

## Interactive comment on "Synthesis of observed air–sea CO<sub>2</sub> exchange fluxes in the river-dominated East China Sea and improved estimates of annual and seasonal net mean fluxes" by C.-M. Tseng et al.

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We thank the reviewers for their helpful comments and suggestions; their critiques have been addressed carefully, and the manuscript improved. The following is mainly detailed point-by-point responses to the reviewer #2's comments.

General comments

2A-1. Tseng et al. conduct 12 mapping cruises in the East China Sea (ECS) shelf between June 2003 and July 2011, as well as an earlier cruise in July 1998. They propose

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an empirical function mainly based on their summer data, which predicts areal mean sea surface pCO2 over the entire ECS from SST and Changjiang river discharges. And then they simulate time series of monthly mean pCO2 and air-sea CO2 fluxes in the ECS over the 14 years from 1998 to 2011. Finally they sum up the model results and deduce their synthesis flux estimation. The fundamental linear relationship between areal mean NpCO2 and Changjiang river discharge has been published elsewhere (Tseng et al., 2011).

We firstly thank the reviewer #1's comments on the empirical findings we proposed. However, the highlights of the manuscript we were focused on are quite different from the previous paper which was related to effect of the Changjiang runoff on the CO2 uptake of the ECS. In our current paper, we constructed the 14-yr time-series CO2 variability, identified distinct seasonal pattern, characterized the long-term changes and further obtained the reliable magnitude of the air-sea CO2 exchange flux in the ECS through the model and observation. Then we did the comparison of our results to other published ones (Peng, et al., 1999; Tsunogai et al., 1999; Wang et al., 2000; Shim et al., 2007; Zhai and Dai, 2009; Chou et al., 2009, 2011; Tseng et al., 2011). The results we provided are more representative and better than previous estimates which were biased by inadequate spatial and/or temporal coverage. In addition, we provided more new data and expanded the study period from 1998 to 2011 to strengthen the algorithms for predicting water pCO2 (pCO2w) in the ECS. Such an amazing and valuable dataset of a decade can't be found elsewhere. The results are thus novel, and precious. It will be attractive to those who are engaging the air-sea CO2 study in the river-dominated marginal seas.

2A-2. If we simply consider the ECS as a whole in a large-scale or even the global context, it is attractive. However, at first they should confirm its regional validity by both data assurance and scientific demonstration.

We are grateful that the reviewer accepts the merits of this study. We did consider the ECS as a time-series standing-point, just like the reviewer mentioned as a whole in a

large-scale or the regional/global context. The reason for doing that is to simplify the issues of spatial and temporal heterogeneity. As we know, the previous estimates of air-sea CO2 exchange existed large biases due to limited tempo-spatial coverage. The potential problems hinder us to effectively achieve the reliable CO2 flux estimates at the same time regarding the field observation. Therefore, our strategy firstly carries out the wide-area CO2 mapping in a seasonal basis for a decade to cover the areal differences and temporal variations as possible as we can. We may then find their general features, controlling processes and weighted-mean contributions. Consequently, we develop an empirical algorithm for computing areal mean sea surface pCO2 over the entire ECS and then to generate the representative values. The modeled pCO2w we calculated were in excellent agreement with observed areal means (r2=0.9, n=13). It indicates the performance of the empirical algorithm applied to the entire ECS is well confirmed. Comparison of modeled and observed data demonstrated the ability of the model to capture the seasonal variability in pCO2w in the ECS.

Results from our algorithm were further validated by our field observations of in-situ underway pCO2 with high spatial and temporal resolution. The model monthly mean fCO2w results agreed well with the observed monthly mean pCO2w (R2=0.90, n=13; Fig. S3), which were based on thousands of individual data. Besides, we have multi-year records revealing significant inter-annual variation. Therefore, this assessment of annual CO2 uptake surpasses all previous estimates (1-3 mol C m-2 y-1) [Chen and Wang, 1999; Peng et al., 1999; Tsunogai et al., 1999; Wang et al., 2000; Shim et al., 2007, Zhai and Dai, 2009] and it is more reliable because our better spatial and temporal coverage as a whole system reduce uncertainties.

In brief, we simplify the complexes, verify the model reliability, provide the reliable timeseries data, first-orderly constrain CO2 fluxes and finally know the role of the riverdominated ECS in absorbing the atmospheric CO2. We believe the data we present no matter what are from observation or by model are high quality and assurance. High resolution pCO2 samplings were carried out by automated underway measurements

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during the ECS cruises. Routine accuracy and precision achieved with this method are about  $\pm 2$  and  $\pm 0.1 \mu$  atm, respectively.

2A-3. As a regional study, they skip several significant issues, such as spatial heterogeneity in the ECS and decadal changes of nutrient discharges from the Changjiang River.

We fully agree the reviewer's critical comments that spatial heterogeneity in the ECS and decadal changes of nutrient discharges from the Changjiang River may influence the air-sea CO2 exchange in the ECS. To consider the ECS as a single unit is based on two considerations: (1) This is a holistic view on the biogeochemical responses of the ECS to external drivers (Changjiang discharge in summer; cooling and wind mixing in winter); and (2) This is a simplified approach to address a very complex system, but we feel that this is such an important issue that a simplified overview is a worthy effort before a full-fledged modeling approach can be launched. Regarding the temporal changes, the changes in nutrient loading should have been implicitly included in the data of water discharge.

Gong et al. (2006) observed the nutrient ratio changes and reduction of primary production in the ECS because of the Changjiang discharge decrease in summer, 2003. Further, the ECS ecosystem responded to changes in the nutrient supply in the plume area. As a result, nutrient levels with the nutrient ratio in relation to the induced phytoplankton biomass growth and further community structure shift shall indeed affect the CO2 uptake in the ECS.

Tseng et al. (2011) highlighted the plume is the most important issue in summer. That means the primary factor, determining the magnitude of the ECS CO2 uptake, is the plume area size which contribution is area-weighted relative to the whole ECS area. The more the plume area presents, the more CO2 uptake area shall be observed. Our further results pointed primarily out the importance of the plume area change, governed by the Changjiang discharge change (see the following figure A2-1). We

observed, for instance, there was a huge rive plume in July of the 2010 flooding year (OR1-932) which area is almost 10 times larger than that in the normal year. The  $\ddot{R}d'pCO2$  in the plume about -135 uatm is slight less than -100 uatm in the normal on average. Further, the plume N+N level (~7  $\ddot{R}M$ ) averaged less than those (~10  $\ddot{R}M$ ) in normal year due to the dilution effect. The same is for the Chl-a (flood, 1.6 mg m-3; normal, 3.8). However, the CO2 uptake on the surface is much larger than that in the normal year since the plume area almost covering the whole ECS. The areal means of N+N and Chl-a by integrating over the survey area in the flooding July are higher than those in normal year as well. That is why we got such a good correlation between river discharge, plume area, and CO2 flux. Apparently, the amount of the CO2 uptake in the ECS shelf in summer is determined by the plume area (Salinity≤31), in which there were high nutrients and high biomass growth. The riverine nutrients that are enriched in the plume fuel rapid phytoplankton growth, drawing water fCO2 down. The results thus highlight that importance of the Changjiang discharge with induced plume area on the summer CO2 uptake in the ECS.

2A-4. Although their rich datasets are valuable, the manuscript fails to expand our understanding of the ECS CO2 flux.

We did collect the rich and valuable dataset over a decade. In the manuscript, we broadly and simply assess the air-sea CO2 exchange flux, improve flux estimates, examine annual and interannual variability, characterize the long-term changes and then recognize the role of the ECS in regulating the anthropogenic CO2 relative to the global ocean. The detailed examination of CO2 cycling enhances understanding of the behavior and fate of carbon within the productive ocean margins will be done cruise by cruise in the following work.

2A-5. I can't recommend it for Biogeosciences to be published as this form. They should be encouraged to re-submit a new version after substantially considering below issues.

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Finally, the manuscript will be revised based on the reviewer's comments suggested below.

Specific comments:

2B-1.Before in-depth analyses and reductive calculation, the primary data set (field measured data) should be fully presented, at least in supplementary materials.

We'll do our best to present the primary dataset with summary information in supplementary materials.

2B-2. They have reported the January 2008 data in Chou et al. (2011; 2013), and the July 2011data in Chou et al. (2013). They also present their summer data from 2003 to 2010 (7 cruises) in Tseng et al. (2011). In this manuscript, they only briefly present the cruise averaged values in Table 2 and Fig. 4, even for their new datasets obtained in July 1998, June 2005, November 2006, and May 2009. The only data map is a composite map of summer surface water pCO2, which is insufficient for readers to assess the data basis of their study.

As mentioned above, we'll present the primary data in figures and tables in supplementary materials.

2B-3. As a seasonality-based synthesis research, their seasonal surveys are quite imbalanced. If possible, more spring/fall/winter data sets should be incorporated, such as pCO2 data in November 2011 (Chou et al., 2013).

There are no problems to add the fall pCO2 data in November 2011 (OR1-980) into the manuscript.

2B-4. The effects of vertical mixing on sea surface pCO2 and air-sea CO2 fluxes also need to be discussed in main text, instead of being presented in supplementary materials.

We'll move the discussion of the mixing effects from supplementary materials into the

main text.

2B-5. The ECS is subject to great heterogeneity (e.g. Chou et al., 2009). And the Changjiang river plume mainly affects the northern ECS and the southern Yellow Sea in summer. The simplified relationship between areal mean NpCO2 and Changjiang river discharges over the entire ECS (i.e. Eq. (3)) should be better examined.

We fully agree with the reviewer's suggestions. In our study, the northern part of the Changjiang river plume is not all covered by our current dataset since many of the cruises cover only half of the ECS. However, the Changjiang river plume is the most important feature in the CO2 uptake processes in the warm season, and has been well observed on our cruises i.e., most of it has been captured in observations. As a first approximation, the peripheral regions to the north and south of the plume are more or less symmetric.

We have, therefore, examined whether the cruise data are representative of the entire ECS. Briefly, the areal mean SST calculated from observed values in cruise survey area correlated well with the average AVHRR-SST data in the entire ECS. Other areal means of pCO2 and hydrographic parameters (e.g., SSS, N+N, Chl-a) were also representatives for the ECS. Accordingly, we may assume the two halves are more or less symmetric and, therefore, the southern shelf is representative of the whole ECS shelf. The simplified relationship between areal mean NpCO2 and Changjiang river discharges over the entire ECS should be further applied and representative.

2B-6. The nutrient basis of Eq. (3) has been briefly discussed by the authors (Tseng et al., 2011). However, in this study they ignore this issue.

We'll recover the issue raised by reviewer. Our results firstly show the water pCO2 well correlated with the magnitudes of the river plume area and of Changjiang discharge and the amounts of N+N and of Chl-a in summer (see Table A2-1). Then, temperature-normalized pCO2 (NpCO2) during the summer productive period also has a good relationship with Changjiang discharge. That means the CO2 change in summer in the

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ECS is mostly related to biological uptake relative to temperature effect (i.e., at a constant temperature range in certain summer months). The riverine nutrients that were enriched in the plume fuel rapid phytoplankton growth, drawing water fCO2 down. The results thus highlight that importance of the Changjiang discharge with induced plume area on the summer CO2 uptake in the ECS. Changes in summer CO2 uptake in the ECS shelf shall be governed by the Changjiang discharge i.e., significant source of CO2 can occur at low flow rate less than ca. 39000 m3/s and correspondingly low plume area below 5x103 km2.

2B-7. Significantly, the filling of the Three-Gorges Dam (since June 2003) have impacts not only on monthly and/or seasonal settings of the Changjiang river discharge, but also on riverine exports of nutrients (e.g. Chai et al., 2009). Since Tseng et al. have only one cruise from 1998 to 2002, the model results during this period should be removed. The Changjiang river plume is also subject to intra-seasonal variations (e.g. Tseng et al., 2011).

We fully agree with the reviewer's comments. The Changjiang river plume is also subject to intra-seasonal variations. Tseng et al. (2011) demonstrated the Changjiang river plume size is directly governed by the Changjiang river discharge. So, intraseasonal variations in plume area were observed due to intra-seasonal changes in the Changjiang river discharge. Additionally, both water pCO2 and NpCO2 during the warm period all well correlated with Changjiang river discharge. The discharge, which governs the nutrient inputs into the ECS, is primary factor to control the CO2 uptake of the whole ECS. In summary, the focus on NpCO2 during the summer productive period is bound to emphasize the river's effect (i.e., high river discharge, causing high nutrient inputs and induced the phytoplankton growth) on biological export. Therefore, although Tseng et al. have only one cruise from 1998 to 2002, the general feature of river effect on the CO2 uptake in the river-dominated ECS system shall be applicable. The model results during this period should not be affected and then draw more attention the 1998 flood event. We believe it doesn't have much influences by the enhanced nutrient loading and induced eutrophication. The study period shall be acceptable and credible.

2B-8. A time lag is needed between Changjiang river discharges at the Datong Station and their biogeochemical effects in the ECS (Kim et al., 2009).

Kim et the al (2009) mentioned there is possibly a time leg according to the remotely sensed data of the Chl-a. Such satellite data in coastal estuarine systems, however, exhibit potential limitations in accuracy so that the estimates had some extents of uncertainties. Anyway, the time lag was roughly averaged about 3 days from the Datong Station to the ECS ( $\sim$ 600 km) at a river flow of 2.5 m/sec which is according to the calculation of averaged summer discharge divided by the surface area of river channel. Once the riverine waters get into the ECS, the nutrients in the plume cause the phytoplankton growth instantly and biological responses are very fast within hours under the conditions of warm, nutrient-enriched supply, and light enough in the surface.

2B-9. Their model data in summer 1998 does not match their real datum in July 1998 (Fig. 4), which is interesting. It is likely that the excess nitrate load during the catastrophic 1998 flood is not effectively transferred into low pCO2, due to the insufficient phosphate supply (Wang and Wang, 2006).

Thanks for reviewer's precious comments. The 1998 cruise we performed was between 6/30 and 7/6 1998, which discharge (ca. 55000 m3s-1) is less than the monthly average in July 1998 (ca. 75000 m3s-1). We found that the discharge actually reached a peak flow occurred from mid-July to the end of July instead of from the beginning of the July. So, it has been questioned whether the information collected in the 1998 cruise can represent an entire month of July. We can be, anyway, sure that great differences between the average discharges during the cruise and in July fairly resulted in data mismatch.

2B-10. The July 1998 data should be separately published.

That will be done in the next work.

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Technical corrections: 2C-1. The statistics are confusing. In their Eq. (1) and Eq. (2), n = 8. In their Eq. (3), n = 7 according to Fig. 3. In Fig. 5, n = 11. In Fig. S4, n=13. Why and how to select data?

We present a total of 10 cruises performed in summer, including nine cruises between 2003 and 2011 and one cruise in 1998. In the Eq (1) & (2), we skipped one cruise affected by Typhoon in Jun 2003 and one in 1998. We may add the data of the 1998 cruise in, so total number of cruises will be nine. In the Eq (3) & Fig 3, we took the monthly averages in January (n=1 cruise), May (n=1), June (n=2), July (n=5), August (n=1), November (n=2) and two separated flood events (n=2) so that there are a total of 8 points. In Fig. 5, n shall be 14 but two points in the cruises of 2006/11 and 2011/7 are too close and overlapped to distinguish each other. In Fig. S4, n will be 14.

2C-2. Materials and methods, why to analyze chlorophyll a and nutrients? Delete them?

We'll add the chlorophyll-a and nutrient data in and made the statistic correlation analyses (Table A2-1) and figures (Fig. A2-1) to demonstrate the biological CO2 exports in warm periods.

2C-3. P. 13981, L. 19: what is Wanninkhof (1993)?

It shall be Wanninkhof and Thoning (1993).

2C-4. Table 3, the two Wanninkhof (1992) equations should be exchanged with each other.

The short-term and long-terms equations have been indicated clearly.

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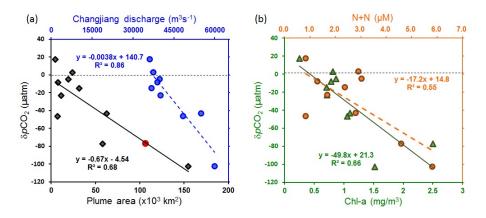


Fig. 1. Figure A2-1. Relationships between the amounts of dpCO2 and magnitudes of the CDW plume area and of the Changjiang discharge (a) and N+N and Chl-a (b) in summer from July 1998 to July 2011 in the ECS

	*Changjiang discharge	Plume area (S<31)	Sea-to-air CO <sub>2</sub> Flux	δpCO <sub>2</sub>	pCO <sub>2waa</sub>	N+N	Chi-a
Changjiang discharge	1.00						
Plume area	0.91	1.00					
Sea-to-air CO2 Flux	-0.97	-0.89	1.00				
δpCO <sub>2</sub>	-0.92	-0.83	0.94	1.00			
pCO <sub>2water</sub>	-0.98	-0.80	0.94	0.96	1.00		
N+N	0.70	0.88	-0.71	-0.75	-0.73	1.00	
Chl-a	0.88	0.60	-0.65	-0.81	-0.82	0.72	1.00

Table A2-1 Correlation coefficient matrix for areal - means of pCO<sub>2</sub> and other environmental variables in the ECS shelf in summer months from 1998 to 2011 (n=10)

 Chl-a
 0.88
 0.60
 -0.65
 -0.81
 -0.82
 0.72
 1.00

 \*Average Changjiang River discharge data collected at Datong station during the LORECS cruise period. Regression results between other variables and Changjiang discharge were obtained for the ten cruises, not including OR1-686 (6/19~6/26, 2003).

 Before that cruise, the typhoon SOUDELOR (6/16~6/18, 2003) passed over east of the ECS shelf toward Korea and Japan. Such a typhoon effect anomalously induced the more diluted plume area through enhanced rainfall in lower watershed of Changjiang River below Datong station. However, high typhoon-induced CRD was not recorded at Datong which is 624 km upstream from the river mouth.

Fig. 2. Table 2A-1

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