

We would like to thank the referee for the thoughtful and insightful comments. We have addressed all of the comments. Our responses are itemized below.

The authors use a surface observations from the IMPROVE network and the chemical transport model GEOS-Chem and its adjoint to estimate concentrations of BC in the atmosphere in the Western U.S. during July-September of 2006. The major finding is that the inversion system accurately predicts total emissions of BC, but cannot distinguish between different sources of BC. Adding additional ‘observations’ of biomass burning-derived BC leads to predictions of higher biomass burning and anthropogenic BC emissions. This result is not improved with additional surface observations of emissions.

I found the manuscript a little hard to read. The manuscript contains a lot of information (22 pages worth + figures). Improving tables 1 and 2, providing additional information about each ‘case’ in the figure headers would greatly improve clarity.

Point well taken. Additional information added as suggested in the 'specific comments'.

The manuscript should be shortened by streamlining the results and discussion section, and removing redundant findings/interpretation of findings. For example, the following statement is repeated multiples times: ‘This indicates that the inversion system lacks the ability to effectively distinguish collocated biomass burning and anthropogenic emissions in the WUS on model grid scales.’ I wonder if the manuscript could be restructured with a stronger emphasis on the results and then a list of reasons for the results rather than being taken step-by-step through a long list of modeling cases.

Point well taken. Revised as suggested in the 'specific comments'.

The manuscript can also be shortened considerably and made easier to follow by moving citations to the end of sentences, rather than starting a lot of sentences with ‘Example et al. (2006) showed ...’.

Agree. The sentences are revised as suggested (Sects. 1, 2, 3.3, 4.1.2, 4.1.3).

Please add a summarizing statement in the introduction and results/summary of how exactly this study differs from Mao et al. 2014. The information is in the text, but it is hard to find.

Added following summary, ‘Previously, linear analytical inversions were applied to optimize sources and source regions (the Rockies, California and the Southwest, and the Pacific Northwest) of BC in the western U.S. for May-October 2006 (Mao et al., 2014). The analytical inversions show factors of 3–5 increase of the biomass burning emissions and a ~50% reduction of the anthropogenic emissions (Mao et al., 2014).’

Title

The title does not convey the main message of the manuscript. I suggest changing the title to something like ‘large uncertainties remain in the magnitude and sources of black carbon emissions in the western United States’ or to make it more clear that ‘variational’ refers to a type of inversion modeling study.

Changed to ‘Estimates of Black Carbon Emissions in the Western United States Using the GEOS-Chem Adjoint model.’

Specific comment (incl. language)

Manuscript

Please use ‘variational’ or ‘adjoint’ throughout the manuscript; using both terms is confusing to readers not familiar with the inverse modeling methods

Now use ‘adjoint’ consistently in the manuscript.

Please use consistent spelling throughout the manuscript, including figures and tables for Western US or western US or WUS.

Now use ‘WUS’ consistently.

Remove spaces between numbers and % or degree signs.

Deleted.

Please spell out abbreviations, including BC if they occur at the beginning of sentences.

Revised (Sect. 1).

Please use italics for a priori and a posteriori – it would make it easier to identify the terms in the text.

Changed.

Introduction

P21867 L4 ‘[. . .] in the IPCC 2007’ should be ‘two times larger than previous estimates (IPCC, 2007).’ (please check the latest IPCC report for updates!)

Changed to ‘The radiative forcing due to BC is 0.64 W m^{-2} globally (IPCC, 2013)’

P21867 L5-7 Remove 'both' in 'also an important agent to both' and remove 'and cause global warming (IPCC, Bond)' – this is repeating the previous statement.

Revised to 'Black carbon is also known as an important agent to both degrade air quality and affect human health.'

P21867 L15 Change 'the severest' to 'the most severe' drought.

Revised.

P21867 L13-17 The sentences 'In the western US. . .' and 'Recently, CA is experiencing. . .' might wrongly suggest that the mountain snowpack in the Sierras is a dominant source of Colorado river water. Please rephrase.

Revised to 'In the recent decades, the western U.S. is experiencing the most severe drought (e.g., Melillo et al., 2014) and the water level of the Colorado River has been decreasing (e.g., Vano et al., 2013).'

P21867 L23-26 In 'Long-term records. . .' Increasing urbanization should be mentioned as another driver of fires in Southern California, not just climate change (which is also partially driven by the urban heat island effect), because most fires in CA are ignited by people.

'Increasing urbanization' now included.

P21868 L6-7 'Top-down inverse methods is' - missing 'The' or change to 'Top-down inverse modeling. . .' More importantly, is it correct to use the term 'top-down' here, since the model is constrained with surface observations (bottom-up)? Maybe it would be more appropriate to just use the term 'inverse'-modeling?

Changed to 'Inverse modeling'.

P21869 L6-14 Please shorten: Starting each sentence with the study author makes this list very wordy. I suggest listing the studies with the references in parentheses at the end of each type of study/model/region.

Revised.

P21869 L15 The term 'variational' is introduced for 'adjoint', please move this to P21867 L9 when inversions are explained.

Moved as suggested.

P21869 L20 'elevated mountainous' makes no sense. Are these sites elevated in anything but relief?

Deleted.

P21869 L17 please remove 'for clarity'

Deleted.

Methods

P21870 L2 How can the update of seasonality in the data be from 2003 (Park) if the data is from 2007 (Bond)? Is Bond 2007 using the wrong seasonality?

Seasonality is not included in Bond et al. (2007). Clarified to "Global annual anthropogenic emissions of BC are from Bond et al. (2007). Seasonal variations of anthropogenic emissions are considered over the U.S. following Park et al. (2003)."

P21870 L3 remove 'emissions' after (GFEDv2)

Deleted.

P21870 L12-13 I suggest removing this statement on the history of the adjoint.

Changed to "We use the GEOS-Chem adjoint (Henze et al., 2007, 2009) to estimate BC emissions in the WUS."

P21870 L26 Why is delta sigma=0.1 used?

Revised as "We use here $\delta\sigma = 0.1$, following Henze et al. (2007)."

P21871 L2 Text uses 'r', but Figure 2 uses 'R' to describe correlations. Please use the same terminology in all instances.

Revised. Now we use 'r' consistently.

P21871 L10 should read X [...] ARE the emissions (not IS)

Revised.

P21872 L16-18 I suggest shortening to: We set the observation error at 30, 50, or 100%, which includes the model, representation, and measurement error.

Revised.

P21873 L1-2 It is not clear from this statement whether scaling emission factors or emissions is the standard practice. Please rephrase.

Revised to 'We optimize here the scaling factors of emissions \mathbf{X}/\mathbf{X}_a (rather than the actual emissions \mathbf{X}), as a standard practice in inversion studies (Henze et al., 2009).'

P21873 L14-18 This can be shortened, to remove redundant information, to 'Here we calculate cost function gradients with a hybrid form of scaling factors (Jiang et al., 2014b) so that the resulting optimization converges equally efficiently for the regions with positive or negative biases.'

Revised as suggested.

Results and Discussion

Typo in title: discussion, not discussions

Revised.

P21874 L22-25 Please provide the a priori emission estimates for July-September for a direct comparison. Unless the reader refers back to the Mao et al. 2011 and 2014 papers, it is impossible to judge how much higher the emissions are a posteriori.

Added. Changed to 'The *a posteriori* emissions are 49.9 Gg at $2^\circ \times 2.5^\circ$ and 47.3 Gg at $0.5^\circ \times 0.667^\circ$ for July–September, substantially higher than the *a priori* (24.3 Gg).'

P21875 L14-15 Please rephrase - I don't understand this sentence/paragraph. Does this mean that positive sensitivities are observed in regions in which a reduction of BC emissions would improve the agreement between the modeling results and the observations? = The model overestimates actual emissions in regions with positive sensitivities? Does this then mean that bb-emissions are underestimated in WA, OH, ID and CA?

Revised to 'negative sensitivities are regions in which the model underestimates actual emissions and an increase of BC emissions would improve model agreement with the observations. The

largest negative sensitivities to biomass burning emissions are in Washington, Ohio, Idaho, and California, where the model severely underestimates the biomass burning emissions and the sensitivities decrease significantly after the inversions.’

P21875 L19-22 I suggest shorting to: ‘For carbon dioxide, a minimum of 10 sites was needed . . . (Gloor et al. 1999). For BC, the number of site is usually smaller.’

Revised.

P21880 L12-13 This statement makes no sense: ‘Pseudos 3–5 are the same as Pseudo 1, but with several differences.’ They are either the same or not.

Revised as ‘Other aspects of Pseudos 3–5 remain the same as those of Pseudo 1’.

P21880 L13-23 This section describes how many observations are needed for successful inversion modeling. The same topic is explored on P21876 L 18-25 to 21877 L1-15 Please provide more explanation of how these two analyses relate / differ from each other.

Added discussions in Sect. 4.2, ‘As we discussed in Sect. 4.1.2, the differences between the inversion results with 69 or 56 IMPROVE sites are essentially small, indicating that the 69 or 56 sites alone (absent other observations) are likely sufficient only for constraining the total emissions of BC, especially at $2 \times 2.5^\circ$ ’.

P21882 L2-8 Please rephrase, I cannot follow and the statements appear to contradict each other. ‘In California, for example, the a posteriori biomass burning emissions at 0.5×0.667 increase in the adjoint inversion but decrease in the analytical inversion. The analytical inversions show factors of 3–5 increase of the biomass burning emissions and 5 a \hat{L}_{ij} 50 % reduction of the anthropogenic emissions (Mao et al., 2014). In contrast, both the biomass burning and anthropogenic emissions in the adjoint inversions increase by two folds (Table 1). The total a posteriori emissions are rather comparable (within 20–50 %) between the two inversions.’

Revised to ‘In California, for example, the *a posteriori* biomass burning emissions at $0.5 \times 0.667^\circ$ increase in the adjoint inversion but decrease in the analytical inversion, relative to the *a priori*. In the WUS, the analytical inversions show factors of 3–5 increase of the biomass burning emissions and a ~50% reduction of the anthropogenic emissions, relative to the corresponding *a priori* (Mao et al., 2014). In contrast, both the biomass burning and anthropogenic emissions in the adjoint inversions increase by two folds (Table 1). The total *a posteriori* emissions are rather comparable (within 20–50%) between the two inversions.

21882 L23-26 This section is a word-by-word repetition of P21873 L20-23. Please streamline. ‘The assumption that a priori errors are spatially uncorrelated hinges on the consideration that

the spatial 25 resolution of the CTM is much larger than the correlation length scale of the individual emission sources (Henze et al., 2009).'

Deleted.

21883 L7 remove 2nd 'by' in by 39 and by 29%.

Deleted.

21883 L10-15 shorten paragraph

Revised as, 'For example, in Montana, Idaho, and Wyoming, the *a posteriori* anthropogenic and biomass burning emissions increase by factors of 2.2 and of 2.7. In Utah, Colorado, Arizona, and New Mexico, the corresponding emissions increase by factors of 1.8 and of 1.3. In California and Nevada, the emissions increase both by a factor of 1.8.'

Summary and Conclusions

It is not necessary to provide another summary. The main questions, activities and findings should be summarized ONLY in the abstract. Please shorten this section to less than 1/3 page of text, focusing on what next steps the work implies.

Revised.

Tables

Table 1 This table is very hard to read, because there are 12 comments underneath the table, and e.g. 'e' refers to Case 1. From looking at a 'Case x' the reader cannot intuitively understand how the difference in the parameters affects the results. It would be easier to understand the different set-ups if the authors added columns for the parameters that are changing: (1) the size of inversion (0.5x0.667 or 2x2.5 degree), (2) the number of IMPROVE sites used, (3) the uncertainty estimate for biomass burning, (4) the uncertainty estimate for anthropogenic emissions, (4) the observational error, and (5) the a priori biomass burning emissions. I also suggest to moving the 'case' column more to the left, next to 'adjoint'. In the adjoint, the uncertainty for biomass burning emissions is set to 500% (Case 1) and then 300% (Case 2), and then the uncertainty for anthropogenic emissions is 30% (Case 3) and then 200% (Case 4). I propose to change the order, so that the uncertainty either increases or declines for each parameter. To the header, please add an explanation for 'giga' (Gg (109 g)).

Point well taken. Revised as suggested.

Table 2 As for Table 1, it is hard to read this table because there is so much information in the comments under the table. It would help if any of the info could be transferred to the table. 'A

posteriori is kind of suspended between columns. Does this refer to inversions and 'ghost' emissions? The label should be moved into the same column as 'a priori'. It is not clear from the header or comments what Delta Emissions and J(X) reduction are.

Revised as suggested.

Figures

This paper has a lot of figures (15). I strongly encourage the authors to consider moving some of the figures to an online supplementing material. Which figures are critical to understanding the main findings? Suggestions for moving to the appendix: Figure 2, Figure 3 & 4 (merged into a two panel figure)

Agree. Revised as suggested.

Figure 1 '.' missing after US ('... western US Also...') Please explain why solid circles differ in diameter.

We add explanation 'We use all the 69 sites in the standard inversion Case 1 (Table 1). Small 56 solid circles represent the sites used in the inversion Case 6 (Table 1)'.

Figure 2 What are the units of finite difference sensitivities; please add this information to the y-axis labels.

Added.

Figure 4 Text in caption does not match y-axis label. Please correct.

Revised.

Figure 5, 6, 7, 10

Figures 5-7: I find it misleading that the same color is used to depict different magnitudes of change, e.g. for Fig. 5 in biomass burning vs. anthropogenic and total emissions. Figure 5: Scale bars under the figure panels are bleeding into each other and some numbers/letters are cut in half. I suggest using only one scale bar for each column, shown at the bottom (or different colors for different magnitudes).

Point well taken. Figures are revised using only one scale bar for each column, as suggested.

Figure 5-7, 9-10, 13 In caption, please briefly explain 'Case', so the reader does not have to refer back to Table 1 to understand the figure.

Added explanations.

Figure 11 Caption mistake: the a priori, should be the a priori.

Revised.

Figure 15 Legend text is misaligned, please correct Taylor Scores.

Revised.