Geosci. Model Dev. Discuss., 7, C815–C827, 2014
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Interactive comment on "A multiresolution spatial parameterization for the estimation of fossil-fuel carbon dioxide emissions via atmospheric inversions" *by* J. Ray et al.

J. Ray et al.

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Received and published: 7 June 2014

At the very outset of the response, we would like to clarify that we view the work described in the manuscript as a methodological first step in the development of an inversion scheme that could be potentially used to estimate ffCO2 emissions. In this paper, we have concentrated on describing a model for very spatially heterogeneous ffCO2 emissions fields and how this model could be used in an inverse problem (it requires a class of optimization methods – sparse reconstruction – that is not widely used in CO2 inversion studies). The inverse problem adopted a number of simplifications e.g. no boundary fluxes into regional domain, very small model-data mismatch errors, an

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Interactive Discussion

Discussion Paper

assumption that the ffCO2 concentration signal at a sensor could be isolated from the biospheric signal – and used a sensor network that is not specifically designed for targeting ffCO2 emissions. Consequently, we do not imply that our method could be quickly adapted for real-data inversions using current transport models, sensor networks and ffCO2 concentration measurement techniques.

It would be reasonable to believe that accurate ffCO2 estimation would require a new sensor network, placed closer to major sources of ffCO2 emissions. Further, while radiocarbon is one way of estimating ffCO2 concentrations at sensors, one could also consider pollutants from incomplete combustion e.g., CO, to "back-out" the ffCO2 signal. However, these topics, though integral to the question of ffCO2 emission estimation, are out of scope in a paper that, as the title suggests, is about a spatial parameterization for ffCO2 emission fields.

Many of the issues required to adapt our method to real-data inversions and/or regional inversions e.g., specification of boundary fluxes, determining their uncertainties and assessing their impact on ffCO2 emission estimates, are identical to those faced by inversion studies for biospheric CO2 fluxes. These issues have been investigated and addressed in the biospheric CO2 context (Gourdji et al, 2012), and may provide starting points for adapting our method (for ffCO2) to a real-world scenario. In addition, the complications introduced by transport (dispersion and dilution) and the impact of transport model errors are identical to those faced by biospheric CO2 inversion studies and may be solved in a similar manner – the papers by Chatterjee et al, (2012) and Gourdji et al, (2012), cited in our manuscript, discuss these issues to a greater extent. Consequently, we consider these topics outside the scope of this paper.

We will add this clarification in the Introduction section.

The reviewer states: "The mathematical framework is rather complicated but is carefully described and can be understood with reasonable effort. However, the numerical tests presented are not realistic, and it is unclear from the present manuscript how

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Interactive Discussion

Discussion Paper

this tool would be applied in practice. Ideally this should be addressed by applying the model to a more realistic case study as described below. If that is not possible, then the limitations of the numerical tests described in section 5 should be clearly described. The introduction or discussion should include a more detailed description of questions/applications for which this framework is appropriate. One such application could be an Observing System Simulation Experiment to evaluate the utility of a dense radiocarbon sampling network or the utility of continuous radiocarbon measurements that have to date been demonstrated with sufficient accuracy only in the laboratory."

Response: We have added a "Discussion" section where we elaborate on what the spatial parameterization could be used for (primarily for Observation System Simulation Experiments, determining the location of sensors, and the frequency with which measurements are obtained) the limitations of the tests performed in Sec. 5, as well as the impact of various numerical and boundary condition approximations on the emission estimates.

The reviewer states: "Model code is provided in Matlab via a website. I'm sorry that I did not have a chance to download this, but I wonder whether it includes the early steps in the wavelet analysis (i.e., corresponding to eqn (3))."

Response: We will include it.

Major concerns

The reviewer states: "The proxy datasets may be strongly spatially correlated with energy use, but not necessarily with fossil-fuel emissions. Many large power plants in the US are located far from the urban areas they serve. For example, large power plants in Wyoming and Ohio serve customers in distant urban areas. In the case of remotely located large power plants, the nightlight and built-up-area index would be unlikely to have intensity proportional to the emissions. In the US and certain other *nations, detailed emissions data are available for large point sources. How could the framework be modified to take advantage of such information? E.g., could these point source emissions be subtracted prior to the inversion)? Is there another proxy dataset that could provide information about large point sources (e.g., perhaps high-resolution thermal imagery?) For areas where reliable emissions point source data are not available, might large point sources complicate or confound the analysis? Please address this in the introduction and/or discussion."*

Response: The reviewer is correct in saying that the use of nightlights and built-up area maps, which correlate with energy use, could lead to an inaccurate random field model. Specifically, we may omit a fine-scale wavelet that corresponds to the large point source. However, the random field model is multi-resolution, implying that another wavelet, at a coarser level, whose support covers the point source, could model it. In doing so, the point source gets "smeared" over a larger area and the estimate of its magnitude may incur an error. However, it will not be totally omitted from the inversion procedure, with its emissions apportioned to other non-neighboring sources. This is a consequence of the multiresolution nature of our MsRF.

In case accurate databases of large point sources exist e.g., CARMA, the impact of the point sources can simply be subtracted out. If another proxy such as infrared images exists, the wavelets in our MsRF could be augmented with the wavelets (of the same family) chosen using the second proxy. If neither exists, the large point sources are smeared, as described above.

We realize that proxies are imperfect markers of ffCO2 emissions. One of the experiments in Section 3 specifically investigates the modeling ramifications of using imperfect proxies to construct the spatial parameterization. Experiments in Section 5 investigate the usefulness of the final model.

We will add this to the "Discussion" section.

The reviewer states: "Fossil fuel CO2 cannot be directly measured. This is acknowl-

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Interactive Discussion

Discussion Paper

edged in the manuscript but treated rather blithely. Radiocarbon measurements provide the most direct measurement-based constraint available for separating biological and fossil fuel CO2. Radiocarbon is a powerful tracer, but unfortunately measurements are expensive and are being made on discrete samples at a subset of the 35 tower measurement sites considered here at a rate of 3 midday samples per week. The errors on fossil fuel CO2 estimated from radiocarbon are ∼ 1ppm (J.B. Miller et al., JGR, 2012). The sampling frequency is overestimated by an order of magnitude and the measurement uncertainty is grossly underestimated by the numerical tests considered here. A technique for continuous measurement of radiocarbon has been demonstrated in the laboratory (D. Murnick, O. Dogru, and E. Ilkmen, 14C Analysis via Intracavity Optogalvanic Spec- troscopy, Nucl. Instrum. Methods Phys. Res B., 2010 April 1; 268(7-8): 708–711. doi:10.1016/j.nimb.2009.10.010.), but field deployment of continuous radiocarbon sensors has not been demonstrated. Operational autonomous field operation will not be plausible for many years. I am curious whether a more realistic numerical test representative of currently available or plausibly augmented radiocarbon data (e.g. 10 - 35 towers, 3-7 mid-afternoon samples per week, 1 ppm measurement errors) would provide a useful constraint if aggregated over a long time period, e.g. 1 year, and limited to the region where the footprints show sensitivity."

Response: The primary difficulty in performing a realistic inversion (1 ppmv noise) is the placement of the measurement towers – they are far from sources of ffCO2 emissions, leading to a ffCO2 concentration signal that is usually no more 2 ppmv on any sensor. Adding a noise with an error variance of 1 ppmv makes them unusable. A true test of our method, under realistic conditions, would also require a sensor network designed and sited to measure ffCO2 emissions. Consequently, in this paper, we have chosen an idealized case and focused on developing the inversion methodology.

The reviewer states: "In any regional inversion, boundary values need to be estimated and may have large uncertainty. Gourdji et al., (2012) showed that boundary/initial condition errors are potentially large enough to preclude reliable quantification of the

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Interactive Discussion

Discussion Paper

net annual ecosystem uptake of CO2 for North America. It is important to consider and discuss the potential complications of assigning fossil fuel CO2 boundary values for the region where fluxes are being estimated, i.e. here the boundaries of CONUS. This seems especially complicated here, given that the impact of emissions from areas outside CONUS but within the rectangular domain would need to be taken into account. The compressive scaling strategy to exclude emissions outside of CONUS as described in the paper is appropriate for the idealistic case considered (synthetic obs), but in a real-data study either (1) accurate 4-dimensional fossil fuel CO2 mole fraction values would be needed along the boundaries of the emission estimation domain (2) accurate 4-dimensional information about fossil fuel CO2 mole fractions along the boundaries of the continent along with a correction for emissions within the rectangular domain but outside the emission estimation area. Other complications arise if a significant number of LPDM particles fail to exit the domain."

Response: The reviewer is correct in stating that our method for regional inversions will suffer from the same boundary condition issues that other regional (biospheric CO2) inversion methods do. This is unavoidable in the absence of good boundary condition data. The choice of option (1) versus (2) above would depend upon where ffCO2 concentrations (to serve as boundary fluxes) were available (around the CONUS boundary or around the continent, as in Gourdji et al, (2012)). In case the boundary fluxes were available at the CONUS boundary, we would use option (1). As in our paper, we would not estimate emissions outside CONUS and use compressive sensing to suppress estimates there. The impact of ffCO2 emissions from OCONUS on the measurements would be imposed by time-variant ffCO2 influx/efflux along the CONUS boundary. In principle this is no different than Gourdji et al, (2012). The question of LPDM particles failing to exist is identical to that faced (and addressed) in Gourdji et al, (2012). However, the issue of boundary fluxes and the impact of those uncertainties on the ffCO2 estimates are outside the scope of the paper.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

Specific concerns

Pg 1280:10 “I recommend expanding the discussion of the potential for using radiocarbon measurements as a (almost) direct tracer for fossil fuel CO2. Also note that accuracy of fossil fuel CO2 estimated from radiocarbon is $\sim$ 1 ppm, measurements are limited because of cost, lack of technology, etc.”

Response: Our manuscript describes a method to estimate ffCO2 emissions predicated on the availability of concentrations of ffCO2 measured at a set of sensors. Radiocarbon is one of the ways of obtaining that measurement, but it could potentially also be derived from joint measurements of pollutants such as CO. Issues related to the cost and feasibility of making radiocarbon measurements are outside the scope of the paper.

Pg 1280: “After discussing radiocarbon, add a short paragraph about the atmospheric transport model describing signatures of emissions are dispersed and diluted and possible errors in simulated transport.”

Response: The complications introduced by transport are no different from those encountered by inversion studies focused on biospheric CO2 fluxes, and these have been addressed in literature. We would consider them to be out of scope of our paper.

Pg 1283:8: “Can wavelets be scaled up as well as shrunk?”

Response: Yes they can. That is clear from the expression for the wavelet at scale s and translation j.

Pg 1283: 2nd to last line.” I don’t understand what is meant where Gothic W(s) then | Goth- icW(s)| (i.e., the |’s don’t appear in the equation, but do appear in the description).”

Response: Gothic W is the set of (I, j) indices of wavelets of scale s. The magnitude of the set (Gothic W within vertical bars) is the number of (I, j) pairs in the set. We will add this clarification in the manuscript.

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

Pg 1284: "Consider defining "random field" and briefly explaining why a random field is useful for representing complex emission maps"

Response: A random field model allows one to generate arbitrary fields based on the values assumed by the model's parameters. Certain characteristics required of the fields can be encoded into the random field model. For example, if the modeled fields are required to be smooth, one can impose a spatial correlation between field values at different locations, e.g. adopt a Gaussian random field model. The correlation function's parameters can be used to control the degree of smoothness. If the modeled fields are known to be rough at certain locations, they can be modeled using wavelets, with fine wavelets restricted to the rough regions and the wavelet weights acting as the model parameters. These parameters can assume arbitrary values i.e., they are random variables, and thus the model can create random fields.

We have added this to the beginning of Sec. 2.1.

Pg 1285: "Explicitly define || ||p notation here instead of or in addition to where it defined on pg 1288 ln 5."

Response: We will do so

Pg 1286:16: " Is there length scale associated with s=3 (i.e. in degrees lat/lon)? Struggling a bit to understand how wavelets manifest in physical space."

Response: Yes there is. The finest wavelets, on the M = 6 hierarchy, are on the 6th level, and have a support of 2 degree X 2 degree. s = 3 wavelets are 3 levels above, and are $2^3 \times 2^3$ larger i.e., 16 degree X 16 degree. We will add this example in the manuscript. Another reviewer also requested it.

Pg 1288:12: "Why does sentence 2 ("Thus, while we . . .") follow from sentence 1 ("Note that the sparse nature. . .")?"

Response: The two sentences were badly framed. What we meant was: "We will use wavelets selected using the (single) nightlight and BUA maps to estimate weekly ffCO2

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

emissions. Our tests above show that they model annually averaged Vulcan emissions adequately, and we assume that while the emissions wax and wane with time, their spatial distribution does not vary sufficiently to require a new wavelet selection. We base this assumption on ffCO2 emissions' correlation with human activities, and static sources like powerplants which do not display large spatial dislocations with time". We will revise the manuscript accordingly.

Pg 1290:13 and Fig. 15: "It looks like the magnitudes of the errors are similar to the magnitudes of the emissions themselves. It would be nice to include a plot of relative error would be interesting perhaps along with a scatter plot."

Response: The relative error plot is not very informative. Locations with large emissions, as predicted by f_v and f_{pr} can be slightly offset; further, since the emission fields are so rough, neighboring locations can have drastically smaller emissions. This leads to division by (almost) zero problems, leading to very large relative errors. These are rare, but they increase the dynamic range of the relative error plots. We will, however, include scatter plots of f_v and f_{pr} for each grid cell, plotted against each other, in the online supplementary material. We see that while there is a strong correlation between the two, they are far from being identical i.e., while the prior fluxes are a "guess" for the true emissions, they are not particularly close.

Pg 1291:2: "Briefly explain here or in the introduction why you are using these 35 tower locations that are ill-suited for fossil fuel estimation. It is sufficient to state that the footprints were available from earlier studies and that it was convenient to use them for method development."

Response: The towers chosen belong to the network that existed in North America in 2008, and therefore represents a realistic network, although far from optimal for the purpose of fossil fuel flux estimation.

Sec. 4.2: "It would be nice to include the figure from Ray (2013) showing a CDF to illustrate impact of non-negativity."

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Response: We will do so.

Pg 1292, 1st paragraph: "8 days seems a short timescale for estimating emissions. Was this selected to minimize aggregation error when computing monthly averages? Even annual estimates would be useful for some applications."

Response: The reviewer is correct. We will add the rationale behind the 8-day estimation period in the manuscript.

Pg 1295:18-22: "As already noted, a radiocarbon-based inversion seems a much more straightforward application for this framework than extending the wavelet approach for simultaneous estimation of bio and fossil fluxes. However, the measurement density is much larger than will be possible for radiocarbon anytime in the next two years. The measurement errors for fossil fuel CO2 are ∼1 ppm. Chatterjee et al. (2012) errors of 0.1 ppm seem optimistic even for total CO2 inversion unless model transport errors are somehow accounted for elsewhere (not an issue for synthetic data studies, but potentially important for real-data inversion). NOAA's CarbonTracker (www.carbontracker.gov) uses much larger sigma values for continental sites though they were assigned somewhat arbitrarily (Peters et al., PNAS, 2007). Also boundary value errors would be significant in any regional inversion."

Response: We agree with the reviewer that a radiocarbon-based inversion is a more straightforward application for our framework than joint biospheric-fossil-fuel CO2 inversion. However, the aim of the paper was to introduce a spatial parameterization, an accompanying sparse reconstruction method and provide evidence of their usefulness in an inversion. We have adopted a number of simplifications to do so, as the reviewer has pointed out. We used sensors from a network that existed in North America in 2008 and whose locations are ill-suited for ffCO2 emission estimation. A sensor network optimized for ffCO2 measurements does not currently exist. Consequently, we have focused on developing the methodology first, under idealized conditions. We introduce a number of methodological and modeling novelties - we could not find any use

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Discussion Paper

of wavelets, sparse reconstruction and non-negativity enforcement in the atmospheric inversion of rough (non-stationary) emission fields. We will add this rationale to the “Discussion” section in our revised manuscript.

Pg 1293:3 “How different is the value of c when Edgar is used instead of Vulcan? How different are the total emissions?”

Response: We have added this to the revised manuscript.

Pg 1296:6 “Please note also that 0.1 ppm is already very optimistic.”

Response: We agree. We will include our rationale in the “Discussion” section, as stated above.

Pg 1299:2-6 and Fig 9(a): “A relative error plots would be useful in addition to difference plot shown in Figure 9a.”

Response: The two estimates differ slightly in the sense that strong ffCO2 sources may be estimated at slightly different locations. Since the spatial distribution of ffCO2 fields is rough, neighboring locations may have very different, i.e., small, emission estimates, leading to large relative errors (division by almost zero). Consequently we have not found relative error plots to be useful – the range of relative errors is set by a few grid-cells where these shifts occur.

Sec. 5, other comments: “A figure showing one or more longitudinal transects of E, fpr, fv, and F (before non-negativity) would be interesting. Perhaps for a 32-day period. It would be nice to see the extent to which sharp spatial transitions are or are/not resolved for these quantities. If at all possible, it would be useful to include another more realistic case study with much sparser data (e.g. daily or thrice weekly samples) and with errors âĹij1 ppm, corresponding to current radiocarbon capabilities.”

Response: We fail to see what these transect would provide, since the fluxes are those before the imposition of non-negativity. As might be expected, the fluxes show positive and negative values. The negative fluxes and their frequency are small and are captured by the Cumulative Distribution Function that the reviewer has asked for. The spatial gradients can be seen in Fig. 6 (and more figures are available in the Technical Report cited in the paper).

Pg 1300:18-21: "This overstates what has been demonstrated in the current study."

Response: We will reword to better reflect our accomplishments.

Pg 1301:5 "Briefly describe what is meant by a dictionary or omit. The discussion of how the framework could be extended to account for biospheric fluxes is too brief to be of much use."

Response: We agree and will remove the discussion.

Fig. 6: "Please consider showing difference plots (from truth) and/or relative errors in addition to estimated emissions. Also, perhaps it would be more useful to show a 32-day average rather than an 8 day average."

Response: The relative error plot is not very informative for the reasons described above for the comments for Pg. 1290:13 and Pg 1299:2-6. We will add a difference plot (between true and estimated emissions) to the paper. We will update the text to point of the comparison with Vulcan emissions.

1280:5 1286:5 The reviewer found grammatical errors.

Response: Will be fixed in the manuscript

1289:15 "Why switch to delta from alpha used earlier?"

Response: alpha is normalized and is [0, 1]. Delta is not.

1301:11 Ray (2013) is a link to the first author's individual webpage at Sandia. It does not seem like a particularly robust long-term repository for the MATLAB code. Perhaps a static version could be included as a supplement to the paper. Ray et al. 2013 SAND Report reference: Is there a long-term repository for DOE technical reports that would *perhaps be a better long-term link*

Response: We will provide a link to the copy of the Sandia technical report stored in Sandia's technical library. The software supplied with this paper is a more difficult challenge. We require permission from the US Department of Energy to license and distribute any software. This may not arrive before the paper is finalized and we may not be able to supply a codebase in time.

Fig. 5, Fig 7 and Fig 9: The reviewer points out that certain markings, axes in figures, colors used for plotting etc. are not very legible, and ought to be magnified, truncated etc. for readability.

Response: We will make the changes in the manuscript.

*Fig. 7 RHS, legend notation seems inconsistent with caption (fk vs Ek?). I'm not convinced that incorporating yobs *clearly* improves the spatial agreement for the 8 day time periods, but agree that 32 day periods show substantial improvement.*

Response: We will correct the manuscript to reflect this.

References

[Gourdji et al, 2012] Gourdji, S. M., Mueller, K. L., Yadav, V., Huntzinger, D. N., Andrews, A. E., Trudeau, M., Petron, G., Nehrkorn, T., Eluszkiewicz, J., Henderson, J., Wen, D., Lin, J., Fischer, M., Sweeney, C., and Michalak, A. M.: North American CO2 exchange: inter-comparison of modeled estimates with results from a fine-scale atmospheric inversion, Biogeosciences, 9, 457–475, doi:10.5194/bg-9-457-2012, 2012.

Interactive comment on Geosci. Model Dev. Discuss., 7, 1277, 2014.