Title: Role of regression model selection and station distribution on the estimation of oceanic anthropogenic carbon change by eMLR.
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General comments

The manuscript shows a careful statistical analysis of the eMLR method which is used to estimate anthropogenic carbon changes. The eMLR method has been applied mainly along repeat hydrographic sections. The authors have evaluated it using known anthropogenic signal from the output of a global circulation and biogeochemistry model. The analysis was done in the North Atlantic arguing that it contains about one third of the anthropogenic global signal, but it should be reminded that it is also affected by strong decadal climate variability (the North Atlantic Oscillation -NAO). The authors claimed that the eMLR produces good results with accuracy typically better than 10 %, although they put attention in the selection of the model regression and the balance of the station distribution. However, very little differences were found between different linear formulations.

The manuscript has a very detailed analysis of many statistical aspects of the eMLR method. The manuscript is well written and structured. The authors use an exhaustive statistical analysis of the optimum regression formulae, and also analyse the problem related with the representativeness of the section in order to get the changes in the anthropogenic CO_2 (Cant) at basin scale. They compared the total CO_2 using the grid of GLODAP centred in 1995 (during a NAO high period) and the CLIVAR grid centred in 2005 (during a period of low NAO) to get the anthropogenic signal. The different physical processes underlying both scenarios put in question one the fundamentals about the use the eMLR methods ("*Assuming that suitable empirical regression models can be found for DIC and that the physical and biogeochemical processes underlying the model are stationary and not affected by the anthropogenic perturbation*"). In fact the results shown in the manuscript described a significant correlation between the natural CO_2 and the Cant. Thus, although the manuscripts tackled about statistical tools to improve the quality of the eMLR method, it did not attack one of the most critical points of this technique that affects clearly the typical underestimation of eMLR method. In fact, the authors have in their hands the information to address this issue that causes the most important biases in the eMLR results.

Specific comment

I would like to stress on three issues: the premises of eMLR described in the manuscript, seasonal variability and the vertical coordinates.

1.- eMLR assumptions. As the authors stated:

"Assuming that suitable empirical regression models can be found for DIC and that the physical and biogeochemical processes underlying the model are stationary and not affected by the anthropogenic signal, the noise can be filtered out and the anthropogenic signal revealed as the difference between model predictions of DIC at different times (Friis et al., 2005)"

However authors show several features that contradicts this assumption. Several figures show that Cant and the anomalies of Cant show some correlation with the natural CO_2. For instance, in Pag 14610 lines (21-26) authors claimed : '*The error pattern of Fig. 7g–i look similar to the column inventory change pattern resulting from the natural carbon run (Fig. 1f), although with absolute errors of overall smaller magnitudes than the vertically integrated natural carbon change. This pattern similarity indicates that, while eMLR accounts for some of the natural variability, large scale natural variability patterns are not fully corrected for, even when the statistically best models are used systematically*'

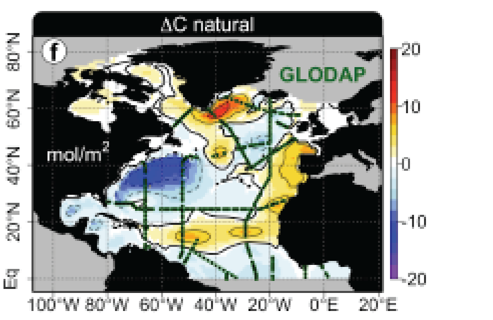


Fig 1f:Changes in natural CO2 inventories

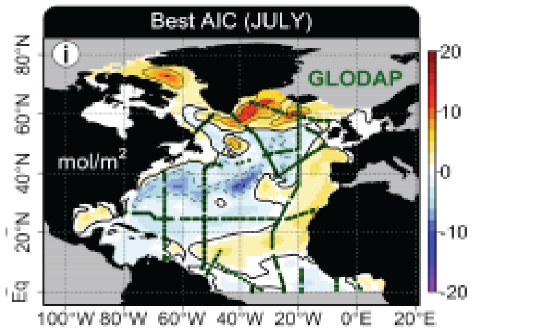
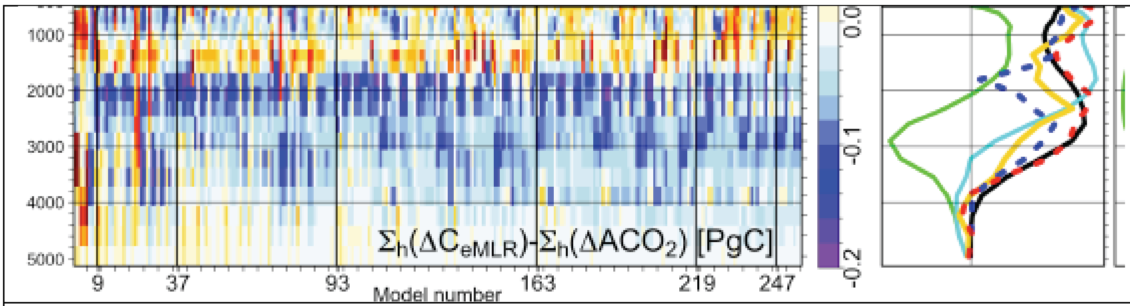


Fig 7 i: Absolute errors in calculated Cant eMLR for 'best AIC'

This fact is associated mainly to the natural variability of LSW in the layer between 1500 and 3500 m, where most of the errors (underestimation, Fig. 6) in the basin-scale inventory change estimates are found. The authors find [Page 14612 (lines 14-21)] a notable correspondence in a layer with high values of AIC around 2000m which corresponds to a layer of systematic underestimations of Cant (Fig. 8a). The underestimation in this layer generates most of the error in the basin-scale inventory change estimates. In this layer, the negative biases of the Cant change inventory (yellow lines vs black lines in Fig8) are associated with the negative layer inventory change of the natural carbon simulation (green line in Fig. 8). The authors claim for '…*clearly shows the effect of the Labrador Sea Water variability and water mass reorganization*…', recognizing that the physical processes are not well isolated in the eMLR model affecting to the Cant change estimates.



Yellow line represents the horizontal integrated Cant (Best AIC) which is lower that the 'true' Cant values (black line) when the changes in natural CO2 is negative (green lines). This is very clear, as the authors commented in the manuscript, in the layer of LSW.

This issue is one of the main disadvantages of applying eMLR in the North Atlantic. Kieke et al (2007) showed the strong variability in the CFCs content in the Labrador Sea Water related to the NAO variability. Steinfeld et al. (2009) evaluated the changes in the Cant using CFC data and the thickness of the main water masses in the North Atlantic and showed a clear decrease in the Cant storage rate due to the volume reduction in the LSW classes between 1997 and 2003. Also, Perez et al. (2008) showed a decrease of the Cant storage rate related with the NAO variability and described a strong correlation between Cant and O_2 in the main water mass (Johnson et al. 2005). This relationship is contradictory to the assumptions needed to apply the eMLR technics. But this inconvenient is not only observed in relation to LSW, also Rodgers et al. (2009) showed that in the North Atlantic, the top 1000m vertical integration of O_2 and total CO_2 does not show any spatial co-variability which suggests that natural and anthropogenic CO_2 concentration should have a negative correlation.

All this evidences and those described in the manuscript indicate that the observed decadal variability of the physical drivers in the North Atlantic between 1995 and 2005, associated with the NAO, are driving changes in both the natural component of the carbon cycle and also the anthropogenic component, generating a cross correlation in both component in contradiction with the assumptions of the eMLR technique.

Kieke, D., Rhein, M., Stramma, L., Smethie, W. M., LeBel, D. A., and Zenk, W.: Changes in the CFC inventories and formation rates of Upper Labrador Sea Water, 1997–2001, J. Phys. Oceanogr., 36, 64–86, 2006.
Steinfeldt, R., Rhein, M., Bullister, J., and Tanhua, T.: Inventory changes in anthropogenic carbon from 1997–2003 in the Atlantic Ocean between 20S and 65N, Global Biogeochem. Cy., GB3010, doi:10.1029/2008GB003311, 2009.
Pérez FF, Vázquez-Rodríguez M, Louarn E, Padín XA, Mercier H, Ríos AF. Temporal variability of the anthropogenic CO2 storage in the Irminger Sea. Biogeosciences, 5: 1669-1679. 2008. 10.5194/bg-5-1669-2008
Johnson, G. C., J. L. Bullister, and N. Gruber (2005), Labrador Sea Water property variations in the northeastern Atlantic Ocean, Geophys. Res. Lett., 32, L07602, doi:10.1029/2005GL022404.

Rodgers, K. B., et al. (2009), Using altimetry to help explain patchy changes in hydrographic carbon measurements, J. Geophys. Res., 114, C09013, doi:10.1029/2008JC005183

2.- Seasonal variability
Why estimated Cant does vary seasonally while the 'true' signal is constant? Authors argued that it originates from the seasonally varying ability of linear models to fit the data. However, a negative bias is found again in the estimated Cant signal, mainly in late winter that could be related to the spatial cross correlation variabilities in the O_2, natural CO_2 and Cant, which seem to be lower in summer and autumn considering the lower AIC numbers. In fact authors noted '*since regression misfits are largest in the summer and early fall, addition of winter and spring data should result in an overall improvement of the fit quality,'* suggesting that better AIC should produce negative biases in the estimated Cant changes because part of the Cant signal is included in the nominal natural signal of the eMLR model.

3.- Vertical coordinates. Authors performed this statistical analysis in horizontal surfaces because of the design of the model outputs. However, they have suggested that the results would be better if the analysis were performed in isoneutral surfaces. It is certainly true that the analysis would be more precise because the variability along isopycnal is dramatically lower than use horizontal surfaces along the whole North Atlantic. In addition, most of the recent works using eMLR did perform the computations over isopycnal layers, so it seems rather evident that the objectives of the manuscript would be better addressed if the analysis were done over isopycnal surface rather than horizontal surfaces from the beginning.

Other specific comments

Authors also suggest the use of using quasi-conservative variables as independent ones in order to optimize the tracer orthogonality. However, the lack of orthogonality is more relevant between natural and anthropogenic CO_2 that produced a negative bias in the estimation of the anthropogenic signal. Following the idea of the authors about quasi-conservative variables, it would be more practical the use of C* or TrOCA parameters as dependent variables to determine the anthropogenic signal instead of using the observed contemporary total carbon to evaluate the decadal change. C* or TrOCA parameters are nearly independent of the natural carbon cycle, thus avoiding the cross correlation observed in the North Atlantic between Cant, natural CO2 and oxygen.