Dear Editor,

We have addressed all of the comments. Our revisions are itemized below, including a point-by-point response and change to the comments and a marked-up manuscript version. Thanks!

Reply to Referee 1

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 (P2 L28, P4 L21-30, P5 L27, L31, P8 L3-5, P11,10-12, P12, 1-2).

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.

(P4 L28-P5 L2) 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 (P5 L27, P6 L15, P18 L22).

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

Now use 'WUS' consistently (P3 L6, 8, 11, 12, 17, 21, P5 L4).

Remove spaces between numbers and % or degree signs.

Deleted throughout the manuscript.

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

Revised (P2 L25, 29).

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

Changed throughout the manuscript.

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!)

(P2 L28) 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.

(P2 L30) 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.

(P3 L8) 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.

(P3 7-9) 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.

(P3 L19) '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?

(P3 L28) 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 (P2 L28, P4 L21-30, P5 L27, L31, P8 L3-5, P11,10-12, P12, 1-2).

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

(P3 L31) Moved as suggested.

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

(P5 L7) Deleted.

P21869 L17 please remove 'for clarity'

(P5 L4) 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?

(P5 L16-17) 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)

(P5 L18) Deleted.

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

(P5 L27-28) 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?

(P6 L7) 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.

(Fig. S1) Revised. Now we use '*r*' consistently.

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

(P6 L20) 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.

(P7 L23-24) Revised.

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

(P8 L3) Revised to 'We optimize here the scaling factors of emissions $\mathbf{X}/\mathbf{X}_a$ (rather than the actual emissions **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.'

(P8 L12-16) Revised as suggested.

Results and Discussion
Typo in title: discussion, not discussions

(P9 L17) 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.

(P9 L21) Added. Changed to 'The *a posteriori* emissions are 49.9 Gg at 2 °×2.5 ° and 47.3 Gg at 0.5 °×0.667 ° 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?

(P10 L6-13) 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.'

(P11 L10-12) 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.

(P14 L23) 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.

(P15 L18-20) 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 °×2.5 °.'.

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 âĹij 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.'

(P16 L2-9) Revised to '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, 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).'

(P16 L23) Deleted.

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

(P16 L30) Deleted.

21883 L10-15 shorten paragraph

(P17 L3-5) 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.

(P18 L22- P19 L13) 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' emisisons? 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.

Fig. S1. Added.

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

Fig. S2. 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).

Figs. 2-4. 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.

Fig. 8. Revised.

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

Fig. 12. Revised.

Reply to Referee 2

This study uses the GEOSH-CHEM adjoint model and BC observations from the IMPROVE network to invert summertime anthropogenic and biomass burning BC emissions over the western U.S. The authors' best estimate for BC emissions are approximately twice the magnitude of the a priori inventories, which is very similar to the results previously obtained by the same group (Mao et al., 2014). In my opinion, the real value of this paper lies in the various sets of sensitivity and pseudo observation experiments, designed to test the adjoint's ability. These methods are novel and of great interest to the community. The paper is generally well written. As a reviewer, I try in general to avoid recommending new experiments. However, in the present case, I do think that two sets of experiments (fortunately, relatively easy to do) would add to the comprehensiveness of the study. The first would be an inversion of the total BC emission, since the adjoint cannot distinguish individual source sectors. The other would a pseudo observation inversion to determine if observations placed at strategic locations would improve the inversion of total emissions or sources.

(P12 L10-21, P15 L12-26) Point well taken. We have conducted the two experiments as suggested and discussed the corresponding results in Sects. 4.1.3 and 4.2.

Page 21870, Line 27-page 21871, line 2: It is reassuring that the derivative of the cost function (J) calculated by the forward model is perfectly correlated with that calculated by the adjoint model. But why is the slope in Fig 2 not 1 (it is close to unity, but there is a 5% bias). For a perfectly linear species such as BC, one would image in the slope to be unity.

Fig. S1. Agree. We have recalculated the sensitivities and found a perfectly linear relationship between the forward model and the adjoint model, as shown in Fig. S1.

Page 21872, lines 15-16; page 21873, line 20-25: I appreciate that characterizing the error structures of a priori emission inventories is difficult. But one would imagine that the errors of the a priori emission inventory would be spatially correlated, perhaps strongly so, particularly

for biomass burning emissions (e.g., underestimation of agriculture fires over extensive farming regions). I see that this is hinted at in page 21873, line 20-25. However, the use of gamma does not completely resolve this issue. A few words about how the spatial correlation of error may impact your inversion would be helpful.

(P7 L19-23) Point well taken. Added discussions, 'The spatial correlations between the *a priori* errors have been proved to improve the inversion, particularly in regions adjacent to strong sources and less directly constrained (Stavrakou and Müller, 2006). For example, the assumption of no spatial correlation between *a priori* errors would underestimate the biomass burning emissions in regions close to the extensive agriculture fires. '

Page 21873, line 14-28: What is this "hybrid" form? Please write out the mathematical form.

(P8 L14) Added as Eq. (4).

Page 21878, lines 3-8: The authors used an indirect method to evaluate the adjoint's ability to distinguish between anthropogenic and biomass burning sources. To me, this is the most interesting experiment in the paper, but the texts here are not very clear. Please re-write.

(P12 L16-22) Revised to 'We conduct another inversion (Case 8) to examine how much the inversion can distinguish the collocated emissions. In each grid box, we add 2.5 Mg (~10% of the maximum emissions among the grid boxes) as a diagnosis to the (*a priori*) biomass burning emissions of BC and examine the degree to which the inversion results change the partitioning of biomass burning versus anthropogenic emissions by comparing the inversion results with those from Case 1 (or Case 7). The differences in the *a posteriori* emissions of BC between Cases 8 and 1 are shown in Fig. 6. '(Sect. 4.1.3)

Page 21878, lines 3-8: Also, if the inversion is unable to distinguish between anthropogenic and biomass burning sources, would it not make sense to do an inversion on the total emission? What are the results?

(P12 L13-16) One experiment Case 7 added in Table 1 and Sect.4.1.3. We find that 'the resulting total *a posteriori* emissions increase by a factor of 2.2 relative to the *a priori* and are within 2% of those from Case 1. '.

Page 21881, lines 13-24: Again, this set of sensitivity tests are very interesting. Would it be possible to find out, using pseudo observations, at which locations would observations be most useful in constraining BC emission totals and sources? For example, I would imagine observations on the eastern slope of the Rockies (leeward of biomass burning emissions and less affected by precipitation) would be most effective.

(P15 L12-26) Point well taken. We have added one experiment Pseudo 9 and used pseudo observations at the grid boxes with more than 5 fire counts in August (covering 50% of surface grid boxes). We find that ' With pseudo observations located at biomass burning source regions (Pseudo 9), the resulting *a posteriori* biomass burning emissions are 5% higher than those from the inversion with similar amount of pseudo observations (covering 50% of surface grid boxes, Pseudo 7), whereas the total *a posteriori* emissions are almost unchanged between Pseudos 7 and 9. Thus, pseudo observations located at source regions would be more effective to constrain sources.'.

Minor comments:
p21866, Line 13: What does 'both' refer to? Please specify. Also 'two-fold'. p21866, Line 14: '... their respective a priori emission inventories, ...'

(P2 L12-13) Revised to ' both anthropogenic and biomass burning emissions in the adjoint inversions increase twofold relative to the respective *a priori* emissions '.

p21866, Line 16 and line 20: What does the 'inversion system' refer to? Are you talking about the adjoint or the analytical inversion system?

(P2 L16 19) Revised to 'adjoint inversion system'.

p21867, Line 7: '... and cause global warming (...)': the global warming effect of BC was already mentioned in the previous sentence.

(P2 L30) Removed.

p21867, Line 24: "increase in fires": are you referring to an increase in fire frequency, burnt area, emissions, or a combination thereof?

(P3 L16-17) Revised to ' an increase in fires in terms of both fire frequency and burned area '

p21868, Lines 17-28: Here and throughout: When you say "inverse modeling" or "inverse system", it is not clear whether you are referring to the analytical inversion method, the adjoint method, or the general inversion problem. Please use consistent nomenclature.

(P4 L7) Revised to 'Inverse modeling in general'.

Estimates of Black Carbon Emissions in the Western United States Using the GEOS-Chem Adjoint model

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Abstract

We estimate black carbon (BC) emissions in the Western United States for July–September 2006 by inverting surface BC concentrations from the Interagency Monitoring of PROtected Visual Environment (IMPROVE) network using a global chemical transport model (GEOS-Chem) and its adjoint. Our best estimate of the BC emissions is 49.9 Gg at 2 °×2.5 ° (a factor of 2.1 increase) and 47.3 Gg at 0.5 °×0.667 ° (1.9 times increase). Model results now capture the observed major fire episodes with substantial bias reductions (~35% at 2 °×2.5 ° and ~15% at 0.5 °×0.667 °). The emissions are ~20–50% larger than those from our earlier analytical inversions (Mao et al., 2014). The discrepancy is especially drastic in the partitioning of anthropogenic versus biomass burning emissions. The August biomass burning BC emissions are 4.6–6.5 Gg and anthropogenic BC emissions 8.6–12.8 Gg, varying with the model resolution, error specifications, and subsets of observations used. On average both anthropogenic and biomass burning emissions in the adjoint inversions increase twofold relative to the respective *a priori* emissions, in distinct contrast to the halving of the anthropogenic and tripling of the biomass burning emissions in the analytical inversions. We attribute these discrepancies to the inability of the adjoint inversion system, with limited spatiotemporal coverage of the IMPROVE observations, to effectively distinguish collocated anthropogenic and biomass burning emissions on model grid scales. This calls for concurrent measurements of other tracers of biomass burning and fossil fuel combustion (e.g., carbon monoxide and carbon isotopes). We find that the adjoint inversion system as is has sufficient information content to constrain the total emissions of BC on the model grid scales.

1. Introduction

Black carbon (BC) is directly emitted from the incomplete combustion of carbonaceous fuels (Bond et al., 2004). Black carbon has substantial impacts on global climate because of its strong absorption of solar radiation (e.g., Horvath, 1993; Ramanathan and Carmichael, 2008), important influences in cloud processes (Jacobson, 2006), and significant impacts on snow and ice albedos (Flanner et al., 2007, 2009). The radiative forcing due to BC is 0.64 W m^{-2} globally (IPCC, 2013), ranked as the third-biggest human cause of global warming. Black carbon is also known as an important agent to both degrade air quality and affect human health (McMurry et al., 2004; Anenberg et al., 2011, 2012). Because of its shorter lifetime relative to long-lived greenhouse

gases, BC reduction may provide an efficient near-term solution to mitigate global warming and to improve air quality and public health simultaneously (Ramanathan and Carmichael, 2008; Bond et al., 2013).

The deposition of BC on glaciers is known to be an important driver to the observed rapid glacier retreat (Xu et al., 2009; Painter et al., 2013) and further impacts the regional hydrological cycle over mountain ranges (Qian et al., 2009). In the Western United States (WUS), mountain snowmelt accounts for at least 70% of the annual stream flow (Qian et al., 2009). In the recent decades, the WUS 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). It is thus imperative to better understand the sources, transport, and deposition of BC in the WUS mountain ranges.

Recent studies have shown that the biomass burning BC emissions in the WUS were underestimated by a factor of two in both the absolute magnitudes and the timing and location of the emissions (Mao et al., 2011, and references therein). The large uncertainty is partially because previous burned area algorithms lacked the ability to detect small fires (Giglio et al., 2010; Randerson et al., 2012). Long-term records have shown an increase in fires in terms of both fire frequency and burned area in the WUS over the past 30 years because of the rising spring and summer temperatures (Westerling et al., 2006; Peterson and Marcinkowski, 2014; Jin et al., 2014) and increasing urbanization (e.g., Cannon and DeGraff, 2009). This upward trend is expected to continue as a result of the warming climate (Spracklen et al., 2009; Yue et al., 2013). Biomass burning emissions will conceivably have an even larger contribution to BC in the WUS in this century, especially considering that North American anthropogenic emissions have been steadily decreasing as a result of effective emission controls (Novakov et al., 2003; Bond et al., 2007; Ramanathan and Carmichael, 2008).

Knowledge of the emissions of a chemical species is imperative for better understanding of its transport, distribution, and removal. Traditional bottom-up emission estimates generally depend on emission factors using socioeconomic, energy, land use, or environmental data (Bond et al., 2007, 2013; Lu et al., 2011). Inverse modeling is able to improve the bottom-up emission estimates by minimizing an error-weighted least squares cost function (Rodgers, 2000). There are two methods to achieve the minimum of the cost function, the so-called analytical inversion and adjoint (i.e. variational) inversion (Kopacz et al., 2009, and references therein). The

analytical method obtains an analytical solution by explicitly constructing a Jacobian matrix. However, the analytical method limits the number of the observations and the number of the sources and source regions that could be optimized because it is computationally expensive. Alternatively, the adjoint method seeks a numerical solution iteratively by using a suitable optimization algorithm (e.g., the conjugate gradient method) and is thus able to handle a very large number of observations and a large state vector resolved on a model grid scale.

Inverse modeling in general is suited for estimating emissions of unreactive or weakly reactive chemical species when their atmospheric concentrations are linearly or weakly non-linearly dependent on emissions (Müller and Stavrakou, 2005). These species include but are not limited to carbon dioxide (e.g., Gloor et al., 1999; Chevallier et al., 2007; Pickett-Heaps et al., 2011), methane (e.g., Hein et al., 1997; Meirink et al., 2008; Wecht et al., 2012), and carbon monoxide (CO) (Stavrakou and Müller, 2006; Arellano et al., 2004, 2006, 2007; Chevallier et al., 2009; Jones et al., 2009). Despite the non-linear complexities of the inversion system for short-lived tracer species, several studies have attempted to constrain emissions for nitrogen oxides (e.g., Martin et al., 2003, 2006; Chai et al., 2009; Lin et al., 2010; Zyrichidou et al., 2013), sulfur dioxide (Lee et al., 2012), and ammonia (Zhu et al., 2013; Paulot et al., 2014). The inverse method has also been used to constrain emission fluxes of aerosols, for instance, inorganic particulate matter (Henze et al., 2009; Xu et al., 2013) and dust (Yumimoto et al., 2008; Wang et al., 2012).

A number of modeling studies have attempted to constrain and attribute BC emissions on regional to continental scales. Several studies have used multiple regressions to estimate annual mean emissions of primary carbonaceous aerosols in the U.S. (Park et al., 2003) and in China (Fu et al., 2012; Wang et al., 2013). A global chemical transport model (CTM) and its adjoint was used to attribute the source regions of BC in the Himalayas and the Tibetan Plateau (Kopacz et al., 2011). Anthropogenic and biomass burning emissions of BC during the Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) (Huebert et al., 2003; Seinfeld et al., 2004) were estimated using a continental-scale CTM (stretched over the Pacific basin) and its adjoint (Hakami et al., 2005). 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 WUS 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, relative to the corresponding *a priori* (Mao et al., 2014).

Here we apply the adjoint inversion method (Henze et al., 2007, 2009) to improve estimates of BC emissions in the WUS (defined hereinafter as 30 °–50 °N, 100 °–125 °W) on model grid scales by inverting the surface BC concentrations from the Interagency Monitoring of PROtected Visual Environment (IMPROVE, Malm et al., 1994) network using the GEOS-Chem global 3-D CTM and its adjoint. We use the observations for 2006 from 69 mostly mountainous sites in the WUS (Fig. 1). We focus our analysis on biomass burning emissions during the large fire season of July–September in the region (Mao et al., 2011, 2014).

2. GEOS-Chem and its adjoint

We apply the GEOS-Chem global 3-D CTM (Bey et al., 2001; with many updates thenceforth) to analyze IMPROVE BC data. Here we use GEOS-Chem version 8-02-01 (available at http://geos-chem.org) driven by GEOS-5 meteorological data. The detailed model configurations are as discussed by Mao et al. (2011, 2014). 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). Biomass burning emissions of BC are from the Global Fire Emissions Database version 2 (GFEDv2) (Randerson et al., 2007; van der Werf et al., 2006), with improved spatiotemporal distributions using the active fire counts from the Moderate Resolution Imaging Spectroradiometer (MODIS) (Mao et al., 2014). For computational expediency, we conduct 'offline' simulations of carbonaceous aerosols (Mao et al., 2011, and references therein) for 2006 at both 2 °×2.5 ° (globally) and 0.5 °×0.667 ° (nested over North America, 40 °–140 °W longitudes, 10 °–70 °N latitudes, cf. Fig. 1 in Wang et al., 2004) horizontal resolutions, following Mao et al. (2014). The first three months are used for initialization and we focus our analysis on July–September. Model results are sampled at the corresponding location and time of IMPROVE observations.

We use the GEOS-Chem adjoint (Henze et al., 2007, 2009) to estimate BC emissions in the WUS. A particular type of application of the adjoint is source attribution of chemical species at individual sites (e.g., Zhang et al., 2009; Kopacz et al., 2011; Parrington et al., 2012; Walker et al., 2012). Here we use the adjoint of the "offline" simulation of BC at 2 °×2.5 ° (globally) and 0.5 °×0.667 ° (nested over North America, Jiang et al., 2015a). The computational cost of the

adjoint simulation is 50% greater than that of the corresponding forward simulation. We validate the adjoint simulation of BC by comparing the adjoint gradients and the forward model sensitivities calculated using finite difference approximation (Henze et al., 2007; Zhu et al., 2013):

$$\wedge = \frac{J(\sigma+\delta\sigma)-J(\sigma)}{\delta\sigma} \quad (1)$$

where J is the cost function (Mao et al., 2014, and references therein), as discussed below in Sect. 3.1, and σ the scaling factor of BC emissions. We use here $\delta\sigma$ = 0.1, following Henze et al. (2007). Specifically we calculate the sensitivity of the BC mass at the surface with respect to the scaling factors of biomass burning and anthropogenic emissions of BC. Fig. S1 shows the results from 1-week simulations for biomass burning (top panel) and anthropogenic emissions (bottom panel) for August 2006. The adjoint and finite difference sensitivities are in excellent agreements ($r \approx 1$), reaffirming the accuracy of the adjoint code.

3. Inversion approach

3.1 The adjoint solution to the inverse problem

Consider the general problem of inferring emissions (state vector $\boldsymbol{X}$) from a set of given observations (observation vector $\boldsymbol{Y}$) with error $\boldsymbol{\varepsilon}$. They are related via a forward model F as follows (Rodgers, 2000):

$$\boldsymbol{Y} = F(\boldsymbol{X}, b) + \varepsilon \quad (2)$$

where $\boldsymbol{X}$ are the monthly biomass burning or anthropogenic emissions of BC in each model grid box in the present study, $\boldsymbol{Y}$ the 24-hour average surface BC concentrations from the 69 IMPROVE sites (Fig. 1), b the model variables not directly retrieved from the inversion, F the GEOS-Chem model, and ε the observation error (measurement and forward model errors). Based on Bayes' theorem and the assumption of Gaussian error distributions (Rodgers, 2000), the optimal or Maximum *A Posteriori* (MAP) solution for $\boldsymbol{X}$ given $\boldsymbol{Y}$ is equivalent to finding the minimum of a cost function $J(\boldsymbol{X})$:

$$J(\boldsymbol{X}) = \frac{1}{2}\gamma_r(\boldsymbol{X}-\boldsymbol{X}_a)^T\mathbf{S}_a^{-1}(\boldsymbol{X}-\boldsymbol{X}_a) + \frac{1}{2}\sum_{i=0}^{N}[\boldsymbol{Y}_i - F(\boldsymbol{X}_i)]^T\mathbf{S}_\varepsilon^{-1}[\boldsymbol{Y}_i - F(\boldsymbol{X}_i)] = \gamma_r J_b + J_o \quad (3)$$

where $\boldsymbol{X}_a$ and $\mathbf{S}_a$ are the *a priori* emissions and the associated error covariance, $\mathbf{S}_\varepsilon$ the observational error covariance, and γ_r the regularization parameter that adjusts the relative

constraints by the observation term (i.e., the prediction term, J_o) and the background term (i.e., the penalty term, J_b) of the cost function (Hakami et al., 2005; Müller and Stavrakou, 2005; Henze et al., 2007; Kopacz et al., 2009). An observation term is added to the cost function for each additional data source during the time interval [t_0, t_N].

The adjoint approach seeks to minimize the cost function $J(\boldsymbol{X})$ numerically and iteratively rather than analytically (Henze et al., 2007, 2009). Starting from an initial guess (i.e., the *a priori* emissions), the adjoint model efficiently computes the cost function gradients. A quasi-Newton L-BFGS algorithm (Liu and Nocedal, 1989) is then used to minimize the cost function iteratively, taking as input the cost function and its gradient. Such iterative optimizations using GEOS-Chem and its adjoint have been discussed in details previously (Henze et al., 2007, 2009; Kopacz et al., 2009, 2010).

3.2 Error specifications

A key aspect of inverse modeling is the specification of the error covariance matrices of variable parameters and observations (Palmer et al., 2003; Heald et al., 2004). We set the *a priori* and observation errors following Mao et al. (2014). We assume for separate inversions presented here an uncertainty of 30, 50, or 200% for anthropogenic BC emissions and 300 or 500% for biomass burning BC emissions. We assume that the *a priori* errors are spatially uncorrelated. The spatial correlations between the *a priori* errors have been proved to improve the inversion, particularly in regions adjacent to strong sources and less directly constrained (Stavrakou and Müller, 2006). For example, the assumption of no spatial correlation between *a priori* errors would underestimate the biomass burning emissions in regions close to the extensive agriculture fires. We set the observation error at 30, 50, or 100%, which includes the model, representation, and measurement errors. Setting these errors in relative terms can become problematic when the observed BC concentrations are vanishingly small. These small values tend to skew the inversion toward matching the minimal errors. We thus set an absolute error of 0.04 μg m^{-3} based on the estimated observation errors. We showed previously that the combination of 50% uncertainty for anthropogenic emissions, 500% uncertainty for biomass burning emissions, and 30% total observation error provided the best retrieval results in the analytical inversions (Mao et al., 2014). We adopt this set of error specifications in the standard inversion in the present study (Case 1, Table 1). The results are compared with those from the analytical inversions of Mao et al. (2014).

3.3 Emission scaling factors $\boldsymbol{X}/\boldsymbol{X}_a$

We optimize here the scaling factors of emissions $\boldsymbol{X}/\boldsymbol{X}_a$ (rather than the actual emissions $\boldsymbol{X}$), as a standard practice in adjoint inversion studies (Henze et al., 2009). The form of the scaling factors in an adjoint inversion is crucial for the inversion to efficiently and rapidly converge to a solution (Jiang et al., 2015b). When the optimization is directly on the scaling factors expressed linearly as $\boldsymbol{X}/\boldsymbol{X}_a$ (i.e., the cost function gradient is computed with respect to $\boldsymbol{X}/\boldsymbol{X}_a$), the regions with strong *a priori* emissions tend to dominate the optimization, manifested in unrealistically large changes of emissions in these regions but limited variations in the regions with weak *a priori* emissions. Alternatively, when the optimization is instead on the logarithm of the scaling factors, $\ln(\boldsymbol{X}/\boldsymbol{X}_a)$ (i.e., the cost function gradient is now computed with respect to $\ln(\boldsymbol{X}/\boldsymbol{X}_a)$), the optimization can potentially result in an unbalanced convergence that is much faster for the regions with positive biases than for the regions with negative biases. Here we calculate cost function gradients with a hybrid form of scaling factors (Jiang et al., 2015b),

$$gradients = \begin{cases} \dfrac{x}{x_a} \le 1 & \dfrac{\partial J}{\partial \ln(x/x_a)} = \dfrac{\partial J}{\partial x} \times x_a \times \dfrac{x}{x_a} \\ \dfrac{x}{x_a} > 1 & \dfrac{\partial J}{\partial \dfrac{1}{2}\left[(x/x_a)^2 - 1\right]} = \dfrac{\partial J}{\partial x} \times x_a / \dfrac{x}{x_a} \end{cases} \quad (4)$$

so that the resulting optimization converges equally efficiently for the regions with positive or negative biases.

3.4 The regularization parameter γ_r

The assumption that a priori errors are spatially uncorrelated hinges on the consideration that the spatial resolution of the CTM is much larger than the correlation length scale of the individual emission sources (Henze et al., 2009). However, the uncertainties of emissions from different model grid boxes (e.g., ~200×250 km^2 at 2°×2.5°) within a region (e.g., a country) are usually correlated (Stavrakou and Müller, 2006). Without explicitly enforcing these correlations, a regularization parameter, which ensures a smooth solution to the inversion, is often used to rectify the aforementioned inconsistency, by ensuring the *a posteriori* emissions remain sufficiently close to the *a priori* values, which themselves reflect such correlations owing to the nature of bottom-up emissions inventories (Rodgers, 2000; Henze et al., 2009). Here we use the regularization parameter γ_r to balance the two terms of the cost function (Eq. 3) (Hakami et al.,

2005; Müller and Stavrakou, 2005; Henze et al., 2007; Kopacz et al., 2009). A large γ_r relaxes the solution toward the *a priori* constraint while limiting the influence of the observation term, resulting in over-smoothing of the solution. In contrast, a small γ_r largely curtails the influence of the *a priori* constraint. To find an optimal γ_r value, we conduct inversions with a range of γ_r (10, 1, 0.1, 0.01, 0.001, and 0.0001). The resulting *a posteriori* cost function values, normalized by the initial value, are shown in Fig. S2 (top). We use $\gamma_r = 0.001$ in the analysis hereinafter, as that provides a small normalized *a posteriori* cost function and a sufficient cost function reduction.

As an example, Fig. S2 (bottom) shows the cost function reduction for August 2006. We regard the minimization as having converged sufficiently when the cost function changes less than 2% during the last three iterations. The cost function converges in 10–20 iterations (35% reduction at 2 °×2.5 ° and 50% at 0.5 °×0.667 °). The cost function values are of the same order as the number of observations used in the inversion (~690). The penalty term (Sect. 3.1) is a mere 3% of the *a posteriori* cost function at 2 °×2.5 ° and 8% at 0.5 °×0.667 °, therefore the influence of the *a priori* is likely rather modest.

4. Results and discussion

Our standard adjoint inversion is at 2 °×2.5 °, with uncertainties of 50% for anthropogenic emissions, 500% for biomass burning emissions, and 30% for the observation (Case 1, Table 1). The *a posteriori* emissions are 49.9 Gg at 2 °×2.5 ° and 47.3 Gg at 0.5 °×0.667 ° for July–September, substantially higher than the *a priori* (24.3 Gg), because the modeled surface BC concentrations are largely biased low at most IMPROVE sites (Mao et al., 2011, 2014).

We focus our discussions hereinafter on August only, unless stated otherwise, for the sake of concision and clarity. Fig. 2 shows the *a priori* and the *a posteriori* monthly anthropogenic and biomass burning BC emissions from the standard adjoint inversion for August. The anthropogenic and biomass burning emissions are adjusted (higher or lower) alike in most grid boxes. Both the anthropogenic and biomass burning emissions increase twofold overall. The biomass burning emissions increase by varying factors (Table 1): 2.3 in the Rockies, 2.8 in California and the Southwest, and 1.5 in the Pacific Northwest – the regions are defined as in Mao et al. (2014).

The sensitivity of the cost function $J(\boldsymbol{X})$ to the BC emissions is a useful metric for evaluating the inversions. Following Henze et al. (2009), we normalize the sensitivity as follows,

$$\frac{\partial J(\boldsymbol{X})}{\partial x_{m,i}} \frac{x_{m,i}}{J(\boldsymbol{X})} \quad (5)$$

It is the percentage of the cost function response to the fractional change in the BC emission source *m* (biomass burning, anthropogenic or total emissions) in model grid box *i*. As such, negative sensitivities are regions in which the model underestimates actual emissions and an increase of BC emissions would improve model agreement with the observations. It is the opposite for positive sensitivities. The results for August are shown in Fig. 3. 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. The inversions generally result in large reductions to both the positive and negative sensitivities (~90% on average at 2 °×2.5 ° and 0.5 °×0.667 °).

4.1 Sensitivity analyses

Here we examine the sensitivity of the adjoint inversions to error specifications, choice of observations, collocated emissions, and the model resolution. These sensitivity analyses also provide a measure of the robustness of the inversions (Mao et al., 2014). For that purpose we conduct adjoint inversions at 2 °×2.5 ° and 0.5 °×0.667 ° (Cases 2–8, Table 1) in addition to the standard inversion (Case 1), with assorted *a priori* and observation errors, different subsets of the IMPROVE data, and collocated anthropogenic and biomass burning emissions. The results are compared and contrasted with those from the standard adjoint inversion in the discussions hereafter, unless stated otherwise. We find that the inversions generally show comparable and consistent results with those from the standard inversion. The ensemble *a posteriori* biomass burning emissions (Cases 1–8) are 4.6–6.5 Gg (a factor of 1.7–2.3 relative to the *a priori*) and anthropogenic emissions 8.6–12.8 Gg (a factor of 1.5–2.2 increase).

4.1.1 Error specifications

We first conduct adjoint inversions (Cases 2–5) to examine the sensitivity of the inversions to the *a priori* and observation errors. The *a posteriori* emissions are 3% lower when we reduce the

uncertainty of the *a priori* biomass burning emissions from 500 to 300% (Case 2). Reducing the uncertainty of the *a priori* anthropogenic emissions from 50 to 30% brings no appreciable change to the *a posteriori* emissions (Case 3). Quadrupling that uncertainty (from 50 to 200%) increases the *a posteriori* emissions by 10% (Case 4). We find that the inversions are more sensitive to the observation error than the *a priori* error. For instance, an increase from 30 to 100% of the observation error (Case 5) results in a 16% decrease in the *a posteriori* emissions.

4.1.2 Choices of observations

A robust inversion critically relies on the spatiotemporal coverage of the observations. For carbon dioxide, a minimum of 10 sites per region were needed in analytical inversions (Gloor et al., 1999). For BC, the number of site is usually smaller. Only ~1000 BC observations were used to optimize ~20000 variables in an adjoint study (Hakami et al., 2005) during ACE-Asia (Huebert et al., 2003; Seinfeld et al., 2004). In this study, we use ~690 observations to constrain ~600 variables at 2 °×2.5 ° and ~10000 at 0.5 °×0.667 °.

Here we conduct two inversions to probe the sensitivity of the inversion to observations by using subsets of the IMPROVE data and comparing the results with those from the standard inversion (Case 1). In the inversion (Case 6), we set aside 13 (~20% of the 69) sites with $\chi^2 > 1.5$ and large model-observation departure (> 0.5 μg m^{-3}). χ^2 is calculated as the square of the ratio of the difference between modeled and observed surface BC concentrations to the observation accuracy. The measurements from these 13 sites are used as independent observations, whereas the measurements from the remaining 56 sites are used in the inversion. We find that the resulting *a posteriori* emissions (Fig. 4) are within 6% of those from Case 1 (Fig. 2). The emissions differ significantly only in ~10% of the grid boxes, mainly in the Pacific Northwest and the Rockies. The resulting surface BC concentrations averaged over the 13 sites are within 15% between the two Cases. There is a ~15% reduction in the mean bias of the surface BC concentrations (averaged over the 13 sites) for Case 6 and 20% for Case 1. In another inversion (not shown), we set aside four (~5% of the 69) sites with $\chi^2 > 2$. The results are also consistent with those from Case 1.

4.1.3 Collocated emissions

In the WUS mountain ranges, biomass burning BC emissions are substantially underestimated (Mao et al., 2011). There are large uncertainties in the temporal variation and spatial distribution of fire emissions (Langmann et al., 2009). Small fires are likely a major source of these uncertainties (Randerson et al., 2012). For instance, small fires can lead to large relative errors (50–100%) in burned area estimates (Korontzi et al., 2006; Giglio et al., 2006, 2010; McCarty et al., 2009; Roy and Boschetti, 2009).

Fig. 5 shows monthly anthropogenic and biomass burning emissions in each 2 °×2.5 ° model grid box in the WUS. Collocated anthropogenic and biomass burning emissions are in most of the grid boxes. The anthropogenic emissions are larger than the biomass burning emissions in 80% of the grid boxes and still significant in the remaining 20%. Here we conduct two inversions (Cases 7–8) to examine the ability of the adjoint inversion system to distinguish collocated biomass burning versus anthropogenic emissions, especially in the regions where the biomass burning emissions are likely underestimated. We conduct one inversion (Case 7) on the total emissions, considering an error of 200% for the *a priori* emissions. We find that the resulting total *a posteriori* emissions increase by a factor of 2.2 relative to the *a priori* and are within 2% of those from Case 1. We conduct another inversion (Case 8) to examine how much the inversion can distinguish the collocated emissions. In each grid box, we add 2.5 Mg (~10% of the maximum emissions among the grid boxes) as a diagnosis to the (*a priori*) biomass burning emissions of BC and examine the degree to which the inversion results change the partitioning of biomass burning versus anthropogenic emissions by comparing the inversion results with those from Case 1 (or Case 7). The differences in the *a posteriori* emissions of BC between Cases 8 and 1 are shown in Fig. 6. The anthropogenic and biomass burning emissions from Case 8 show opposite and disproportional changes, relative to the corresponding emissions from Case 1. Specifically, there is an approximately linear increase (i.e., Case 8 relative to Case 1) in the biomass burning emissions (by more than 5 Mg in nearly every grid box) and a linear decrease (again, Case 8 relative to Case 1) in the anthropogenic emissions (by ~3 Mg, except in California). However, the anthropogenic and biomass burning emissions of BC, when summed over the WUS, each increases twofold (relative to the corresponding *a priori*) in both Cases 8 and 1 – the total emissions remain essentially the same (less than 1% difference) between the two Cases (Table 1). The inversion system tends to overcompensate the deficit of biomass burning emissions by disproportionately increasing anthropogenic emissions instead in the same grid box.

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.

4.1.4 Model resolution

GEOS-Chem simulations generally provide better agreements with observations at 0.5 °×0.667 ° than at 2 °×2.5 ° for CO (Wang et al., 2004; Chen et al., 2009), ozone (Zhang et al., 2011), mercury (Zhang et al., 2012), and BC (Mao et al., 2014). We found in a companion study (Mao et al., 2014) that the *a posteriori* BC emissions from analytical inversions of the IMPROVE observations were considerably lower at 0.5 °×0.667 ° than at 2 °×2.5 °. Intuitively, the larger smearing of the emissions at the coarser resolution results in larger model-observation discrepancies (Chen et al., 2009). The larger discrepancies in turn tend to drive the inversion system toward imposing larger emissions at the coarser than the finer resolutions when minimizing the said discrepancies.

As a comparison to the standard inversion at 2 °×2.5 ° (Case 1), we conduct another adjoint inversion at 0.5 °×0.667 ° (Case 9) with the same error specifications. The resulting *a posteriori* emissions are shown in Fig. 7. The biomass burning emissions are 53% larger in Case 9 than in Case 1 in the Pacific Northwest but 27% lower in the Rockies and 43% lower in California and the Southwest. The total emissions are considerably lower at 0.5 °×0.667 ° (27% lower for anthropogenic and 10% for biomass burning) yet provide better agreement with the observations (Sect. 4.4). The *a posteriori* emissions from the standard analytical inversion (Mao et al., 2014) are also shown in Fig. 7 for comparison. The differences between the analytical and adjoint inversions (Table 1) are slightly larger at 0.5 °×0.667 ° (53%) than at 2 °×2.5 ° (39%). The larger differences reflect in part that the adjoint inversion system has even more difficulty in constraining the emissions at the finer grid scale (Sects. 4.1.3 and 4.2).

4.2 Pseudo observations

We use pseudo observations of BC concentrations in another group of inversions (Table 2) to further examine the sensitivity of the adjoint inversions to collocated emissions, error specifications, and observations. We generate the pseudo observations by increasing the *a priori* biomass burning emissions of BC in each grid box threefold. The total amount of the *a priori* emissions added is 5.6 Gg. The frequency of the pseudo observations are 24-hour averages for

every three days, following the IMPROVE measurements of BC. We then invert the pseudo observations at 2 °×2.5 ° and with the same *a priori* emissions as those used in the standard inversion (Case 1). We examine whether the inversions are able to fully recover the emissions used to generate the pseudo observations. Specifically, we expect the *a posteriori* biomass burning emissions to increase threefold relative to the *a priori*, whereas the anthropogenic emissions remain unchanged.

We first conduct two inversions (Pseudos 1–2) to investigate the ability of the adjoint inversion system to distinguish collocated anthropogenic and biomass burning emissions. We consider two extreme scenarios: the pseudo observations are in every surface grid box (Pseudo 1) and in every grid box in the lowest 15 vertical layers (Pseudo 2). Other aspects of the two inversions remain the same as those of the standard inversion (Case 1). We find that the results are nearly indistinguishable. The *a posteriori* cost function is greatly reduced (by 95% in Pseudo 1 and by 97% in Pseudo 2). The *a posteriori* emissions both increase by exactly 5.6 Gg, fully recovering the added biomass burning emissions. However, the increase is uneven and not limited to the biomass burning emissions. The biomass burning emissions increase by a factor of 2.3 and anthropogenic emissions by a factor of 1.3. The inversions thus falsely impose larger anthropogenic emissions to minimize the large discrepancies between the model predictions and the pseudo observations.

In the next three inversions (Pseudos 3–5), we examine the sensitivity of the inversion system to the constraints for anthropogenic versus biomass burning emissions. The uncertainty of the anthropogenic emissions is reduced to 10% in Pseudo 3. We assume that the anthropogenic emissions are perfect and leave them unchanged in Pseudo 4. In Pseudo 5, we let the biomass burning emissions remain unaltered. Other aspects of Pseudos 3–5 remain the same as those of Pseudo1. We find that the resulting *a posteriori* emissions from the former two (Pseudos 3 and 4) recover fully the added (biomass burning) emissions. The biomass burning emissions increase by a factor of 2.5 in Pseudo 3 (versus 2.3 in Pseudo 1) and by a factor of 2.9 in Pseudo 4. However, the *a posteriori* emissions from Pseudo 5 increase by only 4.2 Gg, recovering just 75% of the added (biomass burning) emissions.

Concurrent measurements of other combustion tracers, for example, CO and carbon isotopes, can conceivably provide additional information to distinguish collocated BC emissions. Previous studies have shown that the ratio of BC to CO significantly varies with the fuel types and thus is

a good indicator for identifying BC sources (Spackman et al., 2008; Han et al., 2009; Subramanian et al., 2010; Reche et al., 2011). Carbon isotopes such as ^{14}C are known to be present at small and more or less constant levels in biogenic emissions but absent in fossil fuels (Schichtel et al., 2008). Ample studies heretofore have shown that ^{14}C is useful for analyzing the source apportionment of atmospheric carbonaceous aerosols (Heal, 2014, and references therein). ^{14}C measurements are currently only available from short-term studies in part because of the relatively high cost of deploying such measurements in routine monitoring networks (Lewis et al., 2004; Bench et al., 2004, 2007; Szidat et al., 2006).

Additionally, we conduct four inversions (Pseudos 6–9) to examine the minimum number of observation sites needed for the inversions. The inversions are the same as Pseudo 1, except that the pseudo observations are randomly distributed in 75% (Pseudo 6), 50% (Pseudo 7), or 25% (Pseudo 8) of the surface grid boxes, or in the surface grid boxes with larger than 5 fire counts per month (covering ~50% of surface grid boxes, Pseudo 9). The resulting *a posteriori* emissions recover 94% (Pseudo 6), 93% (Pseudo 7), 80% (Pseudo 8), and 93% (Pseudo 9) of the added (biomass burning) emissions. Randomly, surface observations covering at least 50% of the model grid boxes are needed to estimate the total BC emissions on the model grid scale. In our case, 69 IMPROVE sites are used to constrain BC emissions in ~100 surface grid boxes at $2°\times2.5°$ and ~1500 at $0.5°\times0.667°$. 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 $2°\times2.5°$. With pseudo observations located at biomass burning source regions (Pseudo 9), the resulting *a posteriori* biomass burning emissions are 5% higher than those from the inversion with similar amount of pseudo observations (in 50% of surface grid boxes, Pseudo 7), whereas the total *a posteriori* emissions are almost unchanged between Pseudos 7 and 9. Thus, pseudo observations located at source regions would be more effective to constrain sources.

4.3 Adjoint versus analytical inversions

The analytical method is limited to constraining emissions over aggregated regions because of computational limitations, whereas the adjoint method is able to describe emission variability on finer scales and more efficiently (Kopacz et al., 2009). There are large differences in the *a*

posteriori emissions between the analytical and adjoint inversions, not only in the spatial distributions but also in the magnitudes (Figs. 5 and 10). 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, 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.

Mao et al. (2014) have examined in detail the quality of the analytical inversions. The robustness of the analytical inversions and the relative consistency in the total *a posteriori* emissions from the two inversion methods therefore imply that the adjoint inversion results, at least the total emissions, are robust on the model grid scale. We will examine the robustness of the adjoint inversions further in Sect. 4.4. The large differences in the *a posteriori* anthropogenic and biomass burning emissions between the two inversion methods are largely because the inversion system has difficulty effectively distinguishing collocated biomass burning and anthropogenic emissions on model grid scales. As a result, the adjoint inversions tend to falsely impose larger anthropogenic emissions in the regions where the collocated biomass burning emissions are too low (Sects. 4.1.3 and 4.2). Jiang et al. (2011) also found that the adjoint inversion system is unable to distinguish CO emissions from collocated combustion and oxidation sources and they therefore lumped the two sectors in their inversions. The differences are also due to the large aggregation errors in the analytical inversions and the assumption of spatially uncorrelated *a priori* errors in the adjoint inversions (Sects. 3.2 and 3.4).

We further separate the anthropogenic-dominated regions to examine the ability of the adjoint inversion system to constrain collocated emissions. In the regions where anthropogenic emissions are dominant, model surface BC concentrations are in good agreement with IMPROVE observations (Mao et al., 2011) and both the *a posteriori* anthropogenic and biomass burning emissions see substantial yet still relatively small increases. For example, the *a posteriori* anthropogenic and biomass burning emissions in Washington and Oregon increase by 39 and 29%. However, in the regions where biomass burning emissions become more important but significantly underestimated, model surface BC concentrations are biased low and both the *a*

posteriori anthropogenic and biomass burning emissions increase dramatically. 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.

4.4 Evaluation against observations

Model simulated surface BC concentrations with the *a posteriori* emissions show significant enhancements and largely reproduce both the synoptic variability and magnitudes of the observed surface BC concentrations, not only at individual sites (Fig. 8) but also on average at four altitude ranges (below 1, 1–2, 2–3, and above 3 km) (Fig. 9). For instance, model surface BC concentrations after the adjoint inversions capture the major fire episodes at Starkey, OR (45.2 °N, 118.5 °W, 1.26 km) and Lassen Volcanic, CA (40.5 °N, 121.6 °W, 1.73 km). The adjoint inversions at 0.5 °×0.667 ° provide better agreements with the observations than the analytical inversion results do at some sites, for example, Three Sisters, OR (44.3 °N, 122.0 °W, 0.89 km) and Pasayten, WA (48.4 °N, 119.9 °W, 1.63 km). At other sites, Jarbidge Wild, NV (41.9 °N, 115.4 °W, 1.87 km), for example, results from the analytical inversions are noticeably better. The two inversion results differ the most at 1–2 km altitudes and to a lesser degree at higher altitudes, for example, Bridger Wild, WY (43.0 °N, 109.8 °W, 2.63 km). The *a posteriori* emissions lead to an average bias reduction of ~50% in the simulated surface BC concentrations at 1–2 km altitudes (Fig. 9). Model simulated surface BC concentrations with the *a posteriori* emissions from the adjoint inversions, especially at 0.5 °×0.667 °, show substantial enhancements during major fire episodes. The enhancements are evident at all altitudes (up to 0.2 μg m^{-3} at 1–2 km and 0.1 μg m^{-3} at 2–3 km). The *a posteriori* emissions lead to large mean bias reductions (34% at 2 °×2.5 ° and 20% at 0.5 °×0.667 ° for August), as shown in Fig. 10. The frequency distributions of the bias of the 24-hour average surface BC concentrations are Gaussian (Fig. 11), as expected. The inversions reduce both the mean (by ~35% at 2 °×2.5 ° and ~15% at 0.5 °×0.667 ° for July–September) and standard deviation of the biases.

Taylor diagram and skill score (*S*) are useful measures of model accuracy. The diagram relates the centered root mean square error (RMSE), the pattern correlation (*r*) and the standard

deviation (σ) of observations and model results (Taylor, 2001). *S* (0–1) increases with increasing correlations and as the modeled variance approaches the observed variance. Fig. 12 presents the resulting diagram and skill scores of the observations and the multitude of model results. Model results with the *a posteriori* emissions are consistently in better agreement with the observations, especially using the nested model. The *a posteriori* emissions lead to higher *r* (by 11–48% on average), larger σ (by 27–122% on average), and lower centered RMSEs, thereby increasing the skill scores (by 43–221%). The *a posteriori* emissions from the adjoint inversion at 0.5 °×0.667 ° show the smallest centered RMSE and largest *r*, whereas the *a posteriori* emissions from the analytical inversion at 0.5 °×0.667 °show the largest σ and *S* values.

There are large uncertainties in the *a posteriori* emissions, as evident in the 20–30% low bias in modeled surface BC concentrations. The uncertainties are partially because of the limitations of the inversion system, in both the nature of the inverse modeling and the spatiotemporal coverage of IMPROVE observations (see Sects. 4.1.3 and 4.2). The adjoint inversion system has sufficient information to constrain the total emissions of BC, especially at the coarse resolution 2 °×2.5 °. The inversion system however has difficulty in partitioning collocated anthropogenic versus biomass burning emissions. Furthermore, comparing localized observations with coarse-resolution model results is inherently problematic (Mao et al., 2011; Fairlie et al., 2007). It is even more so because many of the IMPROVE sites are mountainous and the associated upslope flow is difficult to represent in a global model.

5. Summary and conclusions

We have applied adjoint inversions to estimate biomass burning and anthropogenic emissions of BC in the WUS for July–September 2006 by inverting the surface BC concentrations from the IMPROVE network using the GEOS-Chem chemical transport model and its adjoint. The *a posteriori* emissions of BC differed considerably between the adjoint and analytical inversions (Mao et al., 2014), especially in the partitioning of anthropogenic versus biomass burning emissions. The total was ~20–50% larger in the adjoint inversions than in the analytical inversions. Both the biomass burning and anthropogenic emissions from the adjoint inversions doubled, whereas the analytical inversions showed factors of 3–5 increases in the former and ~50%

reductions in the latter. We attributed these differences to the inability of the adjoint inversion system to effectively distinguish collocated biomass burning and anthropogenic emissions on the model grid scales. That inability resulted in excessively large anthropogenic emissions in the regions where biomass burning emissions were underestimated.

The inversions with various pseudo observations indicated that observations of surface BC concentration covering half of the model grid boxes had sufficient information to constrain the total emissions of BC on the model grid scales. IMPROVE observations of BC have sufficient information to constrain the total BC emissions at the model grid scales, especially at 2 °×2.5 °. The limitations of the adjoint inversion system, including the spatiotemporal coverage of the IMPROVE observations of BC, call for concurrent measurements of other combustion tracers (e.g., CO and carbon isotopes). Other factors may also improve the inversions, e.g., increase measurements in the source regions, or considering the spatial correlation of the *a priori* errors in the inversions.

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Tables and Figures

Table 1. Monthly biomass burning and anthropogenic emissions of BC (unit: Gg) in the WUS for August 2006.

Table 2. Monthly anthropogenic and biomass burning emissions of BC (unit: Gg) in the WUS from the adjoint inversions for August 2006 using pseudo observations.

Fig. 1. IMPROVE sites (data available at http://vista.cira.colostate.edu/improve/; solid circles) in the Western United States (WUS). 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). Also shown are terrain heights (color contours).

Fig. 2. Emissions of BC in the WUS for August 2006: (top two rows) biomass burning, (middle two rows) anthropogenic, and (bottom two rows) total emissions. First column: the *a priori*; second column: the optimized inventory; third column: differences between the *a posteriori* and *a priori*; fourth column: scaling factors. For the purpose of clarity, biomass burning emissions are multiplied by 2 in the figures. Retrieval results are from the standard adjoint (Case 1, Table 1) and analytical (Mao et al., 2014) inversions at 2 °×2.5 °. We assume for inversions uncertainties of 500, 50, and 30% for biomass burning BC emissions, anthropogenic BC emissions, and total observation error.

Fig. 3. Normalized sensitivities of the cost function with respect to the BC emissions (left: biomass burning BC; middle: anthropogenic BC; right: total emissions of BC) before and after the inversions at 2 °×2.5 ° (Case 1, Table 1) and 0.5 °×0.667 ° (Case 9, same as Case 1, except at 0.5 °×0.667 °) for August 2006. For the purpose of clarity, sensitivities of the cost function to biomass burning emissions are multiplied by 10 in the figures.

Fig. 4. Emissions of BC in the WUS for August 2006: (top panels) biomass burning, (middle panels) anthropogenic, and (bottom panels) total emissions. Results are from Case 6 (Table 1): (left column) the optimized inventory, (middle column) *a posteriori* - *a priori*, and (right column) the scaling factors. Case 6 is same as Case1, except with only 56 sites used in the inversion. For the purpose of clarity, biomass burning emissions are multiplied by 2 in the figures.

Fig. 5. Monthly anthropogenic and biomass burning emissions of BC in each 2 °×2.5 ° grid box for August 2006 (unit: kg). Solid line is 1:1 and dashed lines are 1:10 (or 10:1).

Fig. 6. Emissions of BC: Case 8 minus Case 1 (Table 1) at 2 °×2.5 ° for August 2006. Case 8 is same as Case 1, except with the *a priori* biomass burning emissions uniformly increased by 2.5 Mg in every model grid box.

Fig. 7. Same as **Fig. 2**, but at 0.5 °×0.667 ° (Case 9, same as Case 1, except at 0.5 °×0.667 °).

Fig. 8. Observed (red line) and GEOS-Chem simulated 24-hour average surface BC concentrations (μg m^{-3}) at six IMPROVE sites for July-September 2006. Results are for 2 °×2.5 ° (solid) and 0.5 °×0.667 ° (dotted line) with the *a priori* (black line) or *a posteriori* emissions from the analytical (blue line) or adjoint (green line) inversions.

Fig. 9. Observed (red line) and GEOS-Chem simulated 24-hour average surface BC concentrations (μg m^{-3}) for July-September 2006, averaged for the IMPROVE sites (**Fig. 1**) in altitudes below 1 km, 1–2 km, 2–3 km, and above 3 km (3 sites). Results are for 2 °×2.5 ° (solid line) and 0.5 °×0.667 ° (dotted line) with the *a priori* (black line) or *a posteriori* emissions from the analytical (blue line) or adjoint (green line) inversions.

Fig. 10. Differences between GEOS-Chem simulated and observed 24-hour average surface BC concentrations at the 69 IMPROVE sites (**Fig. 1**) for August 2006. Model results are from the adjoint inversions at 2 °×2.5 ° (Case 1, Table 1) and 0.5 °×0.667 ° (Case 9, same as Case 1, except at 0.5 °×0.667 °) with the *a priori* or *a posteriori* emissions.

Fig. 11. Frequency distribution of the bias of GEOS-Chem simulated 24-hour average surface BC concentrations for July-September 2006. Results are for 2 °×2.5 ° and 0.5 °×0.667 ° with the *a priori* or *a posteriori* emissions from the analytical or adjoint inversions. Also shown are the mean, median, standard deviation, and fitted Gaussian distribution.

Fig. 12. Taylor diagram and Taylor scores for GEOS-Chem simulations of BC for July-September 2006 at 2 °×2.5 ° (solid circle) and 0.5 °×0.667 ° (open circle) with the *a priori* (red circle) or *a posteriori* emissions from the analytical (blue circle) or adjoint (green circle) inversions. Values are averages for the 69 IMPROVE sites (**Fig. 1**).

Fig. S1. Sensitivities of surface BC mass to the scaling factors of BC emissions as computed using GEOS-Chem adjoint and the finite difference approximation (Eq. 1, unit: kg / grid box). Results are from 1-week simulations for biomass burning (top panel) and anthropogenic emissions (bottom panel) for August 2006. Solid lines are regression lines.

Fig. S2. (top) Normalized *a posteriori* cost function $J(\boldsymbol{X}_n)/J(\boldsymbol{X}_0)$ as a function of the regularization parameter γ_r (Eq. 3) for August 2006. (bottom) Reduction in the normalized cost function $J(\boldsymbol{X}_i)/J(\boldsymbol{X}_0)$ at 2 °×2.5 ° and 0.5 °×0.667 ° for August 2006.

Table 1. Monthly biomass burning and anthropogenic emissions of BC (unit: Gg) in the WUS for August 2006.

			Inversion configurations						Emissions[a] (Gg (10^9 g))					
									Biomass burning				Anth.	Total
			Model Resolution	Site num.	S_aBB (%)	S_aAnth (%)	S_Σ (%)	*A priori* BB.	RM	CSW	PNW	Total		
A priori									1.3	0.5	1.0	2.8	5.8	8.6
A posteriori	**Analytical**		2°×2.5°	69	500	50	30	2.8	7.6	1.4	0.9	9.9 (3.5[b])	2.8 (0.5)	12.7 (1.5)
			0.5°×0.667°	69	500	50	30	2.8	4.1	0.1	1.9	6.1 (2.7)	3.1 (0.5)	9.1 (1.1)
	Adjoint	**Case 1**	2°×2.5°	69	500	50	30	2.8	3.0	1.4	1.5	5.9 (2.1)	11.8 (2.0)	17.7 (2.1)
		Case 2	2°×2.5°	69	300	50	30	2.8	2.9	1.3	1.4	5.6 (2.0)	11.6 (2.0)	17.2 (2.0)
		Case 3	2°×2.5°	69	500	30	30	2.8	3.2	1.4	1.6	6.2 (2.2)	11.4 (2.0)	17.6 (2.0)
		Case 4	2°×2.5°	69	500	200	30	2.8	3.3	1.5	1.7	6.5 (2.3)	12.8 (2.2)	19.3 (2.2)
		Case 5	2°×2.5°	69	500	50	100	2.8	2.4	1.2	1.0	4.6 (1.7)	10.3 (1.8)	14.9 (1.7)
		Case 6	2°×2.5°	56	500	50	30	2.8	2.8	1.3	1.4	5.5 (2.0)	11.1 (1.9)	16.6 (1.9)
		Case 7	2°×2.5°	69	200	200	30	2.8	3.1	1.4	1.5	6.0 (2.2)	12.5 (2.2)	18.5 (2.2)
		Case 8	2°×2.5°	69	500	50	30	3.0[c]	3.1	1.4	1.5	6.0 (2.1)	11.6 (2.0)	17.6 (2.0)
		Case 9	0.5°×0.667°	69	500	50	30	2.8	2.2	0.8	2.3	5.3 (1.9)	8.6 (1.5)	13.9 (1.6)

[a]See Fig. 1 in Mao et al. (2014) for the geographical definitions of the BC source regions.
[b]Scaling factors are in parentheses.
[c]The *a priori* biomass burning emissions uniformly increased by 2.5 Mg in every model grid box.

Table 2. Monthly anthropogenic and biomass burning emissions of BC (unit: Gg) in the WUS from the adjoint inversions for August 2006 using pseudo observations.

		Inversion configurations			Emissions (Gg (10^9 g))			$J(x)$ reduction[c] (%)
		Pseudo observations	S_aANTH (%)	S_aBB (%)	Anth.	BB	Δ Emissions[b]	
A posteriori	**Pseudo 1**	every surface grid box	50	500	7.7 (1.3[a])	6.5 (2.3)	5.6 (2.0)	95
	Pseudo 2	every grid box in the lowest 15 layers	50	500	7.8 (1.3)	6.4 (2.3)	5.6 (2.0)	97
	Pseudo 3	every surface grid box	10	500	7.1 (1.2)	6.9 (2.5)	5.4 (1.9)	95
	Pseudo 4	every surface grid box	0	500	5.8 (1.0)	8.2 (2.9)	5.4 (1.9)	99
	Pseudo 5	every surface grid box	50	0	10.0 (1.7)	2.8 (1.0)	4.2 (1.5)	55
	Pseudo 6	randomly in 75% of the surface grid boxes	50	500	7.8 (1.3)	6.2 (2.2)	5.4 (1.9)	94
	Pseudo 7	randomly in 50% of the surface grid boxes	50	500	8.0 (1.4)	5.9 (2.1)	5.3 (1.9)	93
	Pseudo 8	randomly in 25% of the surface grid boxes	50	500	7.5 (1.3)	5.3 (1.9)	4.2 (1.5)	96
	Pseudo 9	in the surface grid boxes with fire counts (> 5)	50	500	7.6 (1.3)	6.2 (2.2)	5.2 (1.9)	95
'Ghost' emissions[d]					5.8 (1.0)	8.4 (3.0)	5.6 (2.0)	
A priori					5.8	2.8		

[a]Scaling factors in parentheses.
[b]The increase of total emissions relative to the *a priori* biomass burning emissions (ratio of the total emissions change over the *a priori* biomass burning emissions in parentheses).
[c]The reduction of the *a posteriori* cost function relative to the *a priori*.
[d]Emissions (with anthropogenic emissions unchanged but biomass burning emissions tripled) used to generate the pseudo observations for Pseudos 1-9.

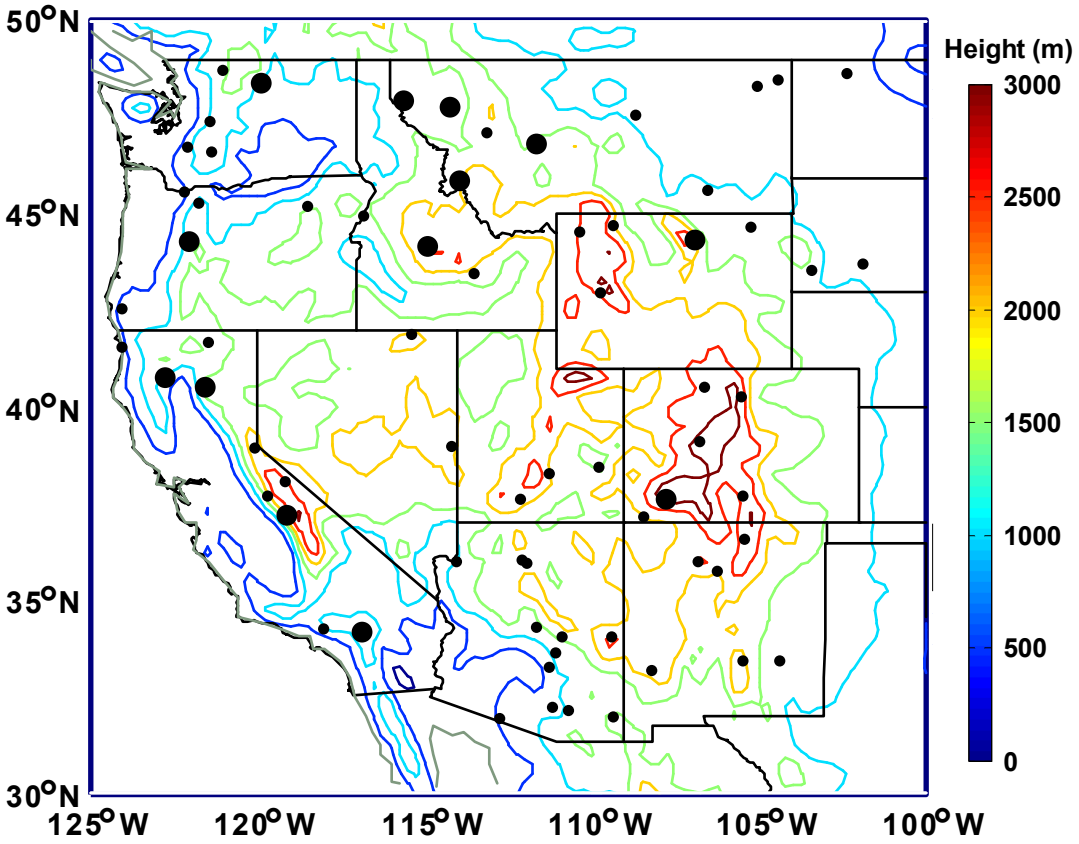
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Fig. 1

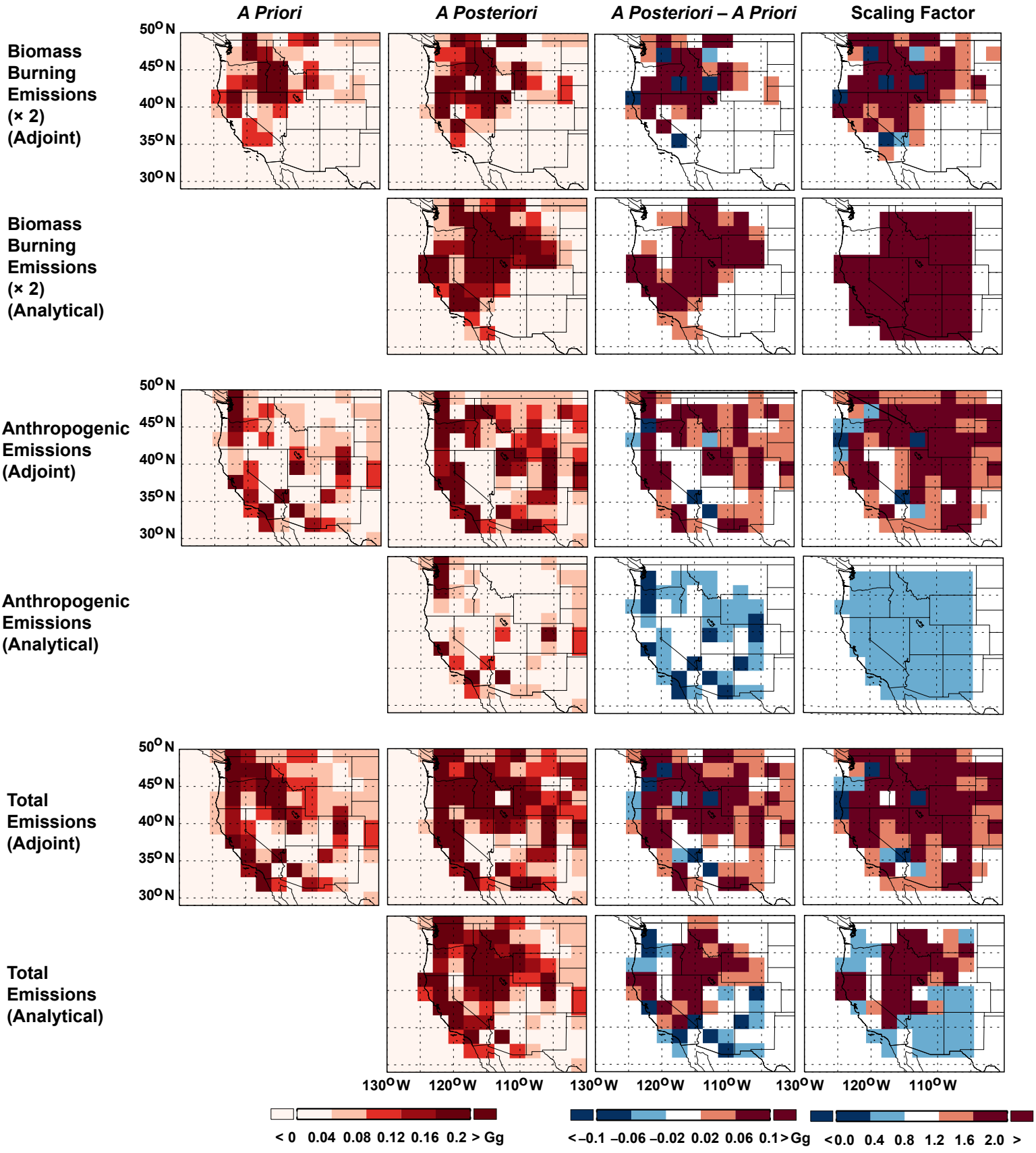
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Fig. 2.

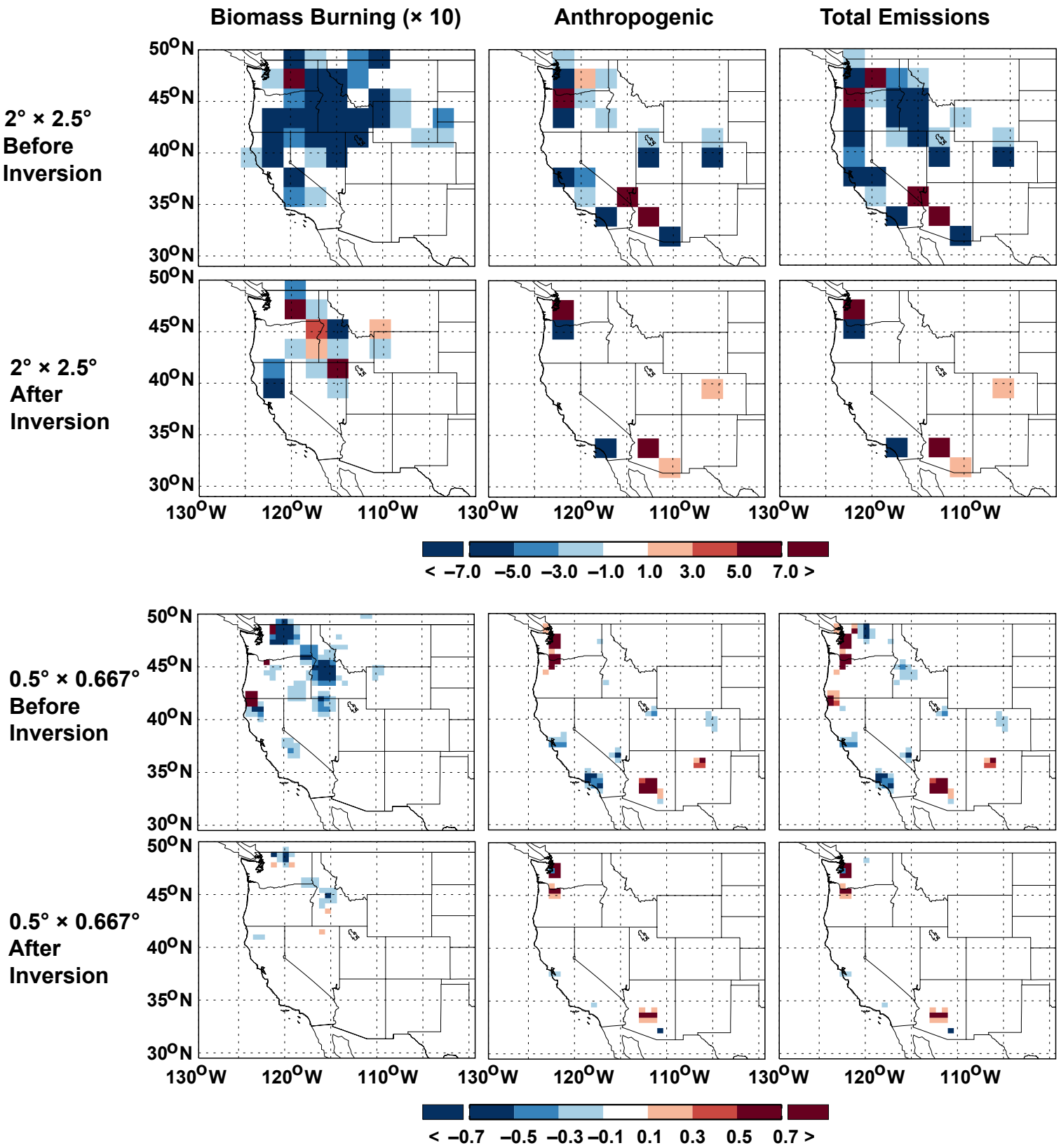
Biomass Burning (× 10)
Anthropogenic
Total Emissions
2° × 2.5° Before Inversion
2° × 2.5° After Inversion
0.5° × 0.667° Before Inversion
0.5° × 0.667° After Inversion
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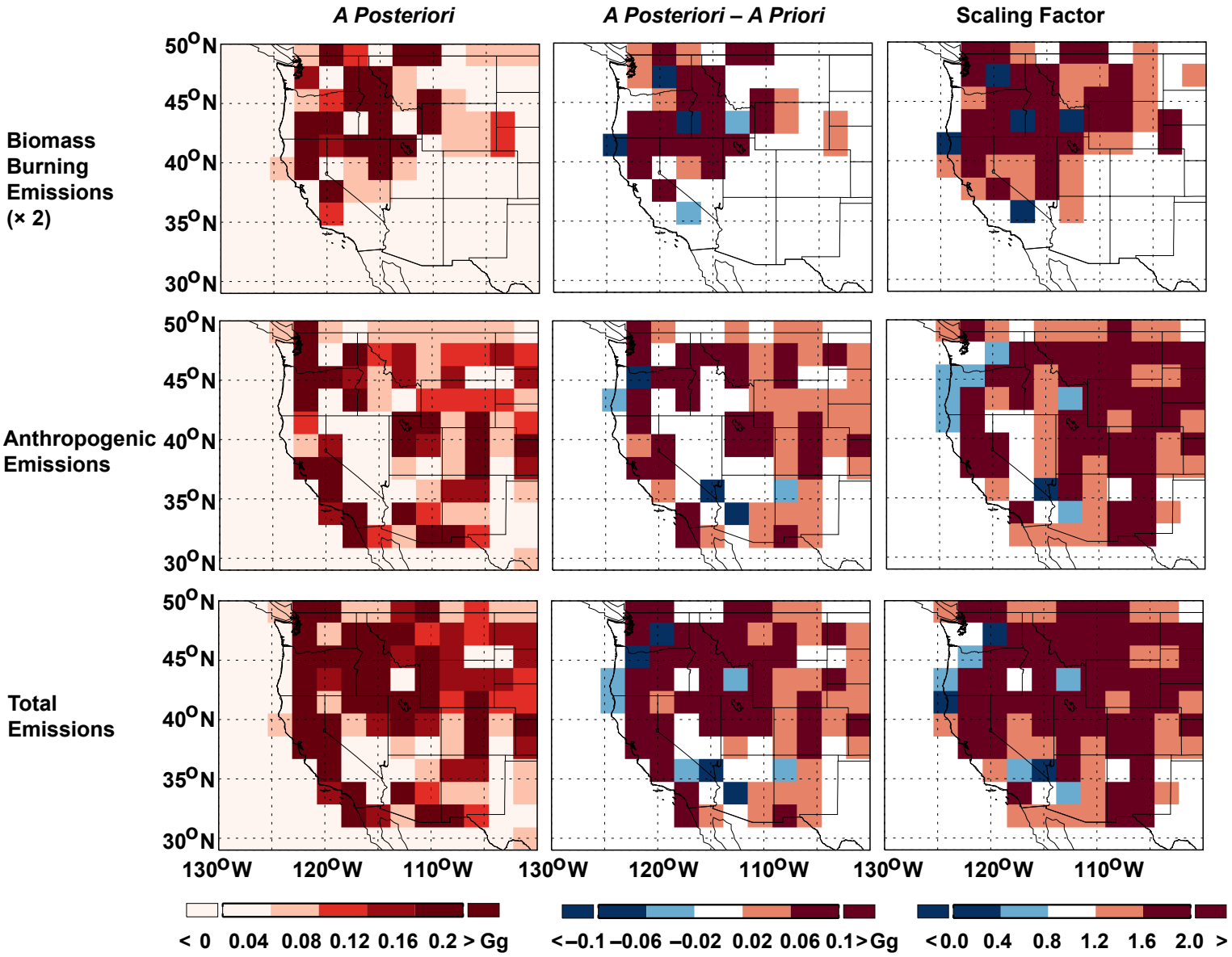
Fig. 3.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<0.0 0.4 0.8 1.2 1.6 2.0 >

Fig. 4.

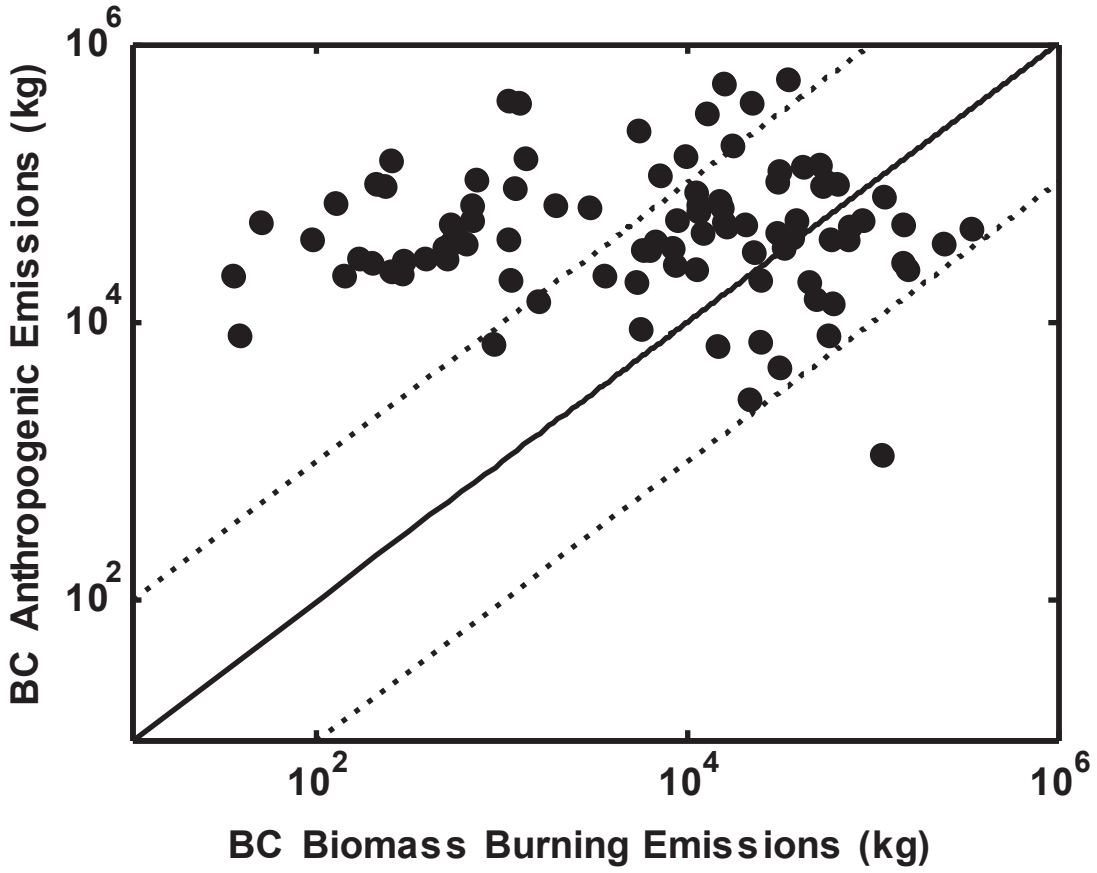

BC Anthropogenic Emissions (kg)
10^6
10^4
10^2
10^2
10^4
10^6
BC Biomass Burning Emissions (kg)

Fig. 5

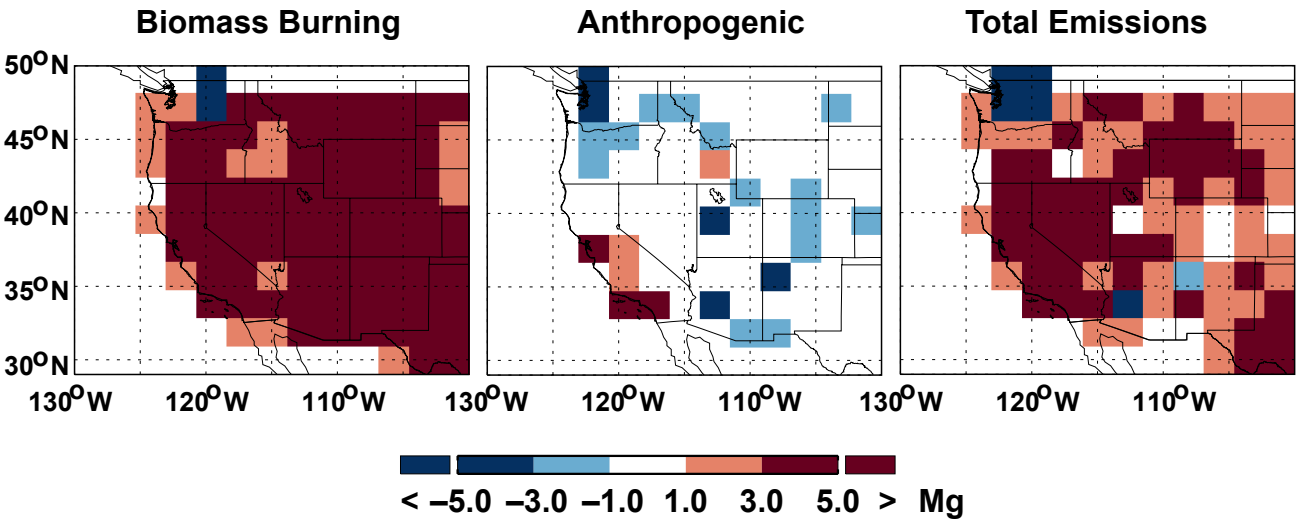

Biomass Burning
Anthropogenic
Total Emissions
50°N
45°N
40°N
35°N
30°N
130°W
120°W
110°W
< –5.0 –3.0 –1.0 1.0 3.0 5.0 > Mg

Fig. 6.

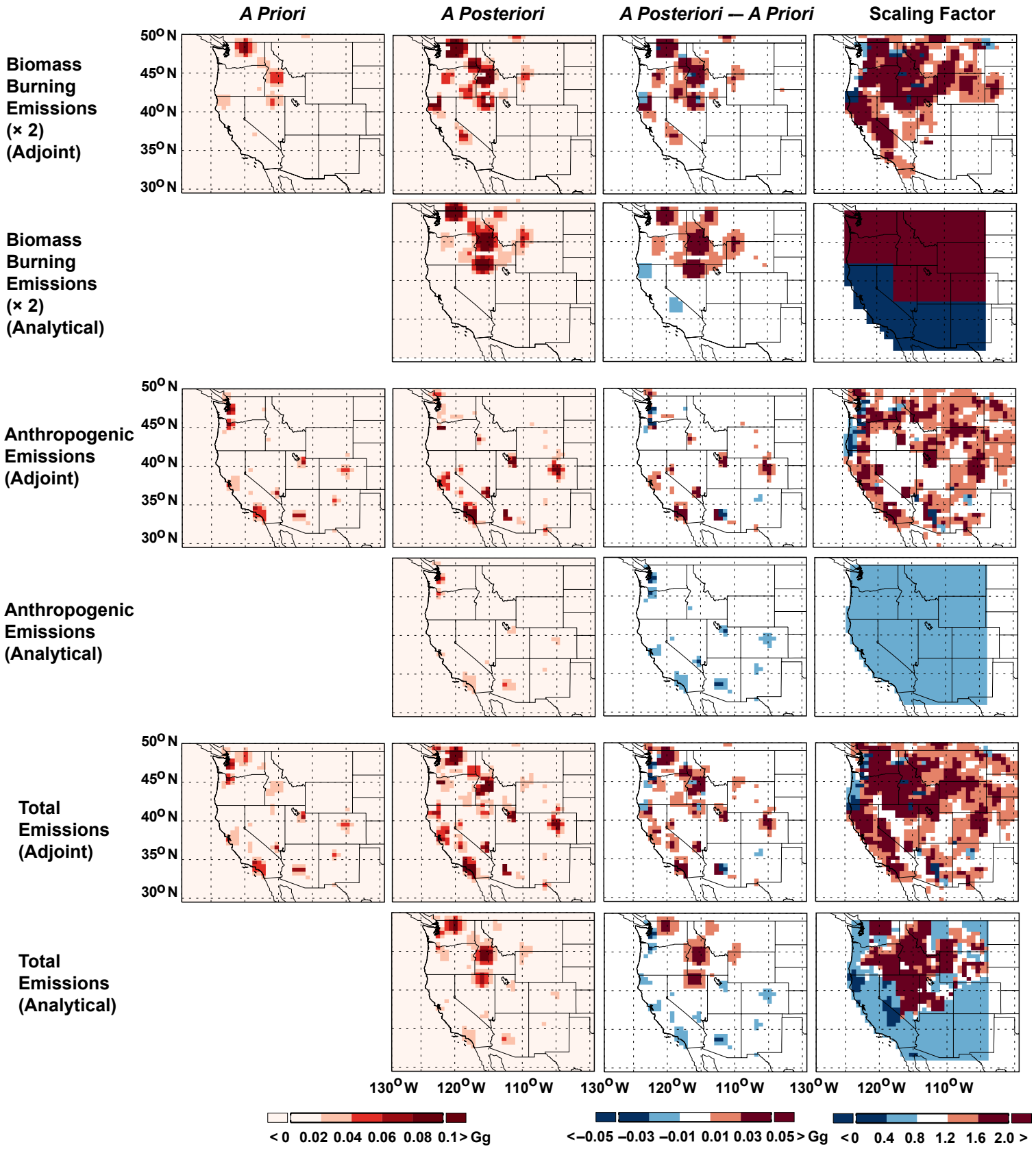

A Priori
A Posteriori
A Posteriori — A Priori
Scaling Factor
Biomass Burning Emissions (× 2) (Adjoint)
Biomass Burning Emissions (× 2) (Analytical)
Anthropogenic Emissions (Adjoint)
Anthropogenic Emissions (Analytical)
Total Emissions (Adjoint)
Total Emissions (Analytical)
50° N
45° N
40° N
35° N
30° N
130° W
120° W
110° W
< 0 0.02 0.04 0.06 0.08 0.1 > Gg
< –0.05 –0.03 –0.01 0.01 0.03 0.05 > Gg
< 0 0.4 0.8 1.2 1.6 2.0 >

Fig. 7.

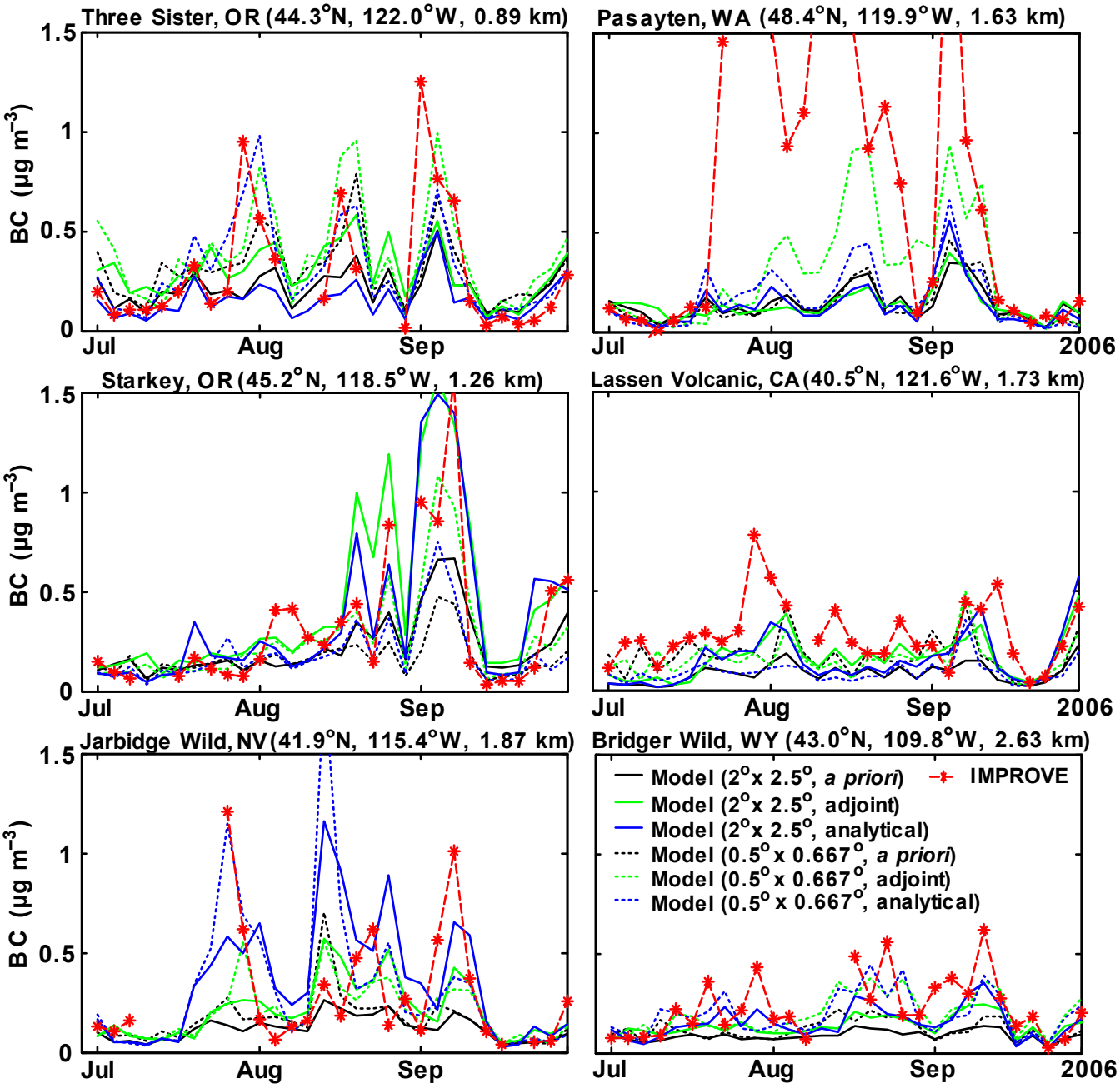

Three Sister, OR (44.3°N, 122.0°W, 0.89 km)
Pasayten, WA (48.4°N, 119.9°W, 1.63 km)
Starkey, OR (45.2°N, 118.5°W, 1.26 km)
Lassen Volcanic, CA (40.5°N, 121.6°W, 1.73 km)
Jarbidge Wild, NV (41.9°N, 115.4°W, 1.87 km)
Bridger Wild, WY (43.0°N, 109.8°W, 2.63 km)
BC (μg m−3)
0
0.5
1
1.5
Jul
Aug
Sep
2006
Model (2°x 2.5°, a priori)
Model (2°x 2.5°, adjoint)
Model (2°x 2.5°, analytical)
Model (0.5°x 0.667°, a priori)
Model (0.5°x 0.667°, adjoint)
Model (0.5° x 0.667°, analytical)
IMPROVE

Fig. 8

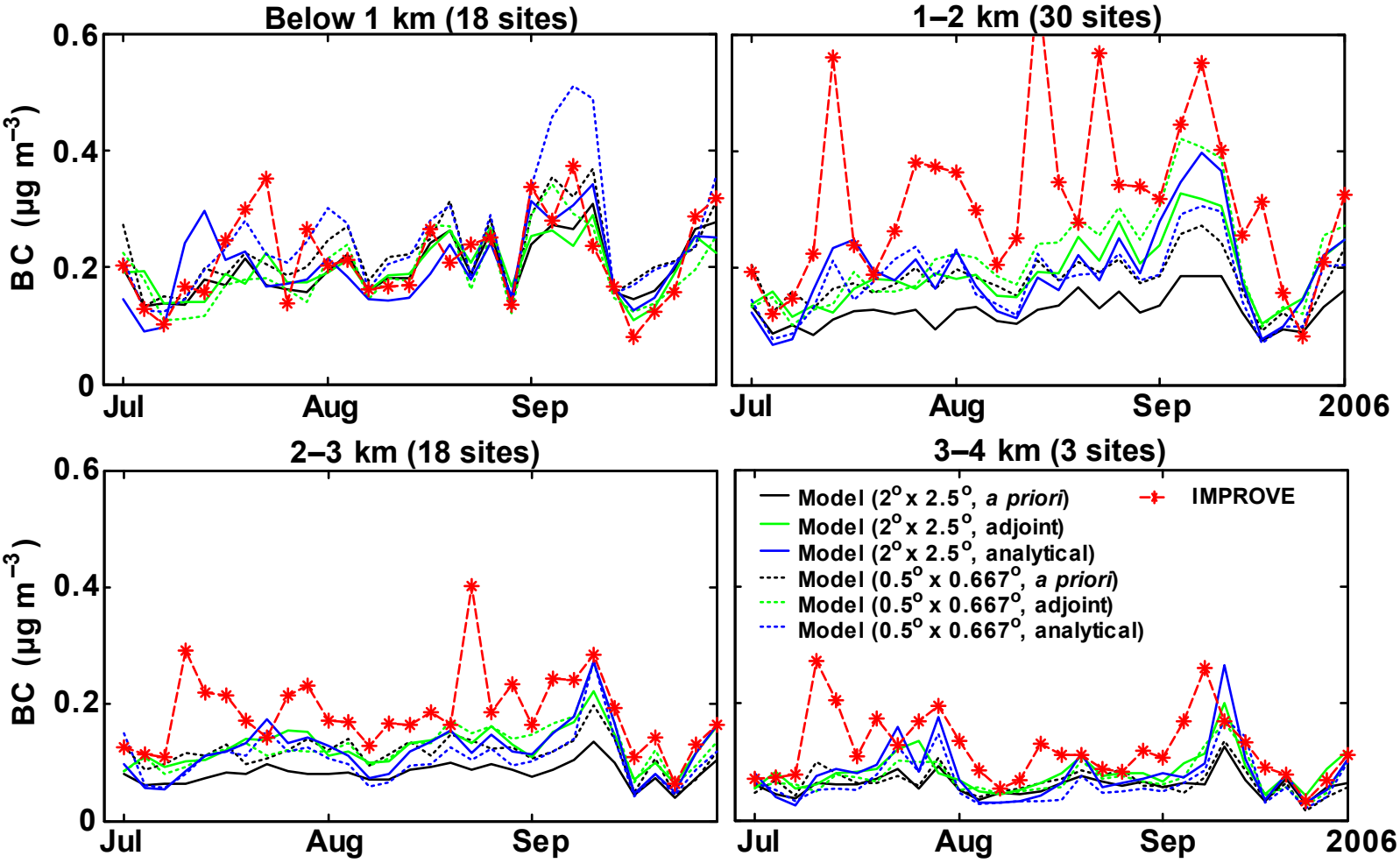

Below 1 km (18 sites)
1–2 km (30 sites)
2–3 km (18 sites)
3–4 km (3 sites)
BC (μg m−3)
0
0.2
0.4
0.6
Jul
Aug
Sep
2006
Model (2° x 2.5°, a priori)
Model (2° x 2.5°, adjoint)
Model (2° x 2.5°, analytical)
Model (0.5° x 0.667°, a priori)
Model (0.5° x 0.667°, adjoint)
Model (0.5° x 0.667°, analytical)
IMPROVE

Fig. 9

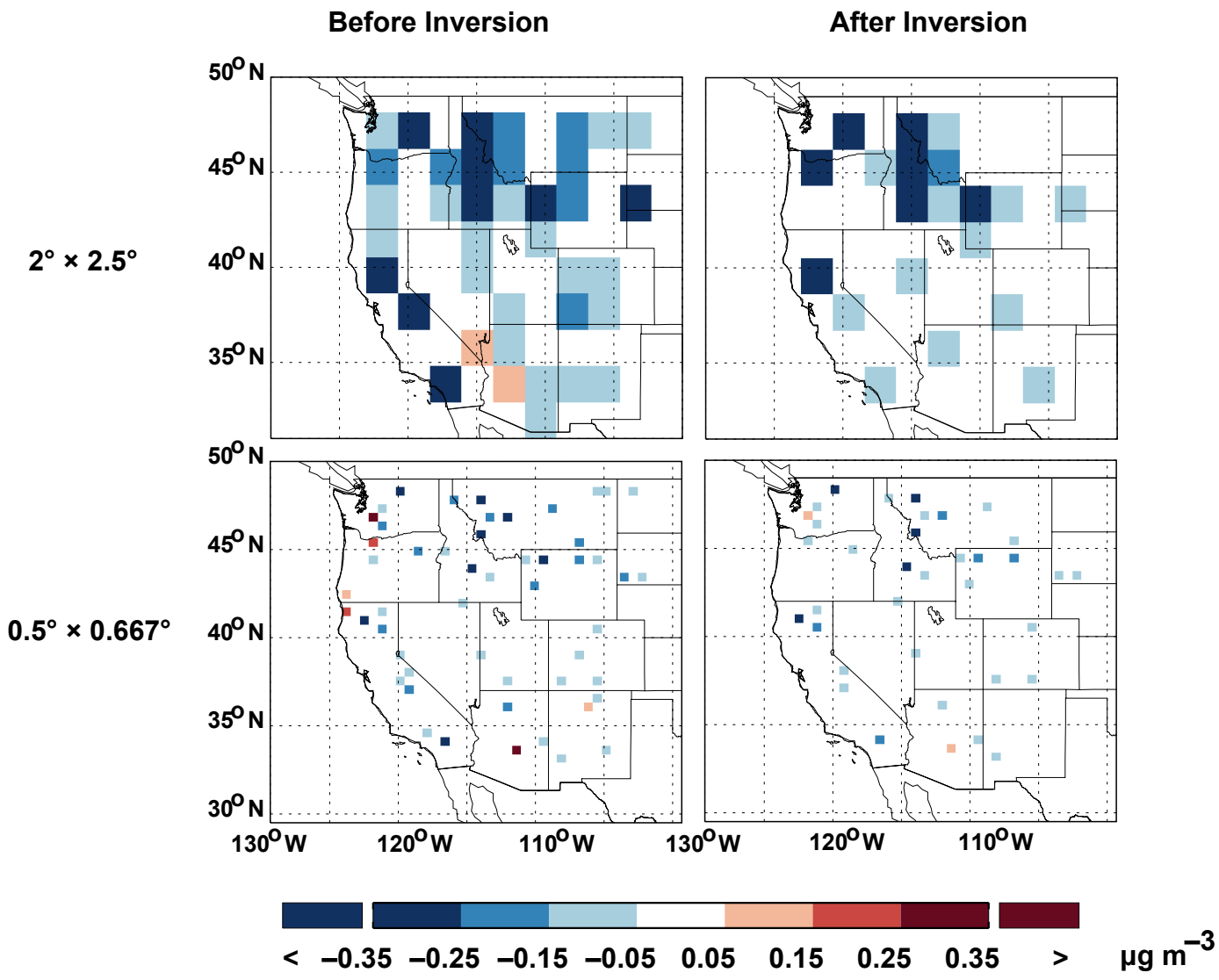

Before Inversion
After Inversion
2° × 2.5°
0.5° × 0.667°
50°N
45°N
40°N
35°N
30°N
130°W
120°W
110°W
< –0.35 –0.25 –0.15 –0.05 0.05 0.15 0.25 0.35 >
μg m–3

Fig. 10.

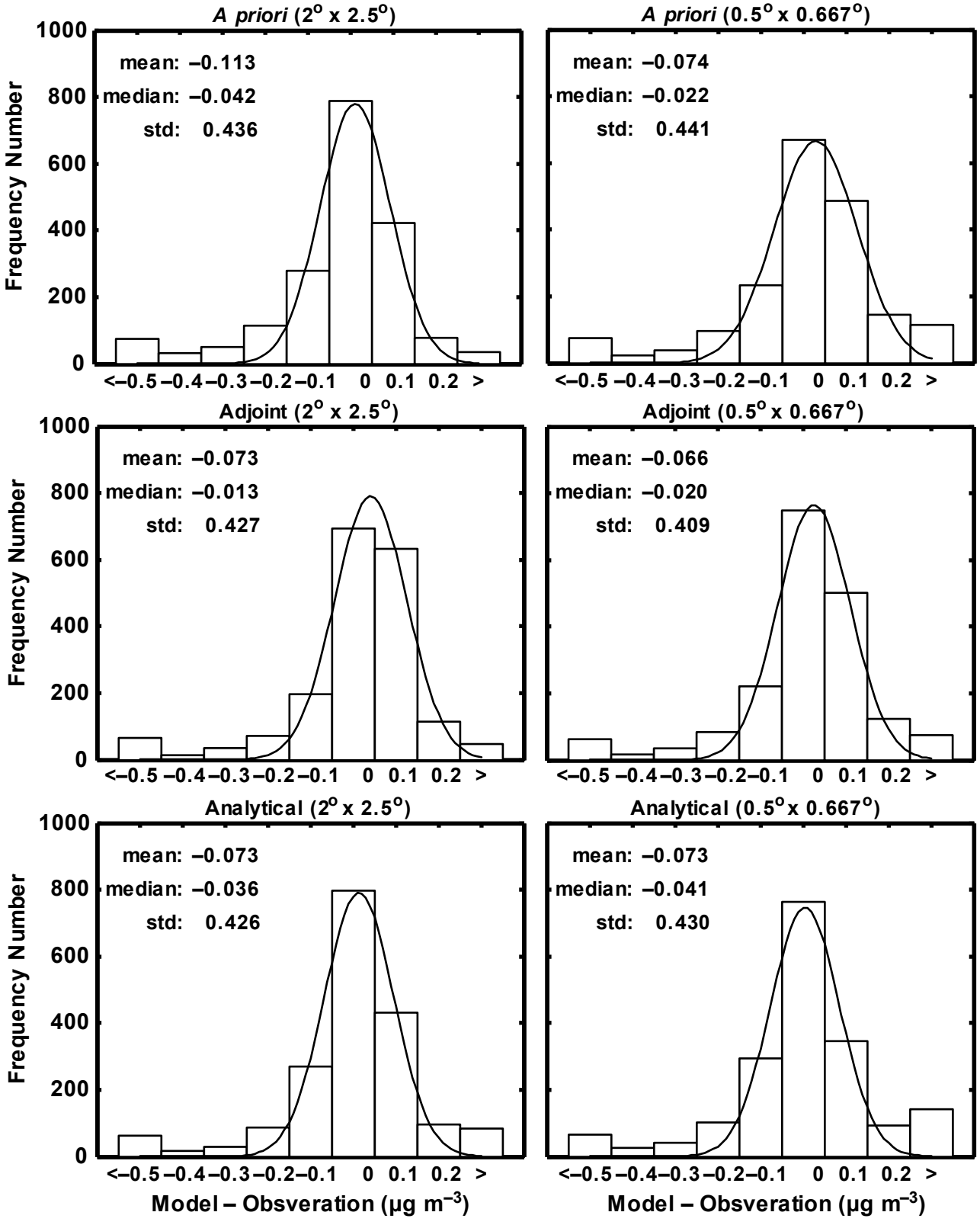

A priori (2° x 2.5°)
mean: –0.113
median: –0.042
std: 0.436
A priori (0.5° x 0.667°)
mean: –0.074
median: –0.022
std: 0.441
Adjoint (2° x 2.5°)
mean: –0.073
median: –0.013
std: 0.427
Adjoint (0.5° x 0.667°)
mean: –0.066
median: –0.020
std: 0.409
Analytical (2° x 2.5°)
mean: –0.073
median: –0.036
std: 0.426
Analytical (0.5° x 0.667°)
mean: –0.073
median: –0.041
std: 0.430
Frequency Number
0
200
400
600
800
1000
<–0.5 –0.4 –0.3 –0.2 –0.1 0 0.1 0.2 >
Model – Obsveration (μg m–3)

Fig. 11

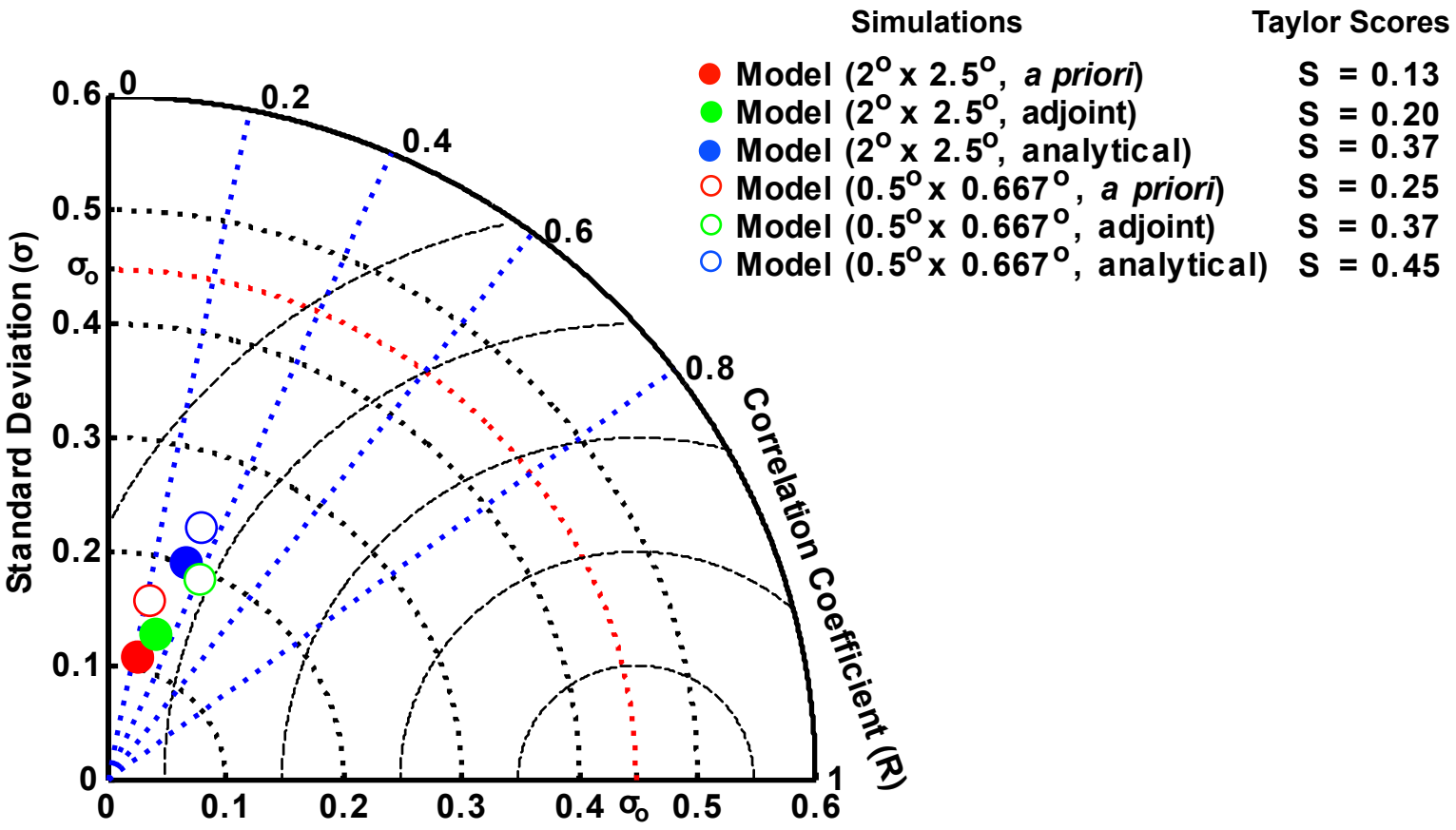

Simulations
Taylor Scores
Model (2° x 2.5°, a priori) S = 0.13
Model (2° x 2.5°, adjoint) S = 0.20
Model (2° x 2.5°, analytical) S = 0.37
Model (0.5° x 0.667°, a priori) S = 0.25
Model (0.5° x 0.667°, adjoint) S = 0.37
Model (0.5° x 0.667°, analytical) S = 0.45
Standard Deviation (σ)
Correlation Coefficient (R)
σo
0
0.2
0.4
0.6
0.8
1
0.1
0.3
0.5

Fig. 12

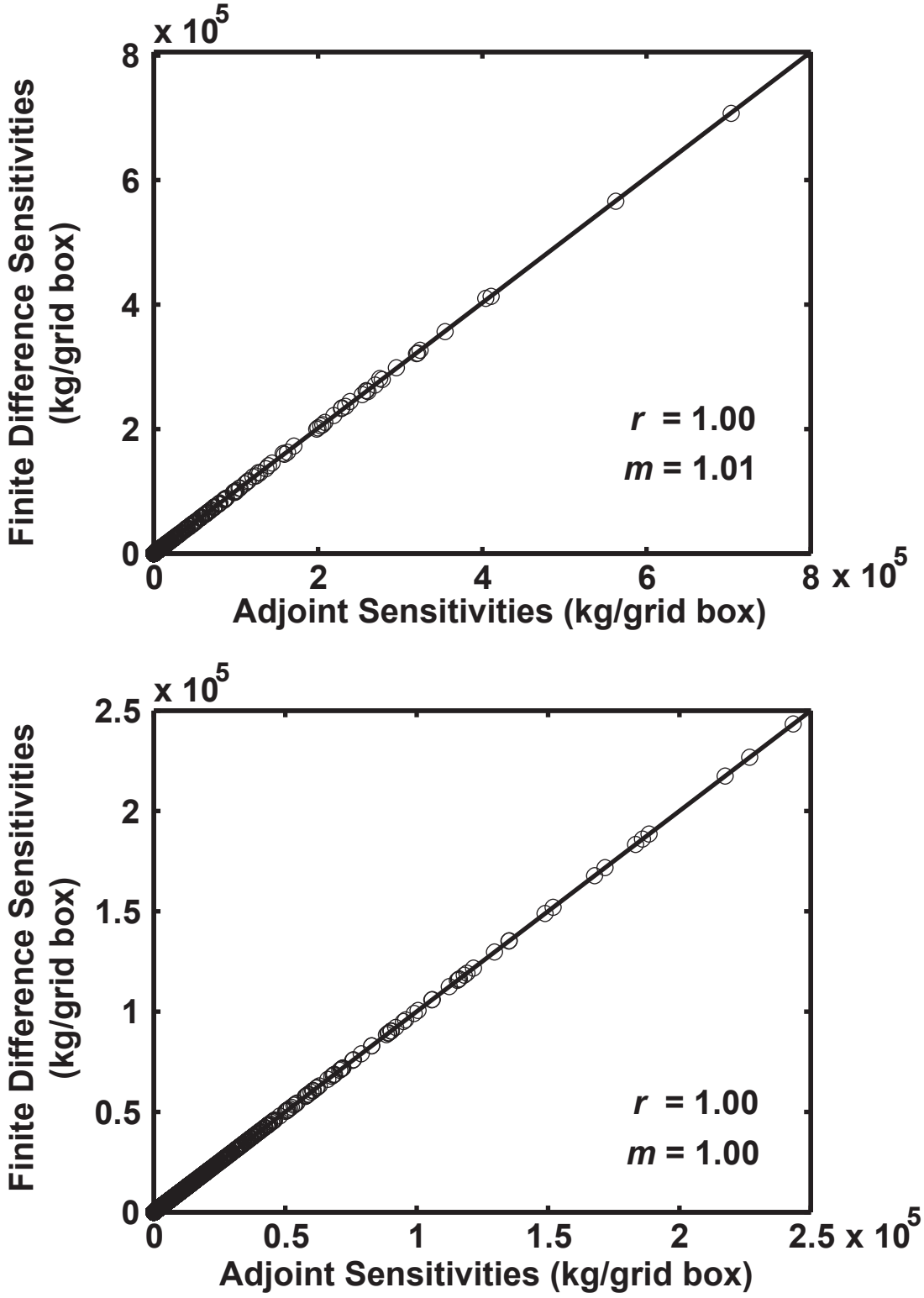
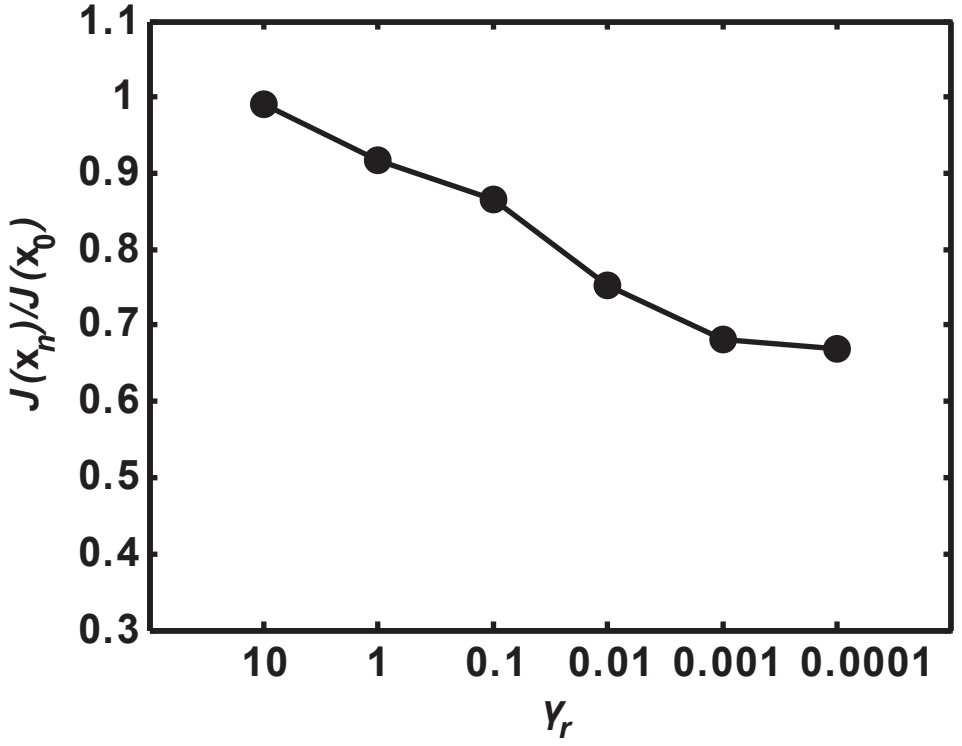

x 10^5
8
6
4
2
0
Finite Difference Sensitivities (kg/grid box)
r = 1.00
m = 1.01
0
2
4
6
8 x 10^5
Adjoint Sensitivities (kg/grid box)
x 10^5
2.5
2
1.5
1
0.5
0
Finite Difference Sensitivities (kg/grid box)
r = 1.00
m = 1.00
0
0.5
1
1.5
2
2.5 x 10^5
Adjoint Sensitivities (kg/grid box)

Fig. S1


1.1
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
$J(x_n)/J(x_0)$
10
1
0.1
0.01
0.001
0.0001
γ_r

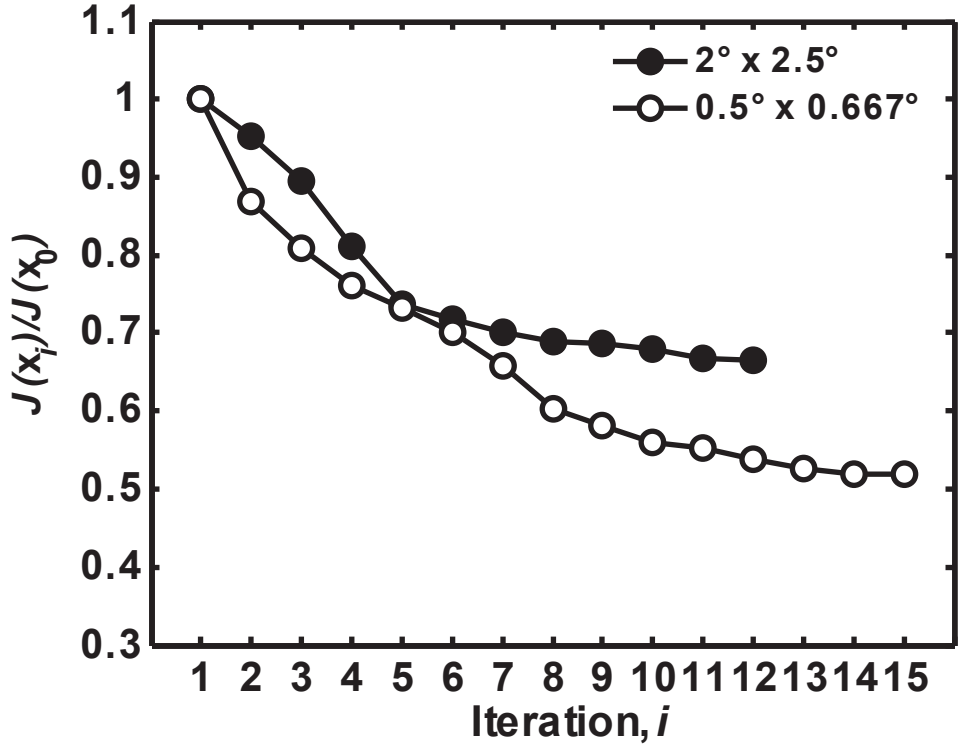

1.1
1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
$J(x_i)/J(x_0)$
2° x 2.5°
0.5° x 0.667°
1 2 3 4 5 6 7 8 9 10 11 12 13 14 15
Iteration, i

Fig. S2