

## ***Interactive comment on “Rapid increasing trend of CO<sub>2</sub> and ocean acidification in the surface water of the Ulleung Basin, East/Japan Sea inferred from the observations from 1995 to 2004” by J.-Y. Kim et al.***

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### General Comments

The manuscript presented twelve mean pCO<sub>2</sub> data from field measurements (four been published before, eight present in this study) between 1995 and 2004. It investigated the CO<sub>2</sub> increase and ocean acidification in the surface waters of the Ulleung Basin (UB) of the East/Japan Sea. The author estimated rates of increase of fCO<sub>2</sub> were 3.36 μatm yr<sup>-1</sup> for the surface ocean and then the ocean acidification trend es-

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timated to be 0.04 pH units decade<sup>-1</sup>. Both estimated rates are double-times faster than the global mean. The estimations, however, seem to be questioned since they may have large uncertainties/errors involved. The reasons are listed as below: (1) The areal mean of the cruise in terms of data accuracy and representativeness may be, for instance, problematic listed in Table 1; (2) That caused the authors with wrong estimations of trend rates and then overestimated only based on the limited periodic survey instead of decadal time-series measurements; (3) They made the big mistakes through the data making from one dataset to generate the one another data. The more data generated from one dataset through statistical method resulted in the final results may not be true. Overall speaking, they over-estimated by the wrong estimation.

RESPONSE: We added more data sets available (data sets observed in November, 2008 and in July, 2009). And re-analysis the data sets using harmonic function analysis not the simple linear regression for the annual increasing trend. Therefore, the values are changed. For example, the long-term fCO<sub>2</sub> increase is changed to 2.7 μatm/yr from 3.36 μatm/yr. The title is also changed to “Long-term trend of CO<sub>2</sub> and ocean acidification in the surface water of the Ulleung Basin, the East/Japan Sea inferred from the underway observational data”. For the reviewers’ information the revised manuscript is attached in the form of a supplement. Please see the supplement attached.

### Specific Comments

1. P9592 in Table 1: data accuracy and representativeness? Is it representative for the cruise fCO<sub>2</sub> mean? The authors did not discuss how they averaged. Since the spatial coverage of the cruise track was not all the same (Fig. 1), it may cause the weighted-mean or arithmetic mean biased. Further, the data in Table 1 for ΔfCO<sub>2</sub> are somewhat doubtful. Please see the following table we made in the column of C-D in red. The ΔfCO<sub>2</sub> data provided by authors are different from those we calculated from the difference between fCO<sub>2</sub>sea and fCO<sub>2</sub>air in A and B column, respectively. The authors shall explain why caused such differences.

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RESPONSE: The values are arithmetic mean. The table caption is modified.  $\Delta fCO_2$  values in the table are averaged value of the  $\Delta fCO_2$  calculated from each  $fCO_{2air}$  and  $fCO_{2sea}$  individually. When we calculated  $fCO_2$ , we applied average value of prior and posterior  $fCO_{2atm}$  data to  $fCO_{2sea}$  data, since we don't have both data at the same time and the  $CO_2$  variation in the atmosphere is much smaller than in the seawater. Hence the numbers could be different.

2. P9596 in Fig. 2: The annual rates of  $fCO_2$  in seawater ca.  $3.36 \mu atm yr^{-1}$  may not be right since the limited sampling in the marginal seas where exhibit large seasonal fluctuations in terms of complex currents and biogeochemical variability and inadequate spatial survey areas where are not the same for each cruises were involved. Therefore, the end results shall be wrong once the increasing rate was not correct.

RESPONSE: It is quite difficult to understand since the data include large seasonal variation as well as long-term trend. Hence we added more data sets available (data sets observed in November, 2008 and in July, 2009). And re-analysis the data sets using harmonic function analysis not the simple linear regression for the annual increasing trend. Therefore, the values are changed. For example, the long-term  $fCO_2$  increase is changed to  $2.7 \mu atm/yr$  from  $3.36 \mu atm/yr$ .

3. P9579 & 9580: Reliability of time-series algorithms of  $fCO_{2sea}$  ? The time-series equation proposed by authors was initiated on a basis of two assumptions: 1. Decadal trend was constant; 2. Seasonal variability constant. Assumption 1 seems to be somewhat acceptable, but annual changes shall be greatly fluctuated in an offshore near coastal area of marginal sea. So, regarding the assumption of constant interannual variability we hold great suspicions. Anyway, authors didn't examine if the assumptions were proper or not before further algorithms establishment. The establishment of time-series algorithm for computing  $fCO_2$  was based on (1):  $fCO_{21995} = fCO_{2in-situ} - ra (Year - 1995)$ . where  $ra$  is  $3.36 \mu atm yr^{-1}$ , the slope of the regression line of  $fCO_2$  against year as shown in Fig. 2. Obviously, it is quite inadequacy of data sampling to have a representative trend, being further used to determine the ocean acidification

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rate. Consequently, the results showed ocean acidification rate in the East/Japan Sea was 2 times higher than the global mean according to the increasing rate of  $3.36 \mu atm yr^{-1}$ . It may be, however, overestimated on a basis of a wrong trend rate. So, if we amend the rate,  $ra$ , to the increasing rate of atmospheric  $fCO_2$ ,  $1.9 \mu atm yr^{-1}$ , equation (2) could be solved in the text:  $fCO_{21995}(t) = C_0 + C_1 \sin(2\pi t) + C_2 \cos(2\pi t) + C_3 \sin(4\pi t) + C_4 \cos(4\pi t)$   $C_0= 333.46, C_1= -38.02, C_2= 18.16, C_3= 5.96, C_4= 23.33$  ( $R^2=0.85$ ) where the  $R^2$  is even better than that for seawater ( $R^2=0.82$ ) observed at the  $ra = 3.36 \mu atm yr^{-1}$ . We can then trace back the in-situ  $fCO_2$  as  $fCO_2^*$  by use of the equation (3):  $fCO_2^* = fCO_{21995}(t) + ra (Year - 1995)$ . The outcome correlated well with the observed data which correlation coefficient is slightly higher than that at the ratio of 3.36. So,  $ra$  proposed in the text could be further refined/ constrained. Fig. 1 In further analysis, assuming observed  $fCO_2$  data are representative, we can solve the optimal solution for  $ra$  in combination of equations (2) and (3)  $fCO_2^* = C_0 + C_1 \sin(2\pi t) + C_2 \cos(2\pi t) + C_3 \sin(4\pi t) + C_4 \cos(4\pi t) + ra (Year - 1995)$  Therefore, we solve the five constant coefficients and  $ra$ , when the observed  $fCO_2$  data present in the text brought into the above formula:  $C_0= 351.10, C_1= -42.42, C_2= 20.49, C_3= -7.98, C_4= 28.32, ra=-1.17$  ( $R^2=0.88$ ) The results showed an annual rate is negative, i.e., a decreasing trend. It indicated that there were problems of insufficient sampling in this study and difficulties of providing enough evidences to assure their assumptions and verify final results.

RESPONSE: To confirm the annual increase of  $fCO_{2sea}$ , we added more data sets available (data sets observed in November, 2008 and in July, 2009). And re-analysis the data sets using harmonic function analysis not the simple linear regression for the annual increasing trend as follows;  $fCO_2(t) = c_0 + c_1 \sin(2\pi t) + c_2 \cos(2\pi t) + c_3 \sin(4\pi t) + c_4 \cos(4\pi t) + c_5 t$  (1) where  $t$  is year. The set of six constant coefficients,  $c_0, c_1, c_2, c_3, c_4,$  and  $c_5$ , ensured a satisfactory fit of the harmonic function of Eq. (1) to the observed data ( $R^2 = 0.78$  for seawater,  $R^2 = 0.96$  for air), when the constant coefficients were equal to these values  $c_0= -4977.46, c_1= -29.97, c_2= 0.88, c_3= -1.45, c_4= 23.41, c_5= 2.66$  (for seawater),  $c_0= -3308.75, c_1= 2.51, c_2= 7.22, c_3= -2.45, c_4=$

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-1.46,  $c_5 = 1.84$  (for air). Among the results, the  $c_5$  value which means annual trend for  $fCO_2^{sea}$  was estimated to be  $2.7 \mu atm yr^{-1}$  and that for the  $fCO_2^{atm}$  was  $1.8 \mu atm yr^{-1}$ .

4.P9580: Is it true the statement that the flux had been decreased about 28 % during the last decade according to comparison results of the data obtained in 1995 with that in 2004? Why were the annual integrated  $CO_2$  fluxes estimated by Choi et al. (2012) and Oh et al. (1999) much larger than your results? Whose values are right? What causes the discrepancies?

RESPONSE: We added a sentence about the discrepancies as; "The transiency of their observations could lead overestimation in  $CO_2$  flux because of fewer observations (less than four times) despite of the complexity of the monthly variability and significant seasonal amplitude in  $fCO_2^{sea}$  in the UB."

Please also note the supplement to this comment:

<http://www.biogeosciences-discuss.net/10/C5260/2013/bgd-10-C5260-2013-supplement.pdf>

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