mg/L) and high particulate organic carbon flux (3900-7300 mg CâĂćm$-$2âĂćd$-$1; Hung et al., 2013) in the bottom waters. Here we just tried to explain the possible reason for the high CH4 value in the bottom and we think that in situ seawater CH4 production was a possible source.

To make the description clear, we rewrote this part.

Q30. Page 16, line 2-3: While I believe this argument, the modelling in 4.2 suggests a rather minor impact of transport and in situ production as a mean driver.

Reply: According to our estimation, the three main CH4 sources might be in situ seawater production, SGD and CH4 import from TWCC in summer, while in winter, the strength of certain sources varied and sediment emission exceeded TWCC, acting as one of the three main sources. However, the above results only indicate that in situ CH4 production in the water column and the groundwater discharge together make greater contribution to CH4 in the ECS compared to water transport process; and it still needs more data to identify which one is the key factor in the biogeochemical CH4 cycle in ECS.

Q31. Page 16, Line 21-22: Which might be problematic, as groundwater can be a major contribution to the methane sources, and will, when neglected, be counted as the only "missing source term", e.g. water column production. The authors neglected groundwater discharge due to a lack of existing data, and also did not consider methane from deep sources, e.g. the Okinawa Trough. In the following part of 4.2, they attribute the mismatch of sources and sinks in the model budget to production in the water column. However, they do not provide any support for this source. Neither there are any production rate measurements in the text, nor a spatial analysis of the water column data pointing to this kind of source.

Reply: We agree with the reviewer′s opinion that it was really improper that we did not take the groundwater into account, even if lack of reference data for calculation. Actually, we measured some groundwater samples along the coast of the ECS in December 2011 and July 2012. During December 2011, these groundwater CH4 samples were collected at twenty-five sites along the Jiangsu and Zhejiang coastal region (120.857∼121.896°E, 30.124∼30.956°N), which can also be considered as the fresh groundwater end-member. CH4 concentrations varied significantly at these stations

and had a great range of 33∼61602 nM, with a median of 271 nM (unpublished data, Zhang et al.). During July 2012, we collected groundwater samples for CH4 measurement along the coast of the ECS (121.371∼121.934°E, 30.733∼31.976°N; n=8). It indicated that the median CH4 concentration was 758 nM in summer, with a great range of 138∼3428 nM, which was much higher than those in winter.

We didn't show these data in the previous edition just because we have no data for SGD, hence we can't estimate the CH4 input via groundwater. In the latest version, we tried our best to collect the information about the groundwater discharge to the ECS and gave a rough estimate of CH4 input via groundwater. We contacted Prof. Jinzhou Du at East China Normal University and obtained the update submarine groundwater discharge (SGD, including submarine fresh water discharge and recirculated saline groundwater discharge) into the ECS to be 0.68×108 m3 d-1 and 0.46×109 m3 d-1 in dry and wet season, respectively, using Ra isotopes as tracers (Wang and Du et al., unpublished data, personal communication). It is hard to measure CH4 concentrations in the recirculated seawater, so we assume that CH4 concentrations in the fresh groundwater and recirculated saline groundwater are the same. Median CH4 concentration was chosen for calculation since it is less susceptible to abnormally high CH4 concentrations observed in the groundwater. Hence, CH4 fluxes via submarine groundwater discharge were estimated by multiplying the median CH4 concentration by the SGD, which yielded a flux of 0.21 mol/s and 4.01 mol/s for winter and summer, respectively. SGD is a mixture of fresh groundwater and recirculated seawater, among which the latter could account for 90% of the discharge or more (Burnett et al., 2006)., but considering CH4 concentration in the porewater (∼0.45 μM) of the ECS usually is much lower than those in fresh groundwater, the above estimation of CH4 flux via submarine groundwater discharge may be overestimated to some extent. However, our results suggest that groundwater discharge might be an important CH4 source for the ECS, especially in summer. We added the above discussions about the groundwater CH4 to part 4.2 in the revised paper and also updated the abstract and conclusion accordingly.

Although the hydrothermal vent is a potential source of CH4 in the deep water, it is only reported sporadically for the Okinawa Trough on the edge of the continental shelf and it's difficult to estimate its emission rate. Hence we think that it is reasonable to ignore such a minor source in our budget and only indicated it might be a minor source for CH4 in the manuscript.

Q32. They cite del Valle and Karl 2014, but would need a larger production term and the process would definitely result in a very distinct surface layer maximum dominated production term in the subsurface layer.

Reply: del Valle and Karl (2014) calculated the CH4 budget in the mixed layer at station ALOHA (in the open sea, north of Hawaii) and estimated the net CH4 production rate in the mixed layer ranged from 0.008–0.047 μmolâĂćm-3âĂćd-1 from September 2012 to September 2013. In this paper, we used to compare the CH4 production rate in ECS with it. However, we deleted the discussion on the net CH4 production rate in the revised manuscript, since it made little contribution to the discussion.

Q33. In the conclusions, the problems with the data interpretation gets really obvious: - the seasonality in distribution and emission might be correct, but the approach does not really support this due to a scientifically unprecise handling of the problem of sampling bias, both for the water column data grid and the sediment flux stations - the production of methane in the water column (attributed to amount for 70% here!!) is not supported by the content of this paper AT ALL. Rather, possible reasons are – uncertainty in the existing numbers; - importance of other external sources such as groundwater seepage or hydrothermal input in the deeper waters; - a problem with the assumption of a steady state for a single year, potentially others.

Reply: In the latest version of manuscript, we changed our data processing method and divided the sampling region into four subareas, including the Yellow Sea, the Changjiang estuary (S<30), the ECS shelf (S >30, depth <200 m) and the ECS slope (S >30, depth >200 m). We discussed the CH4 seasonal variation in ECS shelf, which

was covered by all surveys. Similarly, we also reported the range and mean sea-air flux density for each area during the four cruises (Table 3). An average area weighed sea-air fluxes from the ECS and YS was calculated to avoid the potential spatial bias.

We reconsidered the CH4 sources and sinks in this area, and added some CH4 data for groundwater along the coast of the ECS and estimated CH4 input via submarine groundwater discharge preliminarily. In the revised paper, we also discussed the great uncertainties about this estimation in detail. Firstly, there were potential errors involved in the measurement methods and calculations of sea-air fluxes. Secondly, the estimation of CH4 input via SGD was quite crude due to the limited available data about SGD and groundwater CH4. Thirdly, we performed sediment incubations only at several stations during each survey, and the results were far from representative of sediment emission from the whole ECS due to large spatial and seasonal variations. Finally, some CH4 sources and sinks were neglected in this estimation. For example, aerobic CH4 oxidation has been shown to be a substantial sink of CH4, however, no estimates of methane oxidation extents and rates were made in the water column here. Seepage of thermogenic methane from the sediments was also ignored due to lack of data and hard to estimate. Hence the role of net in situ microbial production might be exaggerated by estimating it as the difference between sources and sinks. Although our method of estimation was not perfect, we demonstrated a variety of CH4 sources and sinks for the ECS and roughly estimated their relative contributions. It suggests that in situ production in the water column and sediment release are major CH4 sources in the marginal shelf seas, while sea-to-air release was the major external sink of CH4 in the ECS. Groundwater might be an important source of CH4 in the ECS, especially in wet seasons. Hence more research work on CH4 production and consumption as well as CH4 in groundwater discharge is needed to further understand the CH4 budget in the ECS.

We rewrote the conclusions thoroughly: CH4 distribution and emission in the ECS and YS had obvious spatial and seasonal variations, and were influenced by various fac-

tors, including mixing of different water masses, water temperature, freshwater input, sediment release, hydrothermal seepage and oxygen levels in the water column. We estimated the CH4 budget of the ECS using a box model, and the results indicated that in situ seawater production and sediment release might be the major sources of CH4, while sea-air exchange was the major external sink of CH4 in the ECS. Groundwater might be an important source of CH4 in the ECS, especially in wet seasons. The ECS and the YS together was estimated to release about 4.09×109 mol of CH4 per year into the atmosphere, which accounts for about 0.45% of the global oceanic emission and was much higher than its corresponding area proportion of 0.32%. Hence the YS and ECS were active areas for CH4 production and emission.

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