Answers to Reviewers' Questions

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January 19, 2014

Dear Editor:

Please find our answers to the questions by the two reviewers below. We sincerely thank you for acquiring these two excellent reviews. Dr. Peter Rayner is one of few experts and pioneers in using 13C for inverse modeling of the CO2 flux, and his comments are highly beneficial to our research. The other anonymous reviewer must also be a highly experienced isotopic researcher, and the comments from this reviewer are also insightful, constructive and helpful. We are greatly indebted to their valuable time and effort in critical reading of our manuscript. We learned a great deal from these experts.

Sincerely,

Jing Chen, Gang Mo and Feng Deng

Interactive comment on "Atmospheric inversion of the surface CO₂ flux with 13CO₂ constraint" by J.

M. Chen et al.

P. Rayner (Referee) prayner@unimelb.edu.au Received and published: 25 November 2013

1 General Comments

this paper presents a new attempt to use a combination of _13CO2 and CO2 measurements in an inverse determination of surface fluxes over land and ocean. In some senses it is more ambitious than previous attempts such as (; ;) in its rather explicit use of a terrestrial model to calculate some of the terms in the 13CO2 budget. It also makes use of a larger set of the GlobalView data product () and especially its _13CO2 counterpart.

In other respects it is a rather conservative advance. It does not use particularly high source resolution nor does it use the much richer datasets of continuous measurements now available. The paper is generally well presented. The methodology, however, is sufficiently unclear that my commentary may be based on a complete misunderstanding of what the authors did.

Answer: We agree that some key steps in our inversion setup are not explained clearly, and we greatly appreciate your effort in trying to understand what we did. We here respond with great admiration for your ability to diagnose the likely shortcoming of our inverse modeling. Your diagnosis might have pinpointed the problem of the low sensitivity to 13C treatment (see more explanation below).

The results of the paper seem striking. Unlike most previous studies, the mean fluxes in the paper seem insensitive to details of the modelling of _13CO2. This is in strong contrast to the results of (;) who needed to include the product of disequilibrium and gross flux (sometimes called the isoflux) in their inversion explicitly. I have struggled to understand the implications of the treatment of _13C in this paper. I'm going to try to summarize what I think the authors have done and comment on it. This will give the authors a chance to correct any misunderstandings but also perhaps give guidance to ways they can improve their explanation. I'm also not sure whether the equation numbering in the text and on the equations is consistent. I think the equations that describe the effective discrimination are in fact (6) and (7) but are referred to as (7) and (8). Eqs. (6) and (7) define an effective discrimination of the net flux. This effective discrimination multiplies the CO2 flux to produce a 13CO2 flux. That suggests that if there were no CO2 flux there would be no 13CO2 flux. This is unphysical, the isoflux is the second largest term in the global atmospheric 13CO2 budget ().

Answer: Sorry, the original Eq. 6 label got lost during the typesetting so every equation is moved up by one after Eq. 6. The 13C fluxes for the ocean and land are calculated with Eq. 6 and Eq. 7 (as in the typeset copy). The first term of these two equations describes the 13C flux due to discrimination, and the second term due to disequilibrium. For the first term, it is indeed the case that the 13CO2 flux is zero when the net CO2 flux is zero. This is because if there is no CO2 flux, there is no discrimination. However, even if no discrimination flux occurs when the net CO2 flux is zero, the disequilibrium flux is still present as this term is multiplied by the one-way flux (respiratory flux for land and ocean-to-atmosphere flux for ocean). The second term should ease the concern of the equations being "unphysical". (It may be easier to read this equations by multiplying the net CO2 flux on both sides of the equation.)

This apparent problem is resolved in Eq. (8) which shows that the _13CO2 values that enter the inversion are observations treated by "presubtracting" the contributions of fossil fuel, the ocean flux, terrestrial flux and biomass burning. Since the ocean and terrestrial terms are taken from BEPS and OPA-PISCES-T the effects of the isoflux term are implicitly included in the inversion, at least from the prior estimates. I presume the CO2 observations are treated the same way although I didn't notice this mentioned in the text. A corollary of this presubtraction treatment is that the prior values of the unknowns f are zero, again I did not see this mentioned. If I have understood up to do not have a general result. If I am misunderstanding they need to explain their setup more carefully.

Answer: You pinpointed to a potential problem in our inversion setup, and you are right that in our inversion, different treatments to the spatial distributions of 13C discrimination and disequilibrium only modified the isotopic ratio while the presubtracted 13C concentrations were not affected by these treatments. This could have caused the low sensitivity to isotopic treatments in our inversion. In this way, we missed an important effect of these treatments on the inversion outcome. We felt greatly indebted to your insightful judgment. We have now begun to rerun our inversion to see the extent of the impact of this problem, and we have obtained a preliminary set of inversion results showing much greater responses to the different treatments. We expect that we will be able to produce a greatly improved set of results in our revised manuscript if we are given a chance to submit a revision.

2 Minor Points

Most of the following are suggestions for extra information needed in the paper.

• Although the paper uses a great deal of data, relatively little of it comes directly from measurements. It would be good to quote how many of the monthly values come from measurements made during that month.

Answer: The direct measurements of CO_2 and ${}^{13}CO_2$ from the GlobalView dataset are used in our inversion. There are 5431 monthly data from 209 sites for 42 months used for CO_2 (5431 out of 8778, i.e. 209×42), and 3066 monthly data from 73 sites for ${}^{13}CO_2$ (i.e. 100% of 73×42 monthly data). Since fewer sites have observations for both CO_2 and ${}^{13}CO_2$, all monthly data at 73 sites are used for ${}^{13}CO_2$. The

missed ¹³CO₂ data are filled with corrected values using the reference data provided in the same GlobalView dataset.

• The _2 test is a good start but it is also interesting to ask whether the _13C measurements are being matched better or worse than the CO2. the algorithm of () as used by () cann provide this.

Answer: We will consider these suggestions in our revision.

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Reviewer 2

This manuscript describes a new framework for joint $d^{1}C / CO_{2}$ inversions, and its application to a recent time period. Novel about this work is the use of a new biogeochemical model (BEPS) to simulate the 13C cycling through the terrestrial biosphere, which is a valuable tool as few of such models currently exist. The use of this model in an inverse framework follows methodology introduced before, but comes to conclusions that are not in line with our knowledge of the current 13C budget. This is very likely due to a number of flaws in the design of the inverse modeling problem, that need to be resolved before the paper can be judged on scientific merits. Since the results are likely to substantially change the conclusions, I can not recommend publication of the current manuscript in ACP and I think that a new manuscript should be created based on all suggestions and new results. I will write detailed comments on the methodology and conclusions below, to guide the authors in re-investigating their approach. I hope they will consider improving their framework, as I think it is important for more modelers, and more different models, to contribute to the effort of interpreting the d13C records.

Detailed comments:

- Please pay close attention to the equation numbering. The matrix formulation is not numbered, but is referred to as eq. 6 (e.g. in 2.1.3., line 18). From there on all other equations are wrongly referred to as 7,8,9,10, etc, but are numbered as 6,7,8,9, etc.

Answer: We apologize for the confusion around the equation numbers. The numbering sequence got shifted during typesetting, and we did not notice that.

- I have strong doubts whether Eq. 6 & 7 are correct. First of all, the units on the left hand side are not consistent with the right hand side. Rj is given as an absolute ratio (13C/12C), whereas Ra, alpha, Rbe

and others are given in permil relative to PDB. These formulations are indeed the same as presented in Ciais et al. (1995), but in their study R is portrait as an absolute ratios 13/12C, and not in permil relative to PDB as done in this manuscript. Alpha should be portrait as the ratio of two carbon ratios in different reservoirs (e.g. alpha_ph=Rb/Ra). For example, the term alpha_ph*Ra should represent the ratio in newly assimilated plant material (Rb). But in this manuscript (at least according to the text), the term is solved as the product of two permil values. Also, I cannot follow the way alpha_ph (photosynthetic fractionation) is calculated from the atmospheric signature (now called d_a and not Ra?!) and plant discrimination (Delta). I would expect the plant discrimination signature being related to alpha_ph through the formulation - Delta/1000+1. I am wondering whether these issues explain some of the unexpected results in this manuscript. I ask the authors to elaborate on this.

Answer: We believe that we implemented Eqs. 6 and 7 correctly. On the both sides of these equations, the unit is the absolute ratio. In the text, we incorrectly described Ra in the unit of permil, but in fact in our calculation, all ratios in these equations were converted to the absolute ratios. We will modify the description in our revision. The parameter alpha_ph for photosynthetic discrimination is already converted to the fractional value with for formulation you suggested. We will make this clear in our revision.

- In those same equations, why are the authors treating net exchange and disequilibrium in one parameter, and not simply as two separate 13CO2 flux terms: one for net exchange, and one for disequilibrium isoflux? If the net exchange flux approaches zero the 1st term will simply disappear (or become very small), but disequilibrium isoflux always remains present. You avoid all problems with dividing through zero NEE, and also make sure that moles of ¹³CO₂ are exchanged even when photosynthesis and respiration balance.

Answer: We are puzzled by the question and might have missed something important. In our view, the Equations 6 and 7 (as in the typesetting) have already separated the 13C fluxes into two terms as suggested by you: one for discrimination and one for disequilibrium, but both sides of the equations are divided by the net flux to become flux ratios. When the net CO2 flux is zero the discrimination flux is zero (the first term becomes the ratio alpha_ph× Ra), but the second term would still be larger than zero because it is multiplied by the one-way flux. As the second term is divided by the net CO2 flux according to the definition, the second term can be very large when the net CO2 flux is approaching zero. We had to limit the magnitude of this term to avoid the abnormally large values in our previous inversion when the ratio was calculated at 1 degree resolution (same as the prior flux) and then the ratio was averaged for a region. We have just found that we can avoid this issue without limiting the range when the flux ratio is calculated using the average fluxes for a region.

- The different case studies investigate the sensitivity of the surface CO2 fluxes to changes in discrimination and/or disequilibrium. If I understood correctly, the authors only make their changes in Eqs. 6 & 7 but not in the presubtracted concentrations. I think this is a wrong assumption, and the residual concentrations used in the inversion will become much larger when for instance the disequilibria fluxes are assumed zero. This must change the flux partitioning quite a lot to still satisfy the carbon balance, as an over- or underestimation of the disequilibrium isoflux by 30% can change the uptake over land and oceans easily with 1-2 PgC/yr (see Alden et al., 2010). So neglecting this disequilibrium flux, either over land or oceans, would cause much larger changes in the fluxes than magnitudes of 0.01-0.02 PgC/yr presented in this manuscript.

Answer: Thank you for pointing out this critical issue with our model setup, and your suggestion affirms that of Dr. Peter Rayner that has prompted us to make changes to the presubtracted concentrations. Preliminary results indicate much larger changes and we are in a process to analyze the results and produce new figures for the revised manuscript.

- Can the authors elaborate on how large the uncertainty is in the R matrix in terms of permil? When I calculate the uncertainties myself from your information the values become very large. For example, a

land station is assigned a standard deviation of 1.25 ppm for CO2. The authors use the relationship Ra*sigma to calculate the uncertainty in 13CO2 in units of ppm. So for a land station I would write: sigma*Ra=1.25*0.011=0.014 ppm. In ppm space, the mean and uncertainty in observed 13CO2 would thus become for instance 4.236 ± 0.014 ppm. In permil space this translates into $-8\pm$ 3.3 permil. I think an uncertainty of \pm 3.3 permil is quite a lot, especially when you consider that measurement precision is ~0.02 permil, and seasonal variability is within 1 permil. This could also be an explanation of why adding ${}^{13}CO_2$ to your inversion only had a minor impact on estimated fluxes.

Answer: We are sorry that we did not describe the calculation of R matrix in our manuscript. The uncertainty in observed ¹³CO₂ is not calculated by the uncertainty in CO₂ *0.011, as mentioned in your question. Our R matrix is calculated with a method based on the site category similar to Peters et al. [2005] and Baker et al. [2006]. By this method, R is defined for month i by $R_i = \sigma_{con}^2 + SD^2$, where σ_{con} is the systematic error for all the data at each site, assigned according to the type of observation sites. For CO₂ sites, the categories (respective σ_{con}) are: Antarctic sites (0.15), oceanic sites (0.30), land and tower sites (1.25), mountain sites (0.90), and aircraft samples (0.75). For ¹³CO₂ sites, 0.011(average ratio of ¹³CO₂/CO₂) of the values for CO₂ is used. SD is the standard deviation of the residual distribution computed monthly from the average monthly variability (var) files of GlobalView dataset. For CO₂ sites, SD is given in ppm. For ¹³CO₂ sites. The average R_i is 4.4ppm for CO₂ sites, 0.0021ppm(~0.5 permil) for ¹³CO₂ sites. If the R matrix could be one of the reasons for the small impact on the inverted CO2 flux by adding ¹³CO₂ to our inversion, we would experiment with other methods for calculating the R matrix.

- In Eqs 8, 9, and 10 I do not see the logic of the calculations, and especially this "VAR" correction term is of unclear origin. What is represented by this term, and why is it needed? Please correct me if I am wrong, but it seems like this has something to do with the smooth-curve nature of GlobalView, forcing you to use some time averaged CO₂ value to back out the 13C/12C ratio from the reported smooth d13C values? If so, this seems very dangerous to me as small variations in CO₂ would drive large changes in 13C/12C which were not actually 'observed', but were part of the separate curve fitting procedures for d13C and for CO₂. And this would mean the 13CO2 signals going into the inversion were not real. Could you please confirm that you have used actual observed d13C and CO₂ pairs from the flasks to back out the 13CO2? If not, can you please explain why your method is valid, and if I even understood it correctly? *Answer: The temporal variation of the observation of 13C available from GlobaView is caused by both 13C sources and sinks but also CO2 variation itself. This is precisely what is described in Eq. 8. The 13C partial derivative with time is used in the inversion, and C-var is the 13C partial derivative with CO2. In our early inversion without considering the effect of CO2 concentration change on 13C concentration (through C_var), the inversion results were quite unreasonable. Thanks for pointing this out, we will elaborate more in the text.*

- The method to simulate the disequilibrium of the land biosphere (Eq 15) is in my view incorrect. The equation states that carbon coming out of todays carbon pools carries the signature of tau years ago, with tau being the pool turnover time. This seems conceptually appealing, but the turnover time tau is not a "lag time" for signatures to return, but rather an integration time of the pools. In other words, carbon coming out of wood pools with a 30-year turnover time have accumulated 30 years of atmospheric signatures, assimilated through NPP, into the wood that is respired. The respiration signature is thus some weighted average of the atmosphere, NPP, and discrimination over each of the past 30 years. That is why one preferably should use a biosphere model to spin-up these pools consistently over time. With the current 'instantaneous' treatment, the isodisequilibrium forcing coefficient (see Scholze et al., 2003, Alden et al., 2010, van der Velde et al., 2013) would likely be too large, and especially too variable from year-to-year. A tabulated comparison of this value to previous work (including a discussion of C3/C4 land-use and NPP fractions, and of global average d13C isofluxes) would be helpful.

Answer: We agree that the turnover time of a carbon pool is not the same as the lag time for the purpose of tracing back the atmospheric signature. However, a pool with a 30-year turnover time does not only

release carbon accumulated over the last 30 years but also that older than 30 years. In other words, the turnover time would approximately equal to the lag time, as carbon older and younger than the turnover time would participate in the total carbon release causing the lag. This difference in the turnover and lad times exists because the variation of atmospheric 13C is not linear, and the proper way would be to do a temporal integration. Thanks for raising this question, although this problem was not raised before in the literature to our knowledge. We will evaluate the difference this may cause on the disequilibrium flux.

- Overall, it would help the readers if the authors can assess the performance of their a-priori modeling system better before being shown the inverse results. For instance by showing observations vs. model results (including uncertainty) from forward modeling of the a-priori fluxes over a few years. One can then quickly see if the simulated ${}^{12}CO_2$ disequilibria match the observed trend of d13C which is most strongly pulled down by fossil fuels. In a forward simulation, cases IV through VI where disequilibria were neglected, should give a rather big offset between observed and modeled 13CO2 mole fractions/signatures. Then in the inverse simulations, readers must be able to see the improvements to the simulated mole fractions/signatures, and have access to some statistics (chi-squared) to judge the chosen uncertainties versus model perfromance.

Answer: We have previously conducted validations of 13C concentration modeling results using observations and have confidence on our flux modeling results. It is difficult to validate the disequilibrium flux though, because the difference in the isotopic signature between photosynthesis and respiration is small, and the seasonal difference is confounded by other factors, such as water stress and stomatal conductance. In analyzing out new inversion results for the different cases, we will also do significance tests as suggested.

- Related to the a-priori mole fractions is the global CO₂ and 13CO2 flux budget. Considering the separate use of GFED2 fires and BEPS biospheric fluxes, I wonder to what extent the global CO₂ balance is closed a-priori and a-posteriori? And is the global 13CO2 balance closed if you add up the global mean values of each of the bottom-up flux terms? The disequilibrium flux (combined ocean + land) is the largest term in the budget after fossil fuels. So, either an over- or underestimation of the disequilibrium isoflux can deteriorate the carbon uptake patterns quite a lot over land and oceans. A closed budget from bottom-up estimates is difficult to create though and often requires additional scaling due to large model uncertainties in respiration, pool turnover and ocean exchange (Alden et al. 2010, van der Velde et al., 2013). The latter authors suggest that matching the observed ¹³CO₂ variability is even harder because models tend to underestimate variations in discrimination. Do you also see this?

Answer: We appreciate this insightful question. We have not examined our posterior CO2 concentrations closely to address this question, but this is definitely something very worth doing. One thing we have gradually come to realize is that our CO2 and 13C combined inversion is not as sensitive to disequilibra as the double deconvolution because the inversion is dominated by CO2 measurements and CO2 and 13C sites are mostly collocated.

- Table 3 is very hard to read, and must be presented differently. Overall, I also would like to see fewer global maps in the figures, and more relevant statistics or temporal behavior of fluxes. Also, the work would profit from considering a longer time period since much information is contained in the year-to-year variations of d13C. What is preventing you from doing this for a full decade?

Answer: Long-term inversion is technically feasible but would involve much more work in terms of global GPP and 13C flux modeling (including LAI and meteorological data preparation) and transport modeling. However, for the objectives of this research for evaluating the 13C information content for use in CO2 flux inversion, we believe the length of time for the inversion is generally sufficient. Our work, albeit not completed yet, has addressed several interesting issues. We will follow your suggestion, trying to present the tables more informatively.