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Top-down estimates of biomass burning emissions of black carbon in the Western United States

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Abstract

We estimate biomass burning emissions of black carbon (BC) in the western United States (WUS) for May–October 2006 by inverting surface BC concentrations from the Interagency Monitoring of PROtected Visual Environment (IMPROVE) network using
⁵ a global chemical transport model. We first improve the spatiotemporal distributions of the BC emissions from the Global Fire Emissions Database (GFEDv2) using 8-day active fire counts from the Moderate Resolution Imaging Spectroradiometer (MODIS) from a 3 yr period (2005–2007). The resulting emissions are then used as the a priori for the inversion analyses. The adjustment primarily shifts emissions from late to early and middle summer (33 % decrease in September-October and 56 % increase in June–August). The adjusted emissions lead to non-negligible increases in the simulated surface BC concentrations in early and middle summer at sites below 2 km. We conduct analytical inversions at both 2° × 2.5° and 0.5° × 0.667° (nested over North America) horizontal resolutions. Simulated surface BC concentrations with the a poste-

- ¹⁵ riori emissions capture the observed major fire episodes at many sites and substantial enhancements at the 1–2 and 2–3 km altitude ranges. The a posteriori emissions lead to substantial bias reductions in the simulated surface BC concentrations (~ 50 % on average) at both resolutions and significant increases in the Taylor skill scores (86 % at 2° × 2.5° and 132 % at 0.5° × 0.667°). We find that the inversion is rather sensitive to the medical matching. The emergencies beginning emissions is rather sensitive to
- the model resolution. The a posteriori biomass burning emissions increase by factors of 4.7 from the inversion at 2° × 2.5° and 2.8 at 0.5° × 0.667°, while as the a posteriori anthropogenic emissions decrease by 48% and 36%, respectively, relative to their corresponding a priori emissions. The two a posteriori estimates differ largest in biomass burning emissions in California and the Southwest (a factor of 5.9) and in the Pacific Northwest (a factor of 2).





1 Introduction

Black carbon (BC), as a component of fine particulate matter, has deleterious effects on human health (e.g., Anenberg et al., 2011, 2012; Smith et al., 2009). BC is also known as one of the only two agents to cause both degraded air quality (e.g., Anenberg et al., 2012).

- ⁵ al., 2011, 2012) and warming due to its strong absorption of solar radiation (e.g., Ramanathan and Carmichael, 2008; Horvath, 1993). BC thus has considerable impacts on global climate (Fuglestvedt et al., 2010; Flanner et al., 2007, 2009; Shindell et al., 2008; Levy et al., 2008; Reddy et al., 2007; IPCC, 2007; Hansen and Nazarenko, 2004; Jacobson, 2001, 2004). BC deposited on snow and ice can significantly decrease the
- ¹⁰ surface albedo (Warren and Wiscombe, 1980) and quicken surface melt (e.g. Flanner et al., 2007; Hansen and Nazarenko, 2004; Zwally et al., 2002). Because of its shorter lifetime relative to long lived greenhouse gases such as carbon dioxide, BC shows a much stronger regional warming effect and its reduction may provide an efficient solution to mitigate near-term climate change and to improve air quality and human health
- ¹⁵ simultaneously (Bond et al., 2013; Shindell et al., 2012; Anenberg et al., 2011, 2012; Kopp and Mauzerall, 2010; Ramana et al., 2010; Jacobson, 2002, 2010; Ramanathan and Carmichael, 2008; Bond and Sun, 2005; Hansen et al., 2005).

The uncertainty in current BC emission estimates ranges from at least ± 50 % on global scales to a factor of 2–5 on regional scales (Ramanathan and Carmichael, 2008;

- Streets et al., 2001, 2003). Mao et al. (2011) estimated that version 2 of the Global Fire Emissions Database (GFEDv2) biomass burning emissions (Randerson et al., 2007; data available at http://daac.ornl.gov/VEGETATION/guides/global_fire_emissions_v2. 1.html) of BC were biased low by a factor of two in the western United States (WUS) for July–October 2006 using the GEOS-Chem model. The discrepancies in the timing
- ²⁵ of the simulated and observed surface BC enhancement suggest that the uncertainties of biomass burning emissions of BC are not only in the absolute magnitudes of fire emissions but also likely in the timing and location of fires. Even though fire emission inventories have been improved considerably in recent years, large uncertainties re-





main in the temporal variations and spatial distributions of fire emissions, particularly from burned area and fuel load (Langmann et al., 2009). Small fires are likely a major source of uncertainty in the estimates of biomass burning emissions of BC (Randerson et al., 2012). For instance, small fires can lead to high relative errors of 50–100% in the burned area estimates (Giglio et al., 2006, 2010). Additionally, the lack of detection of agricultural burnings may be another large uncertainty (Randerson et al., 2012; van der Werf et al., 2010; McCarty et al., 2009; Roy and Boschetti, 2009; Korontzi et al., 2006).

Understanding the distribution of a chemical species in the atmosphere depends on the information of the emissions. The bottom-up emission estimates generally rely on emission factors using socioeconomic, energy, land use or environmental data (Heald et al., 2004). In recent years, there has been an increasing emphasis on the use of inverse methods to characterize the temporal and spatial variability of emissions. Inverse modeling is a standard tool for estimating top-down emissions from the combi-15 nation of observations of atmospheric trace species and bottom-up constraints using a forward model F(*x*). Considering the general problem of estimating a set of emissions

- (assembled in a state vector x), given a set of observed atmospheric concentrations (observation vector y) with error ε , we relate x to y by the following relation (Rodgers, 2000):
- $y = F(x) + \varepsilon$

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Based on Bayes' theorem and the assumption of Gaussian error distributions (Rodgers, 2000), the optimal or Maximum A Posteriori (MAP) solution for x by given y, is equivalent to find the minimum in an error-weighted least squares scalar cost function J(x) (that is, to solve $\nabla_x J(x) = 0$). The cost function describes the error-weighted mismatch between the observed concentrations, y, and those simulated with the forward model, F(x), as well as the error-weighted mismatch between the true state and the a priori estimate x_a (Kopacz et al., 2009). Most of the inverse modeling literature for atmospheric composition has used an analytical solution for $\nabla_x J(x) = 0$. Applications of analytical in-



(1)



verse methods so far have used observational constraints from surface stations. These studies include, for example, Bergamaschi et al. (2000) and Kasibhatla et al. (2002). More recent studies have exploited the high density of observational coverage from aircrafts (Palmer et al., 2003, 2006; Heald et al., 2004) and satellites (Jiang et al., 2011; Jones et al., 2009; Kopacz et al., 2009; Arellano et al., 2006, 2004; Heald et al., 2004).

The goal of the present study is to improve our understanding of sources of BC in the WUS mountain ranges, with a particular focus on BC emissions from biomass burning during May–October 2006, broadly encompassing the fire season in the region. We first improve the spatial distributions and seasonal and interannual variations of the BC emissions from the GFEDv2 using the Moderate Resolution Imaging Spectroradiometer (MODIS) 8-day active fire counts (0.5° × 0.5°, available at ftp: //fuoco.geog.umd.edu) from a 3 yr period (2005–2007). We then apply linear analytical inversions to improve estimates of monthly biomass burning emissions of BC constrained by surface BC concentration measurements from the Interagency Monitor-

- ¹⁵ ing of PROtected Visual Environment (IMPROVE) network (Malm et al., 1994; data available at http://vista.cira.colostate.edu/improve/). We use as the forward model the GEOS-Chem global chemical transport model (CTM) (Bey et al., 2001) at both 2° × 2.5° (globally) and 0.5° × 0.667° (nested over North America) horizontal resolutions. We briefly describe the model in Sect. 2. We then discuss in Sect. 3 improvements to the
 ²⁰ spatiotemporal distributions of biomass burning emissions of BC. In Sect. 4, we de-
- scribe the analytical inversion method. The inversion results are presented in Sect. 5. Summary and conclusions are given in Sect. 6.

2 Model description and simulations

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We use a global 3-D chemical transport model (GEOS-Chem) and its North American nested model to conduct offline carbonaceous aerosols simulations. The GEOS-Chem model is driven by assimilated meteorological observations from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Of-



fice (GMAO) (Bey et al., 2001). We here use GEOS-Chem version 8-03-02 (available at http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_v8-03-02) driven by the GMAO GEOS-5 meteorological data. GEOS-5 meteorological data have a temporal resolution of 6 h (3 h for surface variables and mixing depths) and horizontal resolutions of 2° (latitude) × 2.5° (longitude) globally and 0.5° (latitude) × 0.667° (longitude) over North America, with 47 hybrid eta levels in the vertical column that extending from the surface to 0.01 hPa. The lowest 2 km is resolved by 14 layers centered at approximately 70, 200, 330, 470, 600, 740, 880, 1000, 1160, 1300, 1440, 1600, 2000 m above sea level. The nested GEOS-Chem model simulation over North America was first described by Wang et al. (2004). To provide boundary conditions for the nested model, the global assimilations were provided every three hours with the spatial resolution of 2° (latitude) × 2.5° (longitude). The high-resolution, nested-grid simulation employed the same meteorology, dynamics, and chemistry as the global GEOS-Chem model.

thus allows for consistent propagation of features from the global domain to the nested domain. A detailed description of this one-way nesting in the GEOS-Chem model was

given by Chen et al. (2009). We conducted nested model simulations over the domain

GEOS-Chem has been reported previously by Park et al. (2003) with many updates. Eighty percent of BC and 50% of OC emitted from primary sources are assumed to

be hydrophobic and hydrophobic aerosols become hydrophilic with an e-folding time of 1.2 days (Park et al., 2003; Chin et al., 2002; Cooke et al., 1999). Simulation of aerosol wet and dry deposition follows Liu et al. (2001). Wet deposition includes contributions

from scavenging in convective updrafts, rainout from convective anvils, and rainout and

washout from large-scale precipitation. Dry deposition of aerosols uses a resistance-in-

series model (Walcek et al., 1986) dependent on local surface type and meteorological conditions. Global fossil fuel and biofuel emissions of BC are based upon Bond et

al. (2007). For biomass burning, we use GFEDv2 emissions with a temporal resolution

of eight days (Randerson et al., 2007; van der Werf et al., 2006).

The simulation of carbonaceous aerosols, BC and organic carbon (OC), in the

40–140° W longitude and 10–70° N latitude (cf. Fig. 1 in Wang et al., 2004).

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For the present study, we conducted GEOS-Chem "offline" carbonaceous aerosols simulations (e.g., Mao et al., 2011; Park et al., 2003) for 2006. The first three months were used for initialization and we focused our analysis on the results for May through October. We mainly used the 2° × 2.5° horizontal resolution model for our simulations unless stated otherwise. The 0.5° × 0.667° horizontal resolution model was also included in the sensitivity simulation. Detailed discussions and justifications for the model simulations are provided in the following sections where appropriate. Model results are sampled at the corresponding locations of the IMPROVE sites. Figure 1 shows 69 IMPROVE sites in the WUS used in this study. IMPROVE observations are 24-hour averages sampled every three days and we sample the model accordingly. As pointed out in previous studies (Mao et al., 2011; Fairlie et al., 2007), comparing localized observations such as the IMPROVE data with model results that are representative of a much larger area is inherently problematic. The comparison is further complicated by the fact that many of the IMPROVE sites are mountainous sites and the associated upslope flow is difficult to represent in a coarse-resolution model like the GEOS-Chem 15 model used here. Also shown in Fig. 1 are the three biomass burning regions used to

define the state vector of the inversion analyses. Selections of these three regions are discussed in Sect. 4.

3 Spatiotemporal distributions of biomass burning BC emissions

- ²⁰ The GFED inventory of biomass burning emissions was derived using satellite observations including active fire counts and burned areas in conjunction with a biogeochemical model. Burned area was derived using monthly 0.5° × 0.5° active fire and 500 m burned area datasets from MODIS (Giglio et al., 2006). Total carbon emissions were calculated as the product of burned area, fuel load and combustion completeness. Fuel
- ²⁵ load depends on vegetation type, climate, soil type and time since last fire, while combustion completeness, describing the fraction of the available fuel combusted during a fire, depends on the type of fire, the type of fuel (e.g., stems, leaves and litter) and





its moisture content (Langmann et al., 2009). For GFED, the Carnegie-Ames-Stanford-Approach (CASA) biogeochemical model was used to estimate combustion completeness as well as fuel load and the associated spatial variability (van der Werf et al., 2006, and references therein). BC emissions were then derived from the total carbon emissions based on BC emission factors.

A recent study by Randerson et al. (2012) pointed out that, in the current generation of global burned area products (for example, the GFED), small fires had not been systematically quantified since many of them were well below the detection limit of burned area. However, these fires often generated thermal anomalies that could be detected by satellites, such as from the MODIS active fire counts products. Burned areas in the GFED emissions were mainly derived from MODIS 500 m surface reflectance product, which could detect large fires that leave fire scars greater than 500 m or so (Giglio et al., 2006, 2010). In contrast, active fires, based on thermal anomalies, could detect fires that are an order of magnitude smaller, which may capture many aspects of the

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- spatial distribution and seasonality of burning (Randerson et al., 2012 and references therein). As a result, active fires probably could better capturing smaller prescribed and agricultural fires (Randerson et al., 2012 and references therein). The active fire data are often used as a proxy for area burned due to the lack of long-term global burned area data (Giglio et al., 2006). There are many uncertainties to relate fire counts to ac-
- tual area burned due to inadequate temporal sampling, variability in fuel conditions and cloud cover, differences in fire behavior, and issues related to spatial resolution (Giglio et al., 2006; Kasischke et al., 2003). We here use MODIS active fire counts to scale the spatiotemporal variability of the GFEDv2 biomass burning emissions from a 3 yr period (2005–2007), aiming to capture some of the missing small fires. The assump-
- tion here is that burned area is proportional to fire counts. Based on this assumption, estimated burned areas from fire counts are relatively accurate (Giglio et al., 2006, 2010). Since the relationship between burned area and active fires was non-linear in GFEDv2, the linear scale will give a different pattern of the emissions, which probably could improve the spatiotemporal distributions of the emissions and capture some





of the missing small fires. The GFEDv2 emissions in North America are adjusted for three zones: boreal North America, temperate North America, and Mexico and Central America, based on the geographic regions defined in the GFED inventory (Giglio et al., 2006, 2010). To get BC emissions in each grid box, we kept the total emissions
⁵ the same for each zone during the three years (sum up the monthly emissions) and then redistributed total emissions according to the active fire counts in this grid box. We redistributed fire emissions in three regions for a 3 yr period as a way to reduce the uncertainties in relating fire counts to emissions. The resulting emissions have a temporal resolution of eight days as dictated by the 8-day fire counts. The resulting
¹⁰ emissions were then used as a priori for our analytical inversions to be discussed in Sects. 4 and 5.

Figure 2 shows MODIS active fire counts and GFEDv2 biomass burning emissions of BC, before and after the aforementioned adjustments, summed over the WUS (hereto-fore defined as $30-50^{\circ}$ N, $100-125^{\circ}$ W), from 2005 to 2007. Fire seasons in the WUS

- typically last from late June throughout October even November in terms of both the fire counts and the emissions. The fire season in 2006 is from July to September primarily. Among the three years, 2006 is a relatively large fire year. The adjustment primarily shifts emissions from late to early and middle summer (33 % decrease in September-October and 56 % increase in June–August). Figure 3 compares the spatial distribu-
- tions of monthly biomass burning emissions, before (Fig. 3b) and after (Fig. 3c) the aforementioned adjustments, for July, August, and September 2006, respectively. Also shown are the differences between the adjusted and the standard emissions (Fig. 3d) and MODIS active fire counts (Fig. 3a). The adjusted emissions show the similar spatial distributions and temporal variations as the MODIS active fire counts. The ad-
- justed emissions in August and September significantly increase in the agricultural areas in Washington, Idaho and Oregon states. Figure 4 compares the observed and model simulated daily surface BC concentrations at selective IMPROVE sites for May– October 2006. Model results shown here are from simulations at 2° × 2.5° horizontal resolution and with the standard or the adjusted GFEDv2 8-day emissions. With higher





fire counts during late June through August, simulated surface BC concentrations with the adjusted emissions show enhancements at some IMPROVE sites, for example, UL Bend, MT (47.6° N, 108.7° W, 0.89; up to ~ 100 % increase) and North Cheyenne, MT (45.7° N, 106.6° W, 1.28 km; up to ~ 90 % increase). Since emissions were shifted from

- ⁵ September and October, simulated surface BC concentrations decreased during this period, seen at sites such as Starkey, OR (45.2° N, 118.5° W, 1.26 km; up to ~ 10 % decrease) and Mt. Cabinet, MT (48.0° N, 115.7° W, 1.44 km; up to ~ 10 % decrease). Figure 5 compares the observed and model simulated daily surface BC concentrations for May–October 2006, averaged for sites at the altitude ranges 0–1, 1–2, 2–3, and
- ¹⁰ 3–4 km, respectively. Model results shown here again are from the same simulations as those in Fig. 4. The emissions adjustments lead to small yet significantly relative enhancements of model surface BC concentrations (up to ~ $0.05 \,\mu g m^{-3}$) during late June to August. These enhancements are particularly evident at the 0–1 and 1–2 km altitude ranges.

15 4 Analytical solution to the inverse problem

We here apply linear analytical inversions to estimate the monthly BC emissions constrained by the IMPROVE surface BC concentrations. Measured BC concentrations (assembled in a measurement vector *y*) can be related to the sources of BC (assembled in a state vector *x*) in Eq. (1). The state vector *x* comprises monthly sources
estimates from different geopolitical regions and from different BC source types. The selection of *x* will be discussed later in this section (Fig. 1). *y* is an observation vector and in this study consists of daily BC surface concentrations from the IMPROVE network. Our analyses include 67 IMPROVE sites in the WUS (Fig. 1) as mentioned in Sect. 2, following Mao et al. (2011). These sites are in remote and often elevated mountainous locations. IMPROVE measurements are made every three days and the reported values are 24 h averages. Thermal Optical Reflectance (TOR) combustion method was used for the BC measurements based on the preferential oxidation of





OC and BC at different temperatures (Chow et al., 2004). The uncertainties of the TOR method are still difficult to quantify (Park et al., 2003; Chow et al., 1993). To linearization forward model, the Jacobian matrix **K** (**K** = $\nabla_X F$) is used to relate sources to concentrations in a forward model and describes the sensitivity of the measurement vector

- to finite change in the state vector. The forward model, in our case, GEOS-Chem provides the connection between emissions and species concentrations. K is calculated by dividing the model simulated variation of BC concentrations by the perturbation of monthly mean emissions from each individual source or source region. The linear approximation of the forward model was tested by perturbing monthly mean emissions by
- ¹⁰ 5% and 10%. We found almost linear changes of model BC concentrations resulting from perturbing the monthly mean emissions. The error vector $\boldsymbol{\varepsilon}$, in Eq. (1), includes contributions from errors in the observations, in the forward model, and in the model parameters. From inversion of Eq. (1), involving weighting the error statistics of $\boldsymbol{\varepsilon}$ and $\boldsymbol{\varepsilon}_{a}$ (a priori error), we can obtain an optimal solution of \boldsymbol{x} by given \boldsymbol{y} based on our prior the knowledge \boldsymbol{x}_{a} .

The inverse model describes the best estimate of sources of BC, which is consistent with both the observed BC concentrations and the a priori sources of BC, given their respective uncertainties. Based on Bayes' theorem with the assumption of Gaussian error distributions, the MAP solution for x by given y, is equivalent to finding the minimum in the cost function J(x) (Rodgers, 2000):

$$J(\boldsymbol{x}) = (\boldsymbol{y} - \boldsymbol{K}\boldsymbol{x})^T \boldsymbol{S}_{\Sigma}^{-1} (\boldsymbol{y} - \boldsymbol{K}\boldsymbol{x}) + (\boldsymbol{x} - \boldsymbol{x}_a)^T \boldsymbol{S}_a^{-1} (\boldsymbol{x} - \boldsymbol{x}_a)$$
(2)

where x_a and S_a are the background model state vector and its associated error covariance; S_{Σ} is the covariance of the total observational error. Solution to $\nabla_x J(x) = 0$ yields,

$$\hat{\mathbf{x}} = \mathbf{x}_{a} + (\mathbf{K}^{T} \mathbf{S}_{\Sigma}^{-1} \mathbf{K} + \mathbf{S}_{a}^{-1})^{-1} \mathbf{K}^{T} \mathbf{S}_{\Sigma}^{-1} (\mathbf{y} - \mathbf{K} \mathbf{x}_{a})$$
$$\hat{\mathbf{S}} = (\mathbf{K}^{T} \mathbf{S}_{\Sigma}^{-1} \mathbf{K} + \mathbf{S}_{a}^{-1})^{-1} = (\mathbf{I} - \mathbf{A}) \mathbf{S}_{a} (\mathbf{I} - \mathbf{A})^{T} + \mathbf{G} \mathbf{S}_{\Sigma} \mathbf{G}^{T}$$

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(3) (4) where \hat{x} is the optimized a posteriori state vector and \hat{S} is the a posteriori error covariance matrix, describing the error on \hat{x} ; I is the identity matrix; gain matrix $\mathbf{G} = \frac{\partial \hat{x}}{\partial y}$ describes the sensitivity of the retrieval to the observations; the averaging kernel matrix $\mathbf{A} = \frac{\partial \hat{x}}{\partial x} = \mathbf{G}\mathbf{K} = \mathbf{I} - \hat{\mathbf{S}}\mathbf{S}_{a}^{-1}$ represents the sensitivity of the MAP solution of the true state. The first and second terms of the right side of Eq. (4) are the smoothing error covariance matrix from the a priori and retrieval error covariance matrix from the ob-

- covariance matrix from the a priori and retrieval error covariance matrix from the observational error. In a successful inversion, J(x) should be of the same order as the number of observations, provided that S_{Σ} and S_a are properly specified (Palmer et al., 2003).
- ¹⁰ We here test the matrix of averaging kernel **A** to inspect the ability of the observing system to determine different elements of the state vector, by taking into account the assigned measurement and the a priori state uncertainties (Kasibhatla et al., 2002). Averaging kernel matrix provides the information on the sensitivity of the a posteriori estimates to the unknown true state. In the ideal case, **A** would be an identity ma-
- trix. Averaging kernels peaking at their own state vector element denote a well constrained source, which shows inversion system has enough information to constrain the source categories independently. Starting from five components of biomass burning emissions of BC (the Northern Rocky Mountains, the Southern Rocky Mountains, California, the Southwest, and the Pacific Northwest) in the WUS following the study of
- Jaffe et al. (2008), we used averaging kernels (not shown) to determine which sources or aggregation of sources could be constrained independently with IMPROVE data. We found that biomass burning emissions of BC in the Northern and the Southern Rocky Mountains were too collocated to be retrieved independently (Kasibhatla et al., 2002), and such was the case also for biomass burning emissions in California and
- the Southwest. We aggregated these emissions together as IMPROVE data do not provide independent information on these two regions, the Northern and the Southern Rockies (not shown) as well as California and the Southwest. We thus define a fourcomponent state vector: biomass burning emissions in the Rockies (BBRM), biomass burning emissions in California and the Southwest (BBCSW), biomass burning emis-





sions in the Pacific Northwest (BBPNW), and anthropogenic emissions in the WUS (ANTHWUS). Figure 1 shows these three biomass burning BC source regions in the WUS. Anthropogenic emissions discussed here include both fossil fuel and biofuel emissions from Bond et al. (2007) unless stated otherwise. The averaging kernels are also useful to test the sensitivities of the inversions to the uncertainties of the observations and the a priori. The performances of the averaging kernels are discussed further in later paragraphs.

We conducted inversions with different sets of error specifications as a way to exam the sensitivities of the inversions to the a priori error and the observation error. We will discuss the error specifications in the next three paragraphs. We assumed errors of each element of state vector to be spatially uncorrelated so that background model state vector \mathbf{S}_{a} would be diagonal. Previously, Mao et al. (2011) found that North American anthropogenic emissions in the model were reasonably prescribed, although model was still not prefect to capture the day-to-day variabilities and magni-

- ¹⁵ tudes of observed surface BC concentrations at sites where North American anthropogenic emissions dominated. We thus assumed that the uncertainties of fossil fuel and biofuel emissions were 50 % in the WUS. GFED biomass burning emissions of BC were assigned an uncertainty of 300 or 500 %. Our assumption for biomass burning emissions is based on the fact that the uncertainty of BC emissions estimates gener-
- ally is a factor of 2–5 on regional scales (Ramanathan and Carmichael, 2008). We also believed that GFEDv2 biomass burning emissions of BC were biased low by a factor of two in the WUS during July–October fire season of 2006 (Mao et al., 2011).

The total observation error $\bm{S}_{\bm{\Sigma}}$ includes contributions mainly from transport error in the forward model, representation error, and measurement accuracy. We estimated

the transport errors by computing the variance of the relative difference between the observations and the collocated model BC concentrations (Palmer et al., 2003; Heald et al., 2004). The mean model bias, as diagnosed by the mean relative difference, is due to errors in the a priori sources and the variance of relative residual error is mainly due to errors in the transport. The calculated transport error is about 20% in





the GEOS-Chem and consistent with previous studies (Palmer et al., 2003; Heald et al., 2004). The representation error describes the mismatch between the observations and the corresponding values simulated by the forward model. This error arises because the model only provides concentration data averaged over the grid scale which the

- ⁵ observations do not fully cover. Representation error was about 5–10 % of the observed concentrations, by exam the statistics of the subgrid variability over the 2° × 2.5° GEOS-Chem model grid (Palmer et al., 2003). We assumed representation error to be 5 or 10 % following previous works. As for the instrument accuracy, uncertainties of the TOR method are difficult to quantify and no total error is prescribed for the BC measurement
- from the IMPROVE network (Park et al., 2003; Chow et al., 1993). We assumed the measurement error to be 5–10%. We thus tested the total observation error S_{Σ} using 30 or 50%.

We here use the averaging kernels and the number of degrees of freedom for signal (DOFs) to test the sensitivities of the inversions to the uncertainties of the a priori and

- the inversion system. DOFs is the trace of the averaging kernel matrix (Rodger, 2000). The number of pieces of information from a perfect knowledge of the observing system has an expected value of the size of the state vector. DOFs thus is expected to be close to 4 in our case. Table 1 compares the DOFs from inversions using different error specifications as well as at two different model horizontal resolutions. By inspecting av-
- eraging kernels (not shown) and DOFs (Table 1), we considered the best inversions as those with error specifications of 500% for the a priori biomass burning emissions and 30% for the observations. The inversions with this set of error specifications show better performance of averaging kernels (Fig. 6) and higher values of DOFs (closer to four; bottom two rows in Table 1). Figure 6 compares the averaging kernels for the inversions
- of BC sources for May–October 2006, using GEOS-Chem at 2° × 2.5° and 0.5° × 0.667° horizontal resolutions and with aforementioned best set of error characterizations (bottom two rows in Table 1). Individual lines here are corresponding to the individual rows of averaging kernel matrix **A**. Red line for example, it is the sensitivity of the a posteriori emissions in California and the Southwest to the unknown true state, which shows





that inversion system has enough information to constrain the biomass burning emissions in California and the Southwest uniquely, especially during July to September. Generally, our inversion system could largely constrain the four elements of the state vector independently, especially during July to September and to a less degree in Oc-

- ⁵ tober. Anthropogenic emissions in the WUS could not be separated completely from the three biomass burning emissions elements in May and June. We also compared the averaging kernels and DOFs at different model resolutions. Averaging kernels at both two model resolutions show similar performances, which indicate that inversion system can largely constrain the four elements of state vector independently. DOFs
- ¹⁰ also have acceptable values for inversions at both two resolution models (Table 1). We thus believe our error specifications are appropriate. The retrieval at 0.5° × 0.667° horizontal resolution is better constrained by the observation system, which shows better performance of averaging kernels and higher DOFs values (bottom row in Table 1). Further discussion about the inversions with different set of error characterizations and
- at different model horizontal resolutions are presented in Sect. 5.1 (Fig. 7). The sensitivity of the a posteriori solutions to the assumed uncertainties is also assessed in Sect. 5.1.1. We evaluate a posteriori estimates in Sect. 5.1.3. More discussions about the sensitivity of model resolution to the retrieval are presented in Sect. 5.2.

5 Inversion results and discussions

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20 5.1 A posteriori estimates of BC emissions

5.1.1 Comparisons with a priori emissions

Figure 7 shows the a priori and the a posteriori estimates of monthly BC emissions in the WUS for May–October 2006. For the purpose of clarity, anthropogenic emissions are divided by three in the figures to be compatible with the magnitude of the biomass burning emissions. Error bars here represent uncertainties of the emissions.





The a posteriori estimates of BC sources are from several sensitivity experiments discussed in Sect. 4 (Table 1). Those experiments include inversions using different sets of error specifications, with 300 or 500% for the uncertainties of the a priori biomass burning emissions and with 30 or 50% for the observational error, and at $2^{\circ} \times 2.5^{\circ}$ 5 or 0.5° × 0.667° two different model horizontal resolutions. The cost functions reduce by ~ 40 % after inversions in those experiments. The BC emissions after inversions with different error specifications show similar trend. The a posteriori biomass burning emissions increase dramatically and consistently (about a factor of 3-5 on average) while the a posteriori anthropogenic emissions reduce substantially (\sim 50 %). Detailed analyses are in Table 2 and discussed in the following paragraph. Our retrievals also 10 largely reduce the uncertainties of the emissions, which decline by at least 50% or by even larger than 90%. Those sensitivity experiments thus reflect that our retrievals are reliable. We consider the best retrievals as those with 500% for the error of a priori biomass burning emissions and 30% for the total observational error. The best retrievals include relatively small uncertainties of the a posteriori. The estimates are also 15 consistent with our discussion in Sect. 4, which shows that the inversions with this set of

- error specifications provide largest DOFs values (Table 1) as well as best performance of the averaging kernels. The inversion at $0.5^{\circ} \times 0.667^{\circ}$ horizontal resolution with best error specifications show smallest uncertainties of the a posteriori emissions, which
- ²⁰ again is consistent with analyses based on Table 1 and Fig. 6. This implies that inversion at $0.5^{\circ} \times 0.667^{\circ}$ horizontal resolution with errors of 500 % for the a priori biomass burning emissions and 30 % for the observations provides the best estimates. This inversion is further confirmed to be the best one by analyzing resulting model surface BC concentrations both in the Sect. 5.1.3 and in Sect. 5.3. In the following text, our
- discussion about the a posteriori estimates of BC emissions and the resulting surface BC concentrations are thus based on the retrievals with aforementioned error specifications. For further evaluation, we summarize the monthly BC emissions of a priori and a posteriori estimates from the best inversions in the following paragraph.





Table 2 compares monthly biomass burning BC emissions from the three regions in the WUS and anthropogenic BC emissions over the WUS before and after retrievals for May-October 2006. The a priori GFEDv2 and the a posteriori biomass burning BC emissions are from inversions at 2° × 2.5° and 0.5° × 0.667° horizontal resolutions. In general, the a posteriori biomass burning emissions increase by factors of 4.7 at $2^{\circ} \times 2.5^{\circ}$ and 2.8 at $0.5^{\circ} \times 0.667^{\circ}$ horizontal resolutions in the WUS during July to September, compared with a priori emissions. The a posteriori biomass burning emissions also show large variations from month to month, at different source regions and at different model horizontal resolutions. For example, the biomass burning emissions of BC after retrieval at 2° × 2.5° horizontal resolution during July to September are 6.0 10 and 3.3 times higher than the a priori GFEDv2 in the Rockies and in California and the Southwest, respectively. Monthly biomass burning emissions at 2° × 2.5° horizontal resolution in the WUS may increase by factors of 6.3 in July and 3.6 in August; the correspondingly values are 4.1 and 2.2, respectively, at 0.5° × 0.667° horizontal resolutions. More discussions about the sensitivity of the retrieval to model resolution are pre-15

- sented in the Sect. 5.2. The a posteriori anthropogenic emissions in the WUS decrease by 48 % from the inversion at 2° × 2.5° and by 36 % at 0.5° × 0.667° for May–October 2006 compared to 2000 level (Bond et al., 2007). This decreasing trend of anthropogenic emissions estimates is consistent with the study by Rao and Somers (2010),
- which found that BC emissions in the US had declined by about 30 % from 1990 to 2005 and were expected to decline by an additional 80 % by 2030 compared to 2005 level. Observed BC concentrations from 50 IMPROVE sites in the US also decreased by over 25 % on average from 1990 to 2004, which also implies that emission control have been effective in reducing BC across US (Murphy et al., 2011).

25 5.1.2 Comparisons with GFEDv3 and FLAMBE

Monthly BC emissions from GFEDv3 (van derWerf et al., 2010) and the Fire Locating and Monitoring of Burning Emissions (FLAMBE) inventory (Reid et al., 2009) are calculated over three biomass burning regions for May–October 2006 and compared with a





posteriori estimates (Table 2). GFEDv3 is a new version of GFED and mainly updated by following four aspects. First, the spatial resolution of the global grid was quadrupled from 1 to 0.5°; second, native 500 m MODIS daily burned area maps were applied (Giglio et al., 2009); thirdly, the regional regression trees of the GFEDv2 were largely re-

- ⁵ placed with a local regression approach in producing the indirect, active-fire based estimates of burned area; finally, a revised version of CASA biogeochemical model were used. FLAMBE provides carbon emissions at 1° × 1° spatial resolution and hourly temporal resolution based on both MODIS and Geostationary Operational Environmental Satellites (GOES) fire counts. Hourly emissions from FLAMBE are available from 2005
- (data available at http://www.nrlmry.navy.mil/aerosol_web/arctas_flambe/data_hourly). Fire pixels detections and sub-pixel burning characterizations were computed based on operational NOAA/NESDIS GOES Wild-Fire Automated Biomass Burning Algorithm (WF_ABBA) for most of the Western Hemisphere. For the rest of the globe, the near real time University of Maryland/NASA MODIS fire products from Terra and Aqua were
- ¹⁵ used. The Advanced Very High Resolution Radiometer (AVHRR) derived Global Land Cover Characteristics (GLCC) data base (version 2.0) was used to assign surface emissivity and to screen for false alarms (http://edc2.usgs.gov/glcc/glcc.php). Based on the thermal anomaly and fire radiative power in a subpixel fire, this dataset contains hourly biomass burning areas as well as carbon emissions in a certain location with coordi-
- nated latitudes and longitudes. Total smoke emissions were estimated using average emission factor and fuel loading recommended in FLAMBE. The individual BC and OC emissions were partitioned using the ratio of the GFEDv2 emission factors between BC and OC (Andreae and Merlet, 2001). All emissions were injected into the local planetary boundary layer as defined from the GEOS-5 data (Fisher et al., 2010).
- The two versions of GFED BC emissions generally show quite different spatiotemporal distributions. The GFEDv3 BC emissions increase by 8% in the WUS during July to September 2006. These enhancements are most evident in August (20%) and September 2006 (15%). However, total BC emissions for 2006 in the GFEDv3 are lower than those in its previous version (not shown). Compared with the a posteriori





estimates, GFEDv3 emissions of BC in the WUS are still biased low by factors of 4.4 at $2^{\circ} \times 2.5^{\circ}$ and 2.7 at $0.5^{\circ} \times 0.667^{\circ}$ for July–September 2006. FLAMBE BC emissions in the WUS are much higher than the a priori GFED emissions (a factor of 5.9) and even the a posteriori estimates (factors of 1.3 at $2^{\circ} \times 2.5^{\circ}$ and 2.1 at $0.5^{\circ} \times 0.667^{\circ}$) dur-

- ing July–September 2006. FLAMBE may cover more small fire events due to its hourly temporal resolution. However, simulated BC concentrations with FLAMBE emissions are still problematic and cannot capture the temporal variations of observations well (not shown). Model simulations with FLAMBE also show false high fire contributions to surface BC concentrations, which implies that FLAMBE might be systematically too
- ¹⁰ high. Our analyses are in agreement with previous studies by Wang et al. (2011) and Fisher et al. (2010), which suggested that FLAMBE needed to be reduced to 53 % in Russian and 45 % in the Southeast Asian, respectively, when simulated CO, BC and OC with GEOS-Chem for the NASA Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) (Jacob et al., 2010) and the NOAA ¹⁵ Aerosol, Radiation and Cloud Processes affecting Arctic Climate (ARCPAC) (Brock et
- ¹⁵ Aerosol, Radiation and Cloud Processes affecting Arctic Climate (ARCPAC) (Brock ef al., 2011) campaigns.

5.1.3 Evaluation a posteriori estimates with IMPROVE observations

Figures 8–10 compare the observed and simulated surface BC concentrations at selective IMPROVE sites in the Rockies (Fig. 8), in the Pacific Northwest (Fig. 9), and in

- ²⁰ California and the Southwest (Fig. 10), respectively. Model results shown here are from the simulations with the standard GFEDv2 8-day emissions at 2° × 2.5° horizontal resolution and with the a posteriori estimates at both 2° × 2.5° and 0.5° × 0.667° horizontal resolutions for May–October 2006. Model simulated surface BC concentrations with the a posteriori emissions show significant enhancements for July–September 2006.
- ²⁵ Model surface BC after inversions can capture the major fire episodes at many IM-PROVES sites. Here shown some representative sites in the three regions over the WUS. Model surface BC concentrations with the a posteriori estimates show better agreement with IMPROVE observations, which largely reproduce both synoptic vari-





ability and magnitude of the observed surface BC concentrations. Detailed statistic analyses are in Sect. 5.3. Discussions about the sensitivity of the retrieval to model resolution are presented in Sect. 5.2.

- Figure 11 compares the observed and simulated surface BC concentrations averaged for sites in the Rockies, in California and the Southwest, and in the Pacific Northwest, respectively. Model simulations are same as those in Figs. 8–10. Again, Fig. 11 shows significantly increased model surface BC concentrations with the a posteriori emissions for July–September 2006, especially in the Rockies (up to ~ 100 % increase) and to a less degree in the Pacific Northwest (up to ~ 50 % increase). These improved comparisons between simulated surface BC concentrations and the observations in the
- Rockies may be due to the largest enhancement of biomass burning emissions after inversions. Model simulated surface BC concentrations averaged for sites in California and the Southwest show slight increase compared with those averaged in the other two regions, which may be due to the fact that most of IMPROVE sites in California and the Southwest are not located at the regions with large fires.
 - Figure 12 compares the observed and model simulated daily surface BC concentrations for May–October 2006, averaged for sites at the altitude ranges 0–1, 1–2, 2–3, and 3–4 km, respectively. Model simulations again are same as those in Figs. 8–10. Model simulated surface BC concentrations with the a posteriori emissions show sub-
- ²⁰ stantial enhancements at all altitude ranges, especially at the 1–2 (up to 0.18 μ g m⁻³ increase) and 2–3 km (up to 0.11 μ g m⁻³ increase) altitude ranges. There are also some improvements in late June 2006 (up to 0.06 μ g m⁻³ at the 2–3 km altitude range), which may partially verify our aforementioned adjustment using the MODIS fire counts to improve the spatiotemporal distributions of the GFED emissions. Our previous study by
- Mao et al. (2011) showed that simulated BC concentrations were biased low by a factor of two at elevated mountainous sites during the July–October 2006. The a posteriori emissions lead to an average bias reduction of ~ 50 % in the simulated surface BC concentrations at the 1–2 km altitude range. As widely pointed out in previous studies, part of the discrepancies is because of the model resolution, which is too coarse to





resolve fine regional distributions of BC (Mao et al., 2011; Fairlie et al., 2007), especially at elevated mountainous sites. Lack of detection of small fires may still be a main problem to the aforementioned discrepancies.

5.2 Sensitivity of the retrieval to model resolution

- As discussed in Sects. 4 and 5.1.3, inversions are very sensitive to model horizontal resolution. Both Table 1 and Figs. 6–7 indicate that inversions at 0.5° × 0.667° horizontal resolutions provide better estimates than those at 2° × 2.5°, which include smaller uncertainties of a posteriori emissions, larger DOFs, and better performance of averaging kernels. As for the sensitivity of the resulting BC concentrations to model resolution,
- ¹⁰ the simulated surface BC concentrations using the nested model provide better agreement with IMPROVE observations (Figs. 11, 13, and 14), which are in consistent with previous studies by Wang et al. (2004) and Chen et al. (2009). As suggested by Wang et al. (2004), higher-resolution model allowed for more efficient, advection-related, ventilation of the lower atmosphere, reflecting the localized upward motion not resolved in
- the coarser-resolution simulation. Model results using the nested model thus could provide better agreement with the measurements compared to those from coarse models in both absolute values and distributions of chemical species regionally (Chen et al., 2009). Detailed statistic analyses are in Sect. 5.3.

The largest differences of the estimated emissions at two different resolutions are during July to September 2006. In general, the a posteriori biomass burning emissions using the nested model increase less than those using the coarse-resolution model (Table 2), compared to the a priori emissions. The a posteriori biomass burning emissions increase by factors of 2.8 at 0.5° × 0.667° and 4.7 at 2° × 2.5° in the WUS for July– September 2006. With high-resolution model, the a posteriori biomass burning emissions in the Rockies increase by a factor of 4.8 for July–September 2006; while those at coarse-resolution model increase by a factor of 7.0. The two a posteriori estimates differ largest in biomass burning emissions in California and the Southwest (a factor of 5.9) and in the Pacific Northwest (a factor of 2). Those guite different emission distributions





at two different horizontal resolutions are likely due to the more concentrated emissions in the nested model. Simulated surface BC concentrations with retrieved emissions at different resolutions also show quite different variations and magnitudes from site to site (Figs. 8–10). Simulated surface BC concentrations provide better comparisons with IM-

- ⁵ PROVE observations at 2° × 2.5° resolution at sites such as Craters Moon, ID (43.5° N, 113.6° W, 1.82 km; Fig. 8), Starkey, OR (Fig. 9), and Lassen Volcanic, CA (40.5° N, 121.6° W, 1.73 km; Fig. 10); while more sites show better comparisons at 0.5° × 0.667° horizontal resolutions, such as, Bridger Wild, WY (43.0° N, 109.8° W, 2.63 km; Fig. 8), Three Sisters, OR (44.3° N, 122.0° W, 0.89 km; Fig. 9), and Pasayten, WA (48.4° N, 119.9° W, 1.63 km; Fig. 10). However, if we average these sites together, there are no such large differences (Figs. 11 and 13). Thus, our speculation is that the a posteriori
- such large differences (Figs. 11 and 13). Thus, our speculation is that the a posteriori emissions retrieved by $2^{\circ} \times 2.5^{\circ}$ resolution model are likely overestimated.

5.3 Statistic evaluation of model performance

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We here use the Taylor diagram and Taylor skill score (Taylor, 2001) to further evaluate the model simulations discussed in Sect. 5.1.3. Taylor diagram relates the centered Root Mean Square (RMS) error, the pattern correlation and the standard deviation of observations and model simulations. The centered pattern RMS difference is defined by,

$$E' = \left\{ \frac{1}{N} \sum_{n=1}^{N} [(f_n - \bar{f}) - (r_n - \bar{r})] \right\}^{1/2}$$

²⁰ Where, f_n and r_n are the observed and model BC concentrations with *N* discrete data; \overline{f} and \overline{r} are the mean values. Standard deviation of observation data is plotted along the abscissa. Simulated fields are located in the first quadrant if the correlation with the reference data is positive (Gleckler et al., 2008). For both the observation and model data, the radial distance from the origin is proportional to the standard deviation. The pattern correlation between the simulated field and the observation data is related to



(5)



the azimuthal angle, and the centered RMS error difference between a simulated field and the reference data is proportional to the distance between these two points (i.e., the closer a model is to the observational point, the lower its centered RMS error). Taylor skill score is defined by,

$$S = \frac{4(1+R)}{(\frac{\sigma_f}{\sigma_r} + \frac{\sigma_r}{\sigma_f})^2(1+R_0)}$$

Where *R* is the correlation coefficient between *f* and *r*; σ_f and σ_r are the standard deviations of f and *r*; R_0 is the maximum correlation attainable. As the model variance approached the observed variance, $\sigma_f/\sigma_r \rightarrow 1$, $R \rightarrow R_0$ and skill score approached unity. Under this definition, skill score increases toward value 1 as the correlation becomes larger and larger or as the model variance approaches the observed variance. Model simulation thus shows better performance with higher skill score.

We use Taylor diagrams in Fig. 13 to compare the correlation coefficients, the standard deviations, and the centered RMS error of simulated and observed BC concentrations averaged for 69 IMPROVE sites. Taylor diagram in Fig. 14 is same as Fig. 13 but

- for 69 individual sites. Model standard deviations and centered RMS error for each site are normalized by observed standard deviation at that site in Fig. 14. Three indexes, correlation coefficients, standard deviations and centered RMS error, consistently show that model simulated surface BC concentrations with the a posteriori emissions are in better agreement with the observations, especially using the nested model. Averaged
- ²⁰ correlation coefficient for all 69 sites increases from 0.28 with the standard GFEDv2 emissions to 0.36 with the a posteriori estimates (Fig. 13). Our retrievals also largely improve the dispersion of the model BC concentrations. Standard deviations after inversions at two different resolutions are closer to that of the observations. The averaged standard deviations with the a posteriori emissions increase from 0.11 to 0.16
- at $2^{\circ} \times 2.5^{\circ}$ and to 0.19 at $0.5^{\circ} \times 0.667^{\circ}$. The centered RMS after the retrievals are also lower, which again implies that model simulated surface BC concentrations using the a posteriori estimates are closer to the observations. With the a posteriori emis-



(6)

sions, the resulting skill scores averagely increase by 86% at 2° × 2.5° and by 132% at 0.5° × 0.667°. Model simulated surface BC concentrations thus show large improvement with the a posteriori emissions, especially using the nested model.

6 Summary and conclusions

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⁵ We have applied Bayesian linear inversions to derive top-down estimates of biomass burning emissions of BC in the WUS for May–October 2006 by inverting surface BC concentrations from the IMPROVE network. We conducted analytic inversions using the GEOS-Chem chemical transport model at both 2° × 2.5° (globally) and 0.5° × 0.667° (nested over North America) horizontal resolutions. Model results with both the a priori and the a posteriori emissions were compared to the surface BC concentrations observed from the IMPROVE network.

To capture some missing small fires in the GFED emission inventory, we first improved the spatiotemporal distributions of the BC emissions from the GFEDv2 using MODIS 8-day active fire counts from 2005–2007. The resulting emissions were then used as the a priori for the inversions. The adjustment primarily shifted emissions from late to early and middle summer (33% decrease in September–October and 56% increase in June–August). The adjustments led to significantly relative enhancements of simulated surface BC concentrations at some IMPROVE sites during late June through August 2006. These enhancements were particularly evident at the 0–1 and 1–2 km altitude ranges.

Three a posteriori estimates with different sets of error specifications showed similar monthly emissions, which reflected that our retrievals were reliable. The best retrievals were those with the uncertainties of 500 % for the a priori biomass burning emissions and 30 % for the observations, which included relatively small uncertainties of the a posteriori and largest DOFs values as well as best performance of the averaging kernels. The a posteriori anthropogenic emissions from Bond et al. (2007) in the WUS decreased by 48 % from the inversion at $2^{\circ} \times 2.5^{\circ}$ and by 36 % at $0.5^{\circ} \times 0.667^{\circ}$; while



as the a posteriori biomass burning emissions increased by factors of 4.7 and 2.8, respectively, relative to the a priori emissions during July to September 2006. The a posteriori biomass burning emissions also showed large variations from month to month, at different source regions, and at different model horizontal resolutions. The a poste-

riori biomass burning emissions increase less at $0.5^{\circ} \times 0.667^{\circ}$ than those at $2^{\circ} \times 2.5^{\circ}$. 5 The large differences in the a posteriori emissions at the two model horizontal resolutions were likely due to the better simulated boundary layer and more concentrated emissions in the nested model.

Compared with a priori emissions in the WUS during July to September 2006, the GFEDv3 BC emissions increased by 8% in the WUS, especially evident in August 10 (20%) and September 2006 (15%); while FLAMBE BC emissions were higher by a factor of 5.9. Compared with the a posteriori estimates, GFEDv3 emissions of BC in the WUS were still biased low by factors of 4.4 at 2° × 2.5° and 2.7 at 0.5° × 0.667° for July-September 2006; while FLAMEB were higher by factors of 1.3 and 2.1, respectively. However, FLAMBE emissions might be systematically too high and problematic in its 15

temporal variations.

Model surface BC with the a posteriori emissions captured the major fire episodes at many IMPROVE sites, especially at the 1-2 and 2-3 km altitude ranges. Model surface BC concentrations with the a posteriori estimates provided better agreement with

- IMPROVE observations for July-September 2006, especially in the Rockies and to a 20 less degree in the Pacific Northwest. With the a posteriori emissions, the resulting Taylor skill scores increased by 86 % at 2° × 2.5° and by 132 % at 0.5° × 0.667° horizontal resolutions. Model BC concentrations at two different resolutions also showed largest differences during July to September. The a posteriori emissions led to a bias reduction
- of ~ 50 % on average in the simulated surface BC concentrations. 25

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Table 1. Number of degrees of freedom for signal (DOFs) for inversions (May–October 2006)

 using different error characterizations and at different model horizontal resolutions.

Model	Error	DOFs					
resolution	specification	Мау	Jun	Jul	Aug	Sep	Oct
2° × 2.5°	${f S}_{\Sigma}=50~\%$ ${f S}_{a}~BB=300~\%$	2.36	2.68	3.60	3.85	3.81	2.99
	${f S}_{\Sigma}=30~\%$ ${f S}_{a}~BB=300~\%$	2.87	2.80	3.80	3.93	3.91	3.41
	${f S}_{\Sigma}=30~\%$ ${f S}_{a}~BB=500~\%$	3.44	3.03	3.92	3.97	3.96	3.74
0.5° × 0.667°	$S_{\Sigma} = 30 \%$ $S_{a} BB = 500 \%$	3.57	3.14	3.93	3.98	3.97	3.82

Table 2. Monthly biomass burning BC emissions from three regions in the WUS (see Fig. 1) and anthropogenic BC emissions from the WUS for May–October 2006 (unit: Gg). A priori GFEDv2, the a posteriori biomass burning emissions from inversions at $2^{\circ} \times 2.5^{\circ}$ and at $0.5^{\circ} \times 0.667^{\circ}$ horizontal resolutions are shown. Also shown are GFEDv3 and the Fire Locating and Monitoring of Burning Emissions (FLAMBE) for comparison.

BC Emissions (Gg), 2006				
-Sep total				
3.01				
2.07				
1.67				
6.75				
17.59				
21.2				
8.88				
1.65				
31.73				
9.07				
14.3				
1.51				
3.37				
19.18				
11.24				
2.81				
2.28				
2.14				
7.23				
14.55				
16.79				
8.72				
40.06				

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Fig. 2. MODIS active fire counts (top panel) and the GFEDv2 biomass burning emissions of BC (middle and bottom panels) summed over the Western US (WUS) from 2005 to 2007. The emissions before (middle panel) and after (bottom panel) applying spatiotemporal adjustments based on the active fire counts (see text for details on the adjustments) are both shown. Data shown here has an 8 day temporal resolution. MODIS active fire counts data are available at ftp: //fuoco.geog.umd.edu. GFED data are available at http://daac.ornl.gov/VEGETATION/guides/ global fire emissions v2.1.html.



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Fig. 3. Monthly MODIS active fire counts and biomass burning emissions of BC (unit: $g m^{-2}$) in the WUS for July, August, and September 2006, respectively: **(a)** MODIS active fire counts, **(b)** standard GFEDv2 BC emissions, **(c)** GFEDv2 BC emissions adjusted spatiotemporally based on the active fire counts (see text for details on the adjustments), and **(d)** the difference between **(c)** and **(b)**.













Fig. 5. Observed (red line) and simulated daily surface BC concentrations ($\mu g m^{-3}$) at IMPROVE sites for May–October 2006, averaged for four altitude ranges: below 1 km (averages at 18 sites), 1–2 km (averages at 30 sites), 2–3 km (averages at 18 sites), and above 3 km (averages at 3 sites). Model results are from simulations at 2° × 2.5° horizontal resolution and with the standard (black line) and the adjusted (green line; see Figs. 1 and 2) GFEDv2 emissions.







Fig. 6. The averaging kernels for inversions (May–October 2006) of BC emissions in the WUS, with each line corresponding to an emission source or source region: biomass burning emissions in the Rockies (BBRM, black line), biomass burning emissions in California and the Southwest (BBCSW, red line), biomass burning emissions in the Pacific Northwest (BBPNW, green line), and anthropogenic emissions in the Western US (ANTHWUS, blue line). Results here are from simulations at $2^{\circ} \times 2.5^{\circ}$ (solid line) and at $0.5^{\circ} \times 0.667^{\circ}$ (dotted line) horizontal resolutions and with best set of error characterizations (30 % for observations and 500 % for biomass burning emissions; bottom two rows in Table 1).







Fig. 7. Monthly BC emissions in the WUS for May–October 2006: a priori BC emissions (pink); a posteriori emissions of biomass burning BC from the Rockies (BBRM), from California and the Southwest (BBCSW), and from the Pacific Northwest (BBPNW), and a posteriori emissions of anthropogenic BC from the WUS (ANTHWUS). A posteriori emissions from inversions at $2^{\circ} \times 2.5^{\circ}$ (blue, green, orange) and at $0.5^{\circ} \times 0.667^{\circ}$ (red) horizontal resolutions, color-coded by error characterizations (see text for details), are shown. For the purpose of clarity, anthropogenic emissions are divided by three in the figures. Error bars represent estimated uncertainties of the emissions.







Fig. 8. Observed (red line) and simulated daily surface BC concentrations (μ gm⁻³) at four IMPROVE sites in the Rockies for May–October 2006. Values shown are daily averages for every three days. Shown are results from simulations with the standard GFEDv2 emissions (black line, 2° × 2.5°) and with the a posteriori emissions (green line, 2° × 2.5°; pink line, 0.5° × 0.667°).





Fig. 9. Same as Fig. 8, but for three IMPROVE sites in the Pacific Northwest.





Fig. 10. Same as Fig. 8, but for three IMPROVE sites in California and the Southwest.

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Fig. 11. Observed (red line) and simulated daily surface BC concentrations (μ gm⁻³) averaged at IMPROVE sites in the Rockies (top left panel, 31 sites), in California and the Southwest (top right panel, 28 sites), and in the Pacific Northwest (bottom panel, 10 sites) for May–October 2006. Shown are results from simulations with the standard GFEDv2 emissions (black line, $2^{\circ} \times 2.5^{\circ}$) and with the a posteriori emissions (green line, $2^{\circ} \times 2.5^{\circ}$; pink line, $0.5^{\circ} \times 0.667^{\circ}$).







Fig. 12. Same as Fig. 5, but for simulations with the standard GFEDv2 emissions (black line, $2^{\circ} \times 2.5^{\circ}$) and with the a posteriori emissions (green line, $2^{\circ} \times 2.5^{\circ}$; pink line, $0.5^{\circ} \times 0.667^{\circ}$).





Fig. 13. Taylor diagram and Taylor scores for simulations with the standard GFEDv2 emissions (red dot, $2^{\circ} \times 2.5^{\circ}$), with the adjusted GFEDv2 emissions (blue dot, $2^{\circ} \times 2.5^{\circ}$), and with the a posteriori emissions (green dot, $2^{\circ} \times 2.5^{\circ}$; pink dot, $0.5^{\circ} \times 0.667^{\circ}$) for May–October 2006. Values are averages for the 69 IMPROVE sites in the WUS (Fig. 1).





Fig. 14. Same as Fig. 13, but for the 69 individual IMPROVE sites in the WUS (Fig. 1). The standard deviation for each site is normalized by the standard deviation of the observations at that site.

