

**Biomass burning
contribution to black
carbon**

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Biomass burning contribution to black carbon in the western United States mountain ranges

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Abstract

Forest fires are an important source to carbonaceous aerosols in the western United States (WUS). We quantify the relative contribution of biomass burning to black carbon (BC) in the WUS mountain ranges by analyzing surface BC observations for 2006 from the Interagency Monitoring of PROtected Visual Environment (IMPROVE) network using the GEOS-Chem global chemical transport model. Observed surface BC concentrations show broad maxima during late June to early November. Enhanced potassium concentrations and potassium/sulfur ratios observed during the high-BC events indicate a dominant biomass burning influence during the peak fire season. Model surface BC reproduces the observed day-to-day and synoptic variabilities in regions downwind of and near urban centers. Major discrepancies are found at elevated mountainous sites during the July–October when simulated BC concentrations are biased low by a factor of two. We attribute these biases largely to the underestimated and temporally misplaced biomass burning emissions of BC in the model. Additionally, we find that the biomass burning contribution to surface BC concentrations in the US likely was underestimated in a previous study using GEOS-Chem (Park et al., 2003), because of the unusually low planetary boundary layer (PBL) heights and weak precipitation in the GEOS-3 meteorological reanalysis data used to drive the model. PBL heights from GEOS-4 and GEOS-5 reanalysis data are comparable to those from the North American Regional Reanalysis (NARR). Model simulations show improved agreements with the observations when driven by GEOS-5 reanalysis data, but model results are still biased low. The use of biomass burning emissions with diurnal cycle, synoptic variability, and plume injection has relatively small impact on the simulated surface BC concentrations in the WUS.

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1 Introduction

Black Carbon (BC) is a product of incomplete combustion of carbonaceous fuels (Bond et al., 2004). It is strongly absorptive of solar radiation and has considerable impacts on global climate (Flanner et al., 2007, 2009; IPCC, 2007; Hansen and Nazarenko, 2004; Jacobson, 2001, 2004). BC deposited on snow and ice can significantly decrease the surface albedo (Warren and Wiscombe, 1980). The reduced snow albedo enhances surface snowmelt (Flanner et al., 2007; Zwally et al., 2002) and can potentially change the regional hydrological cycle over mountain ranges (e.g., Qian et al., 2009). Globally the direct radiative forcing due to fossil fuel BC was estimated to be $+0.2 \pm 0.15 \text{ W m}^{-2}$ and the radiative forcing of snow/ice albedo effect due to BC was $+0.1 \pm 0.1 \text{ W m}^{-2}$ (IPCC, 2007). Freshly emitted BC is mostly hydrophobic and becomes hydrophilic by oxidation or by coating with sulfate and organics in about 1–2 days (Park et al., 2003 and references therein). BC is removed from the atmosphere within days to weeks primarily by wet deposition (Jacobson, 2004). Because of its shorter lifetime relative to long-lived greenhouse gases such as CO_2 , BC shows a much stronger regional warming effect and its reduction may provide an efficient short-term solution to combat global warming (Ramana et al., 2010; Ramanathan and Carmichael, 2008; Bond and Sun, 2005; Hansen et al., 2005).

Globally, the annual emissions of BC are mainly from three sources: about 40 % from fossil fuels, 40 % from biomass burning and 20 % from biofuels (Bond et al., 2004; Cooke et al., 1999). The uncertainty in current BC emission estimates ranges from at least $\pm 50\%$ on global scales to a factor of 2–5 on regional scales (Ramanathan and Carmichael, 2008; Streets et al., 2001, 2003). The most recent generation of fire emission inventories is based on a combined approach using burned area and active fire counts from satellites, accompanied by biogeochemical modeling of the available fuel load (van der Werf et al., 2006, 2010; Langmann et al., 2009). Thus, biomass burning emissions were calculated as the product of burned area, fuel loads, combustion completeness and emission factors (van der Werf et al., 2006, 2010). Even though fire

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emission inventories have improved considerably in recent years, large uncertainties remain in the temporal and spatial variations 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. 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 for or under-detection of agricultural burnings in satellite active fire detection algorithms may be another large uncertainty (van der Werf et al., 2010; Korontzi et al., 2006).

Recent studies have shown that the transport and subsequent deposition of BC in the western United States (WUS) mountain ranges may significantly impact the region's climate and hydrological cycle. In the WUS, mountain snowmelt accounts for more than 70 % of the annual stream flows (Qian et al., 2009). A modeling study by Qian et al. (2009) using the WRF-Chem model showed that the BC deposition over the WUS mountain ranges led to increased rain but less snow accumulation in winter. This change in the precipitation pattern resulted in reduced and earlier snowmelt in spring. Consequently runoff from snowmelt between April and June decreased, adversely affecting the supply of fresh water in the western states.

It is thus imperative to better understand the sources, transport, and deposition of BC in the WUS mountain ranges. BC in this region is mainly from North American anthropogenic emissions (Park et al., 2003), transpacific transport of Asian emissions especially during spring (Chin et al., 2007; Hadley et al., 2007; Park et al., 2003), and North American biomass burning emissions during the summer and fall fire season (Spracklen et al., 2009; Park et al., 2003). However, the relative contributions from these sources particularly biomass burning to BC in the WUS are still uncertain. Wildfires are an important source to carbonaceous aerosols in the WUS (Zeng et al., 2011; Spracklen et al., 2007, 2009; Jaffe et al., 2008; Park et al., 2007). The increase of fire frequency and prolonged fire seasons observed in the WUS in recent decades have been linked to increased spring and summer temperatures and an earlier spring snowmelt (Westerling et al., 2006). The modeling study by Spracklen et al. (2009)

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showed that the annual mean area burned in the WUS could increase by 54 % by the 2050s relative to the present under future warming. It is conceivable that fires will be an even larger contributor to BC in the WUS in the 21st century, especially considering that North American anthropogenic emissions are trending down due to aggressive emission reduction regulations (Ramanathan and Carmichael, 2008; Bond et al., 2007; Novakov et al., 2003). For the transpacific transport of Asian emissions, it is likely that Asian BC emissions will continue to increase in the coming decades due to the rapid economic developments in that region (Zhang et al., 2009; Bond et al., 2007; IPCC, 2007; Streets et al., 2003; Novakov et al., 2003). The large uncertainties in BC emissions again warrant better understanding of the sources of BC in the WUS mountain ranges.

The goal of the present study is to improve our understanding of the sources, transport, and deposition of BC in the WUS mountain ranges. Our approach is to apply a global three-dimensional (3-D) chemical transport model (CTM) to analyze surface BC observations over the WUS. We intend to quantify the relative contributions from the different source types and source regions to surface BC concentrations in the WUS mountain ranges. Our focus is on the contributions from fires. Our analysis centers on 2006, a relatively strong fire year in terms of burned area in temperate North America (Giglio et al., 2010). We describe the observations and the GEOS-Chem global 3-D CTM in Sect. 2. We present our results and related discussions in Sect. 3. Conclusions are given in Sect. 4.

2 Observation and model description

2.1 IMPROVE

Long-term measurements of aerosols with chemical species including BC and elemental components of potassium (K) and sulfur (S) are available in the US from the Interagency Monitoring of PROtected Visual Environment (IMPROVE) network since 1987

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for the protection of visibility in Class I remote areas (Malm et al., 1994; data available at <http://vista.cira.colostate.edu/improve/>). Figure 1 shows 67 IMPROVE sites in the WUS. These are remote sites at various elevations. IMPROVE measurements are made every three days for 2006 and twice a week for 1998. 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 organic carbon (OC) and BC at different temperatures (Chow et al., 2004). The uncertainties of the TOR method are difficult to quantify (Park et al., 2003; Chow et al., 1993).

Tanner et al. (2001) showed that the surface concentrations of K and the K/S ratios significantly increased during wildfire episodes and were therefore good tracers of biomass burning. IMPROVE observations of K and K/S are thus particularly useful for identifying fire influence. Additionally, the IMPROVE data also provides surface soil dust concentrations that were calculated as the sum of the soil-derived elements (Al, Si, K, Ca, Ti, Fe) and their normal oxides (Malm et al., 1994). The primary natural dust is wind-blown mineral dust while the main anthropogenic dust is road dust that contains carbon and metals (Wells et al., 2007; Kim et al., 2005). Previous studies have shown that surface dust concentrations in the WUS in spring were influenced by not only local sources (Hwang and Hopke, 2007; Wells et al., 2007) but also the transpacific transport of Asian dust (VanCuren and Cahill, 2002; Husar et al., 2001). Therefore, a combination of high dust concentrations and relative low K concentrations and K/S ratios during relatively high-BC events in the WUS in spring indicates an anthropogenic rather than a fire influence on surface BC concentrations.

2.2 GEOS-Chem description and simulations

GEOS-Chem is a global 3-D CTM driven by assimilated meteorological observations from the NASA Goddard Earth Observing System (GEOS) (Bey et al., 2001). We use here GEOS-Chem version 8-01-04 (available at <http://acmg.seas.harvard.edu/geos/>) driven by GEOS-3, GEOS-4, and GEOS-5 meteorological fields with 6-h temporal resolution (3-h for surface variables and mixing depths), 2° latitude × 2.5° longitude

horizontal resolution, and 30 (GEOS-3, 4) or 47 (GEOS-5) vertical layers from the surface to 0.01 hPa. The lowest model levels are centered at approximately 10, 50, 100, 200, 350, 600, 850, 1250, and 1750 m above sea level in GEOS-3, 60, 250, 600, 1200, 2000 m in GEOS-4, and 60, 200, 300, 450, 600, 700, 850, 1000, 1150, 1300, 1450, 1600, 1800 m in GEOS-5.

Tracer advection is computed every 15 min with a flux-form semi-Lagrangian method (Lin and Rood, 1996). Tracer moist convection is computed using GEOS convective, entrainment, and detrainment mass fluxes as described by Allen et al. (1996a, b). The deep convection scheme of GEOS-4 is based on Zhang and McFarlane (1995), and the shallow convection treatment follows Hack (1994). GEOS-3 and GEOS-5 convection is parameterized using the relaxed Arakawa-Schubert scheme (Moorthi and Suarez, 1992; Arakawa and Schubert, 1974).

GEOS-Chem simulation of carbonaceous aerosols has been reported previously by Park et al. (2003). Eighty percent of BC and 50 % of organic carbon (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). Global fossil fuel and biofuel emissions of BC are based upon Bond et al. (2004) with updated emissions for Asia (Zhang et al., 2009) and North America (Cooke et al., 1999).

Biomass burning emissions of BC are from version 2 of the Global Fire Emissions Database (GFEDv2) (Randerson et al., 2007; van der Werf et al., 2006). GFED was derived using satellite observations including active fire counts and burned areas in conjunction with a biogeochemical model. Carbon emissions were calculated as the product of burned area, fuel loads and combustion completeness. Burned area was derived using active fire and 500-m burned area datasets from the Moderate Resolution Imaging Spectroradiometer (MODIS) as described by Giglio et al. (2006). Figure 2 shows the monthly mean total carbon emissions in the WUS (100–125° W, 30–50° N) for 2006. The fire season started in April and lasted through November. The GFEDv2 inventory has a multitude of temporal resolutions from monthly, 8-day, to 3-hourly with

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diurnal cycles, as reported previously by Chen et al. (2009). We provide a brief summary here. The 8-day emissions were re-sampling of the standard GFEDv2 monthly emissions to an 8-day time step according to MODIS 8-day active fire counts. To account for the strong diurnal cycles of forest fires (Giglio et al., 2006), 3-hourly diurnal coefficients were then multiplied with the 8-day emissions to derive a diurnal GFEDv2 inventory. Additionally, the variations of synoptic weather conditions may influence forest fires and the associated emissions. Such synoptic, day-to-day variability was superimposed onto the diurnal inventory. The resulting synoptic GFEDv2 inventory thus combined both diurnal and synoptic variations.

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.

For the present study, we conducted GEOS-Chem “offline” carbonaceous aerosols simulations (Park et al., 2003) for 2006, driven by GEOS-4 and GEOS-5 meteorological fields. We use GFEDv2 8-day emissions unless stated otherwise. In addition, we conducted two model simulations for 1998 driven by GEOS-3 and by GEOS-4 meteorological fields. In the last two simulations, emissions including those from biomass burning were exactly the same as those used by Park et al. (2003). Detailed discussions and justifications for these model simulations are provided in the following sections where appropriate.

3 Results and discussions

3.1 Seasonal and daily variations of surface BC

Figure 3 compares the seasonal variations of simulated and observed daily surface BC concentrations during 2006 at selected IMPROVE sites. Values shown are daily averages for every three days. Model results shown here are from simulations driven

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by GEOS-4 data with GFEDv2 8-day emissions unless stated otherwise. We sampled model results at the time and location of IMPROVE observations. In addition to a standard simulation where all emissions were included, we also conducted sensitivity simulations by shutting off separately sources of BC from North American anthropogenic emissions, Asian anthropogenic emissions, and global biomass burning emissions. The differences between results from the standard simulation and those from the sensitivity simulations therefore represent the contributions to surface BC concentrations from the aforementioned BC source types and source regions. These relative contributions are also shown in Fig. 3. GEOS-Chem BC reproduces both the synoptic variability and magnitudes of surface BC concentrations at sites downwind of urban areas and near urban centers. Figure 3 includes two such sites, Meadview, AZ (36.0° N, 114.1° W, 0.90 km) (Fig. 3a) and San Gabriel, CA (34.3° N, 118.0° W, 1.79 km) (Fig. 3b). The San Gabriel site is on the northern edge of the Los Angeles Basin. Meadview is about 100 miles to the east of Las Vegas. North American anthropogenic emissions dominate at these sites as seen in the model results. The agreements at these sites indicate that North American anthropogenic BC emissions in the model are reasonably prescribed.

Both the observations and model results show broad maxima of surface BC concentrations during summer and fall at some sea-level sites (e.g. Fig. 3c) and at elevated sites (Fig. 3d–p). The seasonal variations of BC vary considerably from site to site. Some of the highest BC concentrations are seen during August and September at most of the sites. Relatively small fires (in terms of BC emissions) are apparent in late April and early May at sites such as Kalmiopsi, OR (42.6° N, 124.1° W, 0.08 km) and Three Sisters, OR (44.3° N, 122.0° W, 0.89 km). At the 0–1 km altitude range, model results reproduce largely the peaks of BC concentrations observed during the fire season at some IMPROVE sites, e.g., Kalmiopsi, OR and North Cascades, WA (48.7° N, 121.1° W, 0.57 km). At elevated sites, however, model results significantly underestimate surface BC concentrations by a factor of at least two during summer and fall (Fig. 3e–p). The discrepancies exist not only in the magnitudes of BC concentrations but also in the timing of the enhanced BC concentrations. In particular, some

of the observed large enhancements due to biomass burning during June and July are completely missing in the model results. For example, at sites Hells Canyon, OR (45.0° N, 116.8° W, 0.66 km) and Three Sisters, OR, observed large enhancements to the surface BC are in middle July. Though simulated BC concentrations show small yet significant relative enhancements, the large peaks of simulated BC concentrations do not occur until September. The discrepancies are larger at the 1–3 km altitude range. The simulated BC concentrations at IMPROVE sites Flathead, MT (47.8° N, 114.3° W, 1.58 km), Craters Moon, ID (43.5° N, 113.6° W, 1.82 km), and Mt. Gates, MT (46.8° N, 111.7° W, 2.39 km) are biased low by a factor of three or more. Part of the discrepancies is because of the model resolution, which is too coarse to resolve fine regional distributions of BC. Comparing localized observations with model results that are representative of a much larger area is inherently problematic.

Figure 4 compares the observed and model simulated daily surface BC concentrations averaged for sites at the altitude ranges 0–1, 1–2, 2–3, and 3–4 km, respectively. Model results shown here are from simulations driven by GEOS-4 meteorological data and with 8-day GFEDv2 emissions. Again, Fig. 4 shows significantly underestimated surface BC concentrations in the model during summer and fall, especially at the 1–2 km and 2–3 km altitudes. The contributions to surface BC concentrations in the WUS from North American anthropogenic emissions show rather small variations throughout the year at all four altitude ranges. Figures 3 and 4 also show small yet significant relative enhancements of BC concentrations (up to 50%) during February to March and April to early May. These enhancements are particularly evident at the 0–1 and 1–2 km altitude ranges and to a lesser degree at 2–3 km. Our model results show that these enhancements are dominated by North American anthropogenic emissions but with significant contributions from Asian anthropogenic emissions. This relatively large Asian influence during this time of the year is consistent with our understanding that the transpacific transport of Asian pollution is strongest in spring (Jacob et al., 2010; Report of National Research Council, 2009; Liu et al., 2005, 2003).

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3.2 BC correlations with K, K/S, and dust

In this section we examine the correlations between surface BC and K, K/S, and soil dust to further verify the large influence of biomass burning on surface BC concentrations in the WUS during summer and fall. As an example, Fig. 5 shows time series of observed surface concentrations of BC and K as well as K/S ratios at the IMPROVE site Flathead, MT. The large BC concentration peaks ($0.4\text{--}1\ \mu\text{g m}^{-3}$) during August–October are strongly correlated with either high K concentrations (up to $0.3\ \mu\text{g m}^{-3}$) or high K/S ratios (up to 0.9) or both. We find similar strong correlations during the summer and fall fire season at most of the mountainous IMPROVE sites (not shown). These correlations suggest that biomass burning emissions dominate the broad maxima of surface BC concentrations in summer and fall. That biomass burning emissions are the dominant source to surface BC concentrations in the WUS mountain ranges during summer and fall is consistent with our model results (Sect. 3.1).

Also shown in Fig. 5 are time series of IMPROVE surface soil dust concentrations. The relatively high surface BC concentrations (up to $0.4\ \mu\text{g m}^{-3}$) during middle March and April to early May correspond with high soil dust concentrations (up to $2\ \mu\text{g m}^{-3}$) and relatively low K concentrations and K/S ratios. As discussed in Sect. 2.1, the strong BC-soil dust correlations thus indicate significant anthropogenic contributions to the surface BC at the site. Our examination of BC-soil dust correlations at the other IMPROVE sites shows similar results (not shown). The significant springtime anthropogenic contributions to the surface BC in the WUS mountain ranges are in agreements with our model results that show dominant contributions from North American anthropogenic emissions (Sect. 3.1).

As discussed in Sect. 3.1, North American anthropogenic emissions prescribed in the model appear to be reasonable. The broad maxima of surface BC concentrations in the WUS mountain ranges during summer and fall are dominated by biomass burning emissions. Thus the large discrepancies between our model results and the observations in the summer and fall fire season can be attributed in large part to the

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biomass burning emissions of BC being underestimated in the model. In addition, the discrepancies in the timing of the observed and simulated surface BC enhancements suggest that the uncertainties of biomass burning emissions of BC are not only in the magnitudes of fire emissions but also likely in the timing and location of fires.

3.3 Sensitivity of surface BC to PBL height and precipitation

A previous study by Park et al. (2003) using the GEOS-Chem model driven by GEOS-3 reanalysis data estimated the contribution of Asian emissions to surface BC concentrations in the US in 1998. Fire activities in temperature North America were considerably weaker in 1998 than in 2006 in terms of burned area (Giglio et al., 2010). Their model results showed very good agreements with IMPROVE BC observations ($r^2 > 0.8$) including those in the summer and fall fire season. Interannual biomass burning emissions in that study were from Duncan et al. (2003). To reconcile the apparent differences between results from our simulation driven by GEOS-4 data and those of Park et al. (2003), we conducted a simulation for 1998 using the same GEOS-Chem configurations, including GEOS-3 reanalysis data and the various emissions (biomass burning included), as used by Park et al. (2003) (the line “GEOS-3 Interannual” in Fig. 6). In addition, we also conducted a model simulation for the same year but driven by GEOS-4 data and with the same Duncan et al. (2003) biomass burning emissions as used by Park et al. (2003) (the line “GEOS-4 Interannual” in Fig. 6). Figure 6 compares the monthly mean surface BC concentrations for June–December 1998 from these two simulations against IMPROVE observations at two IMPROVE sites Mt. Rainier, WA (46.8° N, 122.1° W, 0.44 km) and Three Sisters, OR. Also shown in Fig. 6 are results from our standard model simulation driven by GEOS-4 data and with GFEDv2 8-day biomass burning emissions (the line “GEOS-4 GFEDv2 8-day” in Fig. 6). We are able to reproduce the results reported by Park et al. (2003). The results from the simulation driven by GEOS-3 data are in good agreements with IMPROVE observations. The results from the two simulations driven by GEOS-4 data are very similar, despite the different biomass burning emissions used. However, the results from both of these

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simulations show considerably lower surface BC concentrations than those from the simulation driven by GEOS-3 meteorological data and from IMPROVE observations.

Part of the discrepancy seen in Fig. 6 can be attributed to the different planetary boundary layer (PBL) heights. We compare PBL heights from GEOS-3 and GEOS-4 against those from NCEP North American Regional Reanalysis data (NARR) (Mesinger et al., 2006; data available at <http://www.esrl.noaa.gov/psd/cgi-bin/data/narr/plothour.pl>). The NARR data have a horizontal resolution of 32 km. We extracted PBL heights at 13:00 local time at IMPROVE sites from all three datasets. Figure 7 shows as an example the comparison for August–September 1998 for Mt. Rainier. PBL heights are considerably lower in GEOS-3 than in NARR. In contrast, GEOS-4 PBL heights are in good agreement with NARR data. Other IMPROVE sites show similar comparisons. It is thus clear that the unusually shallow boundary layer in GEOS-3 data partly results in artificially high surface BC concentrations in the model simulation driven by GEOS-3 reanalysis data hence a false good agreement with IMPROVE observations.

Additionally, we examine the differences of precipitation between GEOS-3 versus GEOS-4 meteorological fields. Figure 8 shows the comparisons for August 1998 over the WUS. We compare monthly mean precipitation from these two reanalysis datasets to those from the Climate Prediction Center Merged Analysis of Precipitation (CMAP) (Xie and Arkin, 1997; available at <http://www.esrl.noaa.gov/psd/data/gridded/data.cmap.html>) and the Global Precipitation Climatology Project (GPCP) (Adler et al., 2003; data available at <http://www.esrl.noaa.gov/psd/data/gridded/data.gpcp.html>). Both CMAP and GPCP are monthly means from combined satellite and station data available since January 1979. The resolution of both CMAP and GPCP data used here is 2.5° latitude × 2.5° longitude. Precipitation is much weaker in GEOS-3 (Fig. 8a) than in both CMAP (Fig. 8c) and GPCP (Fig. 8d). Precipitation in GEOS-4 (Fig. 8b) is in much better agreement with CMAP and GPCP data. The weak precipitation in GEOS-3 data results in weak wet scavenging of BC in the model simulation driven by GEOS-3 data (not shown). The large differences in precipitation between GEOS-3

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and GEOS-4 are mainly due to the different convective precipitations (Fig. 8e and f). These differences are largely explained by the different convective parameterizations – relaxed Arakawa Schubert (Moorthi and Suarez, 1992) in GEOS-3 and the scheme by Zhang and McFarlane (1995) in GEOS-4.

Thus the unusually shallow boundary layer and weak precipitation in GEOS-3 resulted in artificially high surface BC concentrations in the WUS in the GEOS-Chem model. The artificially high BC concentrations largely explain the good agreement between the simulated surface BC concentrations in Park et al. (2003) and IMPROVE observations. Because the US fossil fuel emissions of BC prescribed in GEOS-Chem are reasonable (Sect. 3.1), our model simulations driven by GEOS-4 data therefore suggest that biomass burning emissions of BC were likely significantly underestimated in Park et al. (2003), too.

We conducted an additional GEOS-Chem simulation for 2006, driven by GEOS-5 meteorological fields and with GFEDv2 8-day biomass burning emissions. The results are compared against IMPROVE observations and those from the standard simulation driven by GEOS-4 data. Figure 9 shows surface BC concentrations from model results and IMPROVE observations, averaged for IMPROVE sites at the 0–1, 1–2, 2–3, and 3–4 km altitude ranges. During the fire season, the simulation driven by GEOS-5 data shows slightly improved comparison with the observations, especially at sites in the 0–1 km altitude range (Fig. 9a). Part of this improvement is because of the better-resolved boundary layer in GEOS-5 than in both GEOS-4 and GEOS-3 (Sect. 2.2). The two model simulations driven by GEOS-5 and by GEOS-4 meteorological fields show very similar results at the higher altitude ranges. Both model results are biased low, particularly at the 1–2 and 2–3 km during the fire season (Fig. 9b, c). The largest discrepancies are seen at 1–2 km.

Again, we compare the PBL heights and precipitation fields between GEOS-4 and GEOS-5 at IMPROVE sites. As an example, Fig. 10 compares PBL heights from GEOS-4 and GEOS-5 with NARR data at Mt. Rainier for August and September 2006. Both GEOS-4 and GEOS-5 PBL heights are in reasonable agreements with NARR

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data. Figure 11 compares GEOS-4 and GEOS-5 precipitation fields against CMAP and GPCP data. In the WUS, precipitation is significantly larger in GEOS-4 than in CMAP and GPCP for August 2006. In contrast, GEOS-5 precipitation is weaker than those in CMAP and GPCP. Further examination shows that the precipitation in the WUS during August 2006 is predominantly convective precipitation, both in GEOS-4 and GEOS-5. The convective precipitation is considerably stronger in GEOS-4 (Fig. 11e) than in GEOS-5 (Fig. 11f). The different precipitation led to different wet scavenging of BC in the WUS (not shown).

3.4 Sensitivity of surface BC to improved and finer temporally-resolved biomass burning emissions

Giglio et al. (2006) showed that the burned area estimated in GFEDv2 had low biases of 17 % in Alaska and 30 % in western Canada. In a recent modeling study using GEOS-Chem and GFEDv2 emissions, Chen et al. (2009) scaled up GFEDv2 emissions of CO and BC by 20 % over North America to correct for these low biases. We conducted a sensitivity GEOS-Chem simulation driven by GEOS-4 data where we scaled the 8-day GFEDv2 total carbon emissions by a factor of 1.5 globally. Figure 12 compares the results with IMPROVE observations. Near linear increases in the model simulated surface BC concentrations are evident. The largest increases are at the 1–2 km altitude ranges in the summer and fall fire season (Fig. 12b). The increased emissions have rather small impacts on the simulated surface BC concentrations at the 0–1 km (Fig. 12a) and 3–4 km (Fig. 12d) altitude ranges. However, simulated BC concentrations are still significantly lower than IMPROVE observations. Clearly not only the total biomass burning emissions of BC as prescribed in the model are likely too low but also the spatiotemporal distributions of the emissions are less than accurate. Small fires are likely a major source of uncertainty in the estimates of biomass burning emissions of BC (Giglio et al., 2006, 2009, 2010). Since agricultural burnings are usually small fires therefore difficult to detect from space, agricultural burnings may be another large uncertainty (van der Werf et al., 2010; Korontzi et al., 2006). Furthermore, that some

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fires were obscured by clouds or vegetation, or were not actively burning at the time of the satellite overpass introduces additional uncertainties (Giglio et al., 2009).

Chen et al. (2009) have also shown that finer temporally-resolved biomass burning emissions had significant impacts on GEOS-Chem simulated surface carbon monoxide and BC concentrations, especially in the biomass burning source regions in Alaska and western Canada. We conducted two simulations driven by GEOS-4 and by GEOS-5 reanalysis data, both with synoptic GFEDv2 emissions. Additionally, we conducted two simulations driven by GEOS-4 and by GEOS-5 reanalysis data, but with 8-day GFEDv2 emissions. All four simulations are for 2004, the last year for which both GEOS-4 and GEOS-5 data are available to us. Figure 13 compares the results with IMPROVE observations from April to December 2004. Since 2004 is a relatively weak fire year in terms of burned area in temperate North America (Giglio et al., 2010), the agreement between the model simulations and IMPROVE observations is better in 2004 than in 2006. With the synoptic GFEDv2 emissions, model results are in slightly better agreements with the observations. The largest improvements are seen at below 2 km altitudes, but model results still underestimate surface BC concentrations during the fire season. Results from the simulation driven by GEOS-5 meteorological data and with GFEDv2 synoptic emissions are in slightly better agreement with IMPROVE observations. The results from the simulations driven by GEOS-4 and by GEOS-5 on average are comparable and almost indistinguishable.

3.5 Sensitivity of surface BC to vertically injected biomass burning emissions

Ample evidence has shown that biomass burning smoke plumes can be injected into the free troposphere (Mims et al., 2010; Kahn et al., 2008). Modeling studies also showed that vertically injected biomass burning emissions can significantly improve model comparisons with the observations (Leung et al., 2007; Turquety et al., 2007). To investigate the impact of smoke plume vertical injection on the surface BC, we conducted two simulations with vertical injection of 8-day GFEDv2 emissions. In the first simulation, GFEDv2 emissions were evenly distributed throughout the PBL. Obviously,

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this approach underestimated emissions injected into the free troposphere. In the second simulation, GFEDv2 emissions were uniformly (in mass mixing ratio) distributed throughout the tropospheric column up to 200 hPa. This approach represented an extreme scenario in which certain percentages of emissions from each forest were injected to the middle and upper troposphere. We find that the inclusion of plume vertical injection has relatively small impact on the simulated surface BC concentrations in the WUS mountain ranges during the fire season (not shown). As expected, the simulated summer-fall BC concentrations from the second simulation show significant decreases compared with the first simulation at sites below 2 km: Sula Peak, MT(45.9° N, 114.0° W, 1.90 km), Trinity, CA (40.8° N, 122.8° W, 1.01 km), Starkey, OR (45.2° N, 118.5° W, 1.26 km), and North Sawtooth, ID (44.2° N, 114.9° W, 1.99 km) (not shown) simply because of more emissions are injected to above the boundary layer.

3.6 Sources of surface BC in the WUS mountain ranges

Table 1 shows the annual and seasonal contributions to surface BC concentrations at IMPROVE sites in the WUS in 2006 from North American anthropogenic emissions, transpacific transport of Asian anthropogenic emissions, and global biomass burning emissions. The contributions are shown both as relative contributions and percentages. The results are from the simulations driven by GEOS-4 data and with 8-day GFEDv2 emissions. Annually, the contributions to surface BC concentrations from North American anthropogenic emissions, Asian anthropogenic emissions, and global biomass burning emissions account for 79.0 %, 9.4 %, and 9.9 %, respectively. It is important to point out that the contributions from global biomass burning emissions are significantly underestimated in our model results, both annually and during the summer and fall fire season. We will discuss these three source types and source regions separately in the following paragraphs.

Among the three sources, North American anthropogenic emissions provide the dominant contributions year-round and are relatively invariable throughout the year, consistent with previous studies (e.g., Park et al., 2003). The observed surface BC concentrations in winter and spring were considerably lower in 2006 (this study) than

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in 1998 (Park et al. (2003) study). The decreases likely reflect the reduction in anthropogenic BC emissions in North America during the 8-yr span (Ramanathan and Carmichael, 2008; Bond et al., 2007; Novakov et al., 2003). North American anthropogenic emissions show larger contributions to surface BC concentrations at the lower altitudes close to the surface BC source. The annual percentages of surface BC concentrations from North American anthropogenic emissions are 83.2 % at the 0–1 km altitude range and 67.7 % at 3–4 km, respectively.

Hadley et al. (2007) used Chemical Weather Forecast System (CFORS) model to estimate the transpacific transport of Asian BC in North America. Their simulations showed that, across the 130° W longitude, about 30 % of the BC flux near the surface and more than 75 % of the BC flux above 3 km were from Asia in spring. The percentages of the Asian BC flux across the western Pacific Ocean in spring from our simulation are comparable with the results by Hadley et al. (2007), with about 20 % of the BC flux near the surface and about 80 % of the BC flux at the 3–4 km altitude range are from Asia (not shown). We are also interested in the Asian contribution to surface BC concentrations over the US. Table 1 shows that the percentages of Asian contribution to surface BC concentrations are 7.7–28 % in winter and 13.5–24.4 % in spring 2006 at the 1–4 km altitude range. Figure 14 shows observed and model simulated monthly mean surface BC concentrations at IMPROVE sites (Fig. 1) at different altitudes in April and May 2006. The simulation was driven by GEOS-4 reanalysis data and with GFEDv2 8-day emissions. Also shown are simulated relative contributions and percentages from Asian anthropogenic emissions averaged with altitude. Both Fig. 14 and Table 1 show that the contribution from the transpacific transport of Asian anthropogenic emissions becomes more important with increasing altitude. The annual percentages of Asian contribution are about 9.4 % for all the IMPROVE sites in the WUS and 9–20.5 % for sites at the 1–4 km altitude range. Park et al. (2003) showed that transpacific transport BC from Asian anthropogenic emissions amounted to less than 10 % of the annual surface BC concentrations over the US in 1998. Our results are in broad agreements with those of Park et al. (2003).

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Table 1 shows that the largest contribution from biomass burning emissions is at 40–50° N latitude, the Pacific Northwest where the largest fires in temperate North America tend to occur (Giglio et al., 2010), with an annual contribution of 15 % of surface BC concentrations. Biomass burning emissions are most important at the 1–2 km altitude range in the WUS. The percentages of biomass burning contributions are 11 % averaged in 2006, 14.4 % in summer, and 20.0 % in fall at the 1–2 km altitude range. Our results so far have shown that, the contributions from global biomass burning emissions are significantly underestimated in our model, likely by more than a factor of two during summer and fall. Park et al. (2003) showed that annually about 30 % of surface BC concentrations in the US were from biomass burning. It is likely that their estimate is biased low, too. Recent studies have projected increased fire activity in the WUS in the 21st century (Spracklen et al., 2009; Westerling and Bryant, 2008), which portend to even larger contributions from biomass burning to BC in the WUS.

4 Summary and conclusions

We have used a global 3-D chemical transport model driven by assimilated meteorological data (GEOS-Chem) to examine the sources of the surface black carbon (BC) in the western United States (WUS) mountain ranges. We conducted simulations of BC for 2006 with 2° × 2.5° horizontal resolution and compared model results to surface BC concentrations observed from the IMPROVE network. Sensitivity simulations were used to estimate the relative contributions from North American anthropogenic emissions, Asian anthropogenic emissions, and global biomass burning emissions to surface BC concentrations in the WUS.

Observed concentrations of BC over the WUS showed strong enhancements during summer and fall of 2006. Observed concentrations of potassium and potassium to sulfur ratio, both tracers of biomass burning, indicated that these enhancements of BC concentrations were largely influenced by fire emissions. Model results were strongly sensitive to the assimilated meteorological observations, particularly the boundary

layer schemes and moist processes in the assimilation systems used to generate these meteorological data. The unusually shallow planetary boundary layer and weak precipitation in GEOS-3 significantly and artificially increased model surface BC concentrations in the WUS. In contrast, model simulations driven by GEOS-4 and GEOS-5 meteorological observations with reasonable boundary layer heights and stronger precipitations, showed significantly lower surface BC concentrations. Observed BC concentrations during the summer and fall fire season were often a factor of two higher than the corresponding model results from simulations driven by GEOS-4 and GEOS-5 meteorological data. Largest discrepancies were seen at elevated mountainous sites (above 1 km altitude). Improved temporal variation including diurnal and synoptic variability and plume vertical injection in the biomass burning emission inventory were found to have relatively small impact on the simulated surface BC concentrations at the mountainous IMPROVE sites during the fire season.

Surface BC concentrations in the WUS were dominated by North American anthropogenic emissions year-around (79%). Transpacific transport of Asian anthropogenic emissions became more important with increasing altitude and accounted for about 28% of surface BC concentrations in winter and 24% in spring at the 3–4 km altitude range. The large low bias of model results during summer and fall was a result of the low biomass burning emissions of BC used in the model. Biomass burning emissions contributed about 14–20% at the 1–2 km altitude range to surface BC concentrations during summer and fall, but these estimates were likely biased low by a factor of two.

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Table 1. Annual and seasonal contributions to surface BC concentrations in the western US in 2006 from North American anthropogenic emissions, transpacific transport of Asian anthropogenic emissions, and global biomass burning emissions derived from GEOS-Chem simulations (Units: $\mu\text{g m}^{-3}$).

Time	Altitude (# of sites)	North American anthropogenic emissions (%)	Asian anthropogenic emissions (%)	Global biomass burning (%)
Annual	All sites (69)	0.083 (78.99)	0.010 (9.44)	0.010 (9.85)
	0–1 km (18)	0.115 (83.21)	0.010 (6.91)	0.012 (8.65)
	1–2 km (30)	0.086 (77.91)	0.010 (9.11)	0.013 (11.29)
	2–3 km (18)	0.053 (74.76)	0.010 (14.06)	0.006 (8.67)
	3–4 km (3)	0.031 (67.71)	0.009 (20.45)	0.004 (7.62)
	30–40° N (32)	0.094 (84.15)	0.010 (9.30)	0.005 (4.77)
	40–50° N (37)	0.073 (73.92)	0.009 (9.57)	0.015 (14.85)
	125–115° W (23)	0.100 (78.41)	0.011 (8.54)	0.015 (11.56)
	115–100° W (46)	0.074 (79.38)	0.009 (10.05)	0.008 (8.68)
DJF	All sites	0.083 (88.38)	0.008 (8.30)	0.002 (1.95)
	0–1 km	0.124 (92.08)	0.007 (5.21)	0.003 (1.93)
	1–2 km	0.087 (89.40)	0.007 (7.70)	0.002 (1.70)
	2–3 km	0.046 (79.43)	0.009 (15.29)	0.001 (2.49)
	3–4 km	0.021 (62.01)	0.009 (28.14)	0.001 (4.30)
	MAM	All sites	0.079 (78.27)	0.014 (13.78)
0–1 km		0.109 (82.63)	0.014 (10.31)	0.007 (5.59)
1–2 km		0.082 (78.67)	0.014 (13.51)	0.006 (5.89)
2–3 km		0.052 (71.29)	0.014 (19.39)	0.005 (6.43)
3–4 km		0.034 (63.16)	0.013 (24.39)	0.004 (8.04)
JJA		All sites	0.078 (74.98)	0.011 (10.37)
	0–1 km	0.100 (78.27)	0.011 (8.71)	0.015 (11.39)
	1–2 km	0.082 (73.51)	0.011 (10.28)	0.016 (14.43)
	2–3 km	0.055 (73.33)	0.010 (12.75)	0.009 (11.89)
	3–4 km	0.034 (72.20)	0.008 (16.60)	0.004 (7.92)
SON	All sites	0.092 (75.85)	0.007 (5.91)	0.020 (16.56)
	0–1 km	0.130 (80.32)	0.007 (4.08)	0.023 (14.51)
	1–2 km	0.095 (72.62)	0.007 (5.65)	0.026 (19.96)
	2–3 km	0.059 (75.96)	0.007 (9.51)	0.009 (12.11)
	3–4 km	0.035 (72.28)	0.007 (14.56)	0.004 (9.14)

* Percentage contributions are included in parentheses.

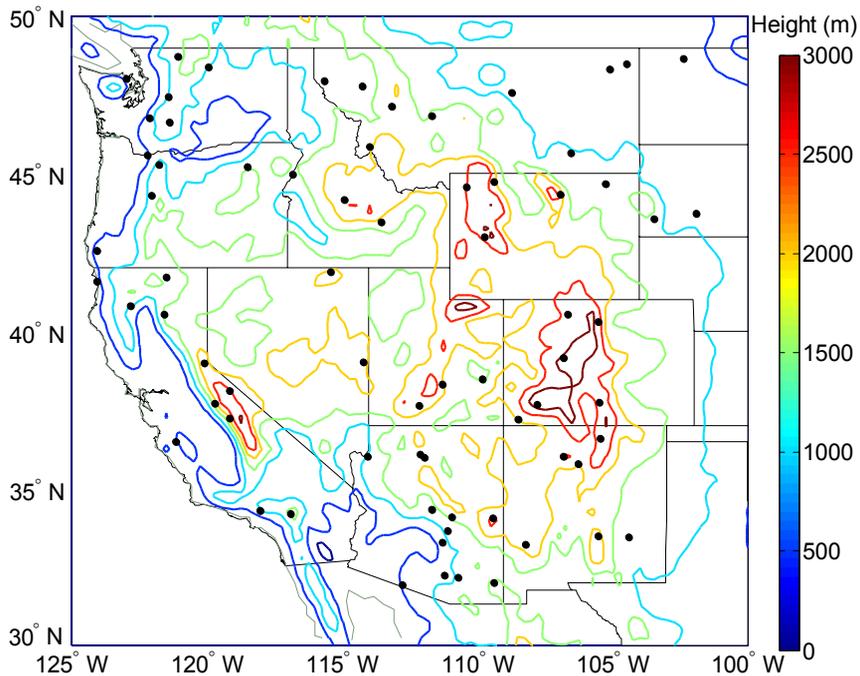


Fig. 1. IMPROVE sites (black dots; data available at <http://vista.cira.colostate.edu/improve/>) in the western US. Also shown are terrain heights (color contours).

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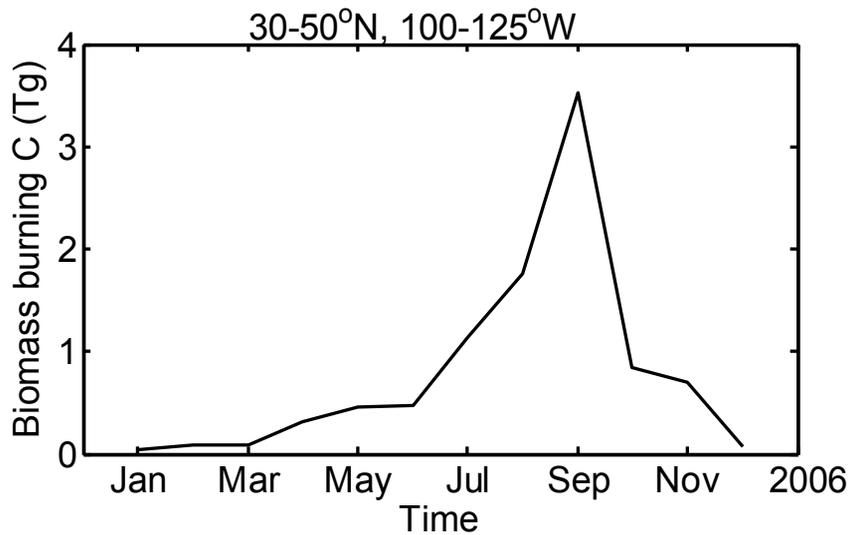


Fig. 2. GFEDv2 monthly total carbon emissions from fires in the western US (100–125° W, 30–50° N) for 2006.

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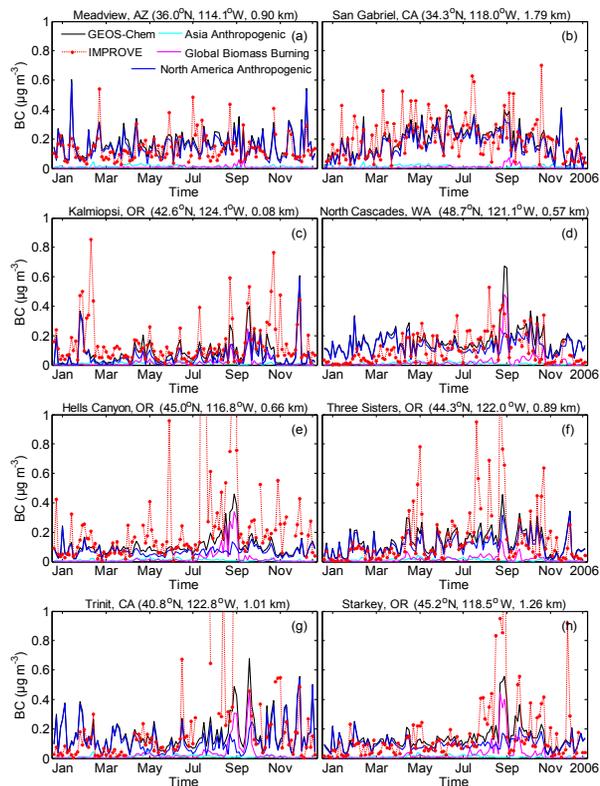


Fig. 3. Simulated (black line) and observed (red line) daily surface BC concentrations at representative IMPROVE sites (Fig. 1) in 2006. Values shown are daily averages for every three days. Simulations are driven by GEOS-4 reanalysis data and with 8-day GFEDv2 emissions. Model results are sampled at the time and location of IMPROVE observations. Also shown are relative contributions to surface BC concentrations from Asian anthropogenic emissions (green line), global biomass burning emissions (pink line), and North American anthropogenic emissions (blue line).

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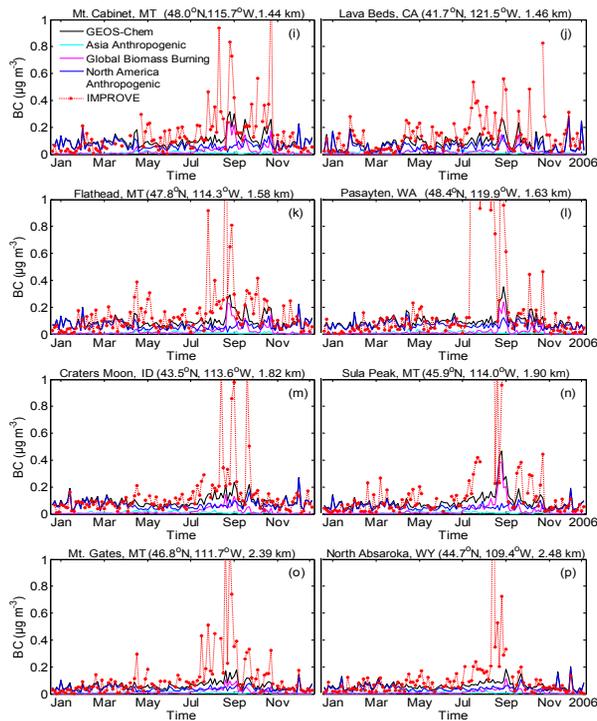


Fig. 3. Continued.

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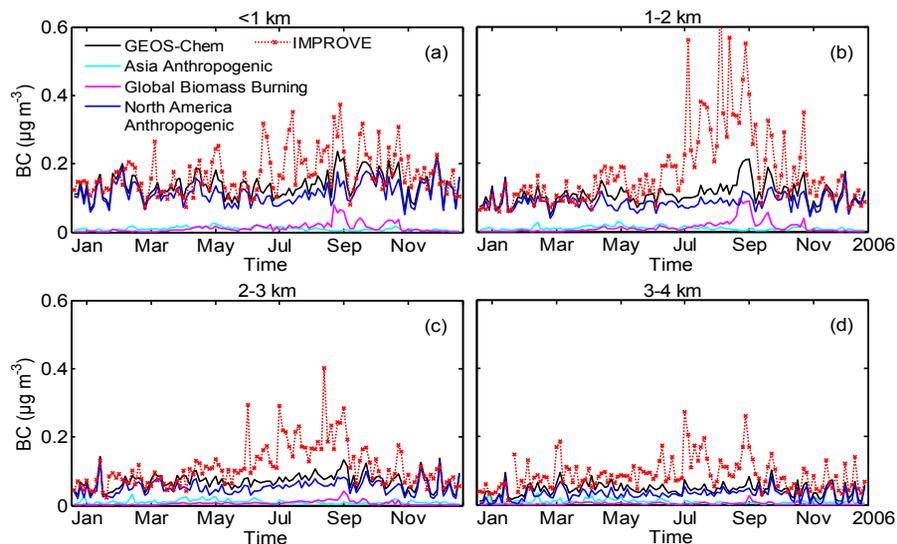


Fig. 4. Simulated (black line) and observed (red dots) daily surface BC concentrations at IMPROVE sites (Fig. 1) for 2006, averaged for four altitude ranges: **(a)** below 1 km (18 sites), **(b)** 1–2 km (30 sites), **(c)** 2–3 km (18 sites), and **(d)** above 3 km (3 sites). Simulations are driven by GEOS-4 reanalysis data and with 8-day GFEDv2 emissions. Also shown are simulated relative contributions to surface BC concentrations from Asian anthropogenic emissions (green line), global biomass burning emissions (pink line), and North American anthropogenic emissions (blue line).

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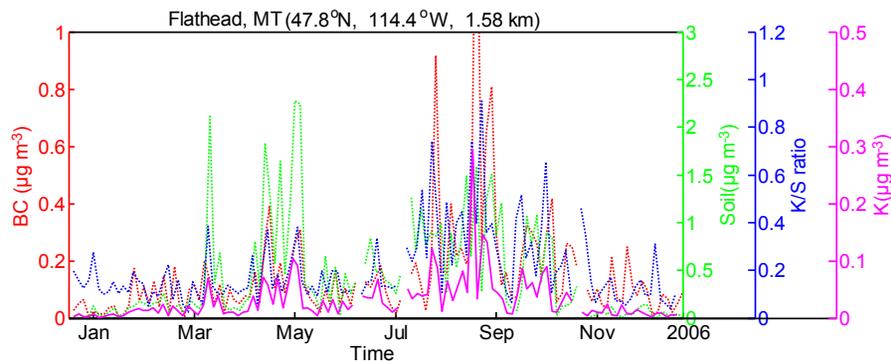


Fig. 5. Daily surface concentrations of BC (red line), soil (green line), potassium (K, pink line), and potassium to sulfur (K/S) ratio (blue line) at IMPROVE site Flathead, MT (47.8° N, 114.3° W, 1.58 km) for 2006.

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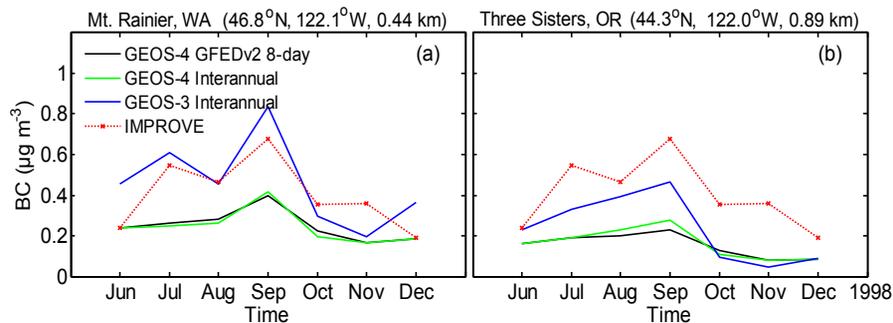


Fig. 6. Monthly mean surface BC concentrations from June to December 1998 at (left) Mt. Rainier, WA (46.8°N , 122.1°W , 0.44 km) and (right) Three Sisters, OR (44.3°N , 122.0°W , 0.89 km). Red lines: IMPROVE observations; black lines: model driven by GEOS-4 data and with 8-day GFEDv2 emissions; green lines: model driven by GEOS-4 reanalysis data and with Duncan et al. (2003) interannual biomass burning emissions; blue lines: model driven by GEOS-3 reanalysis data and with Duncan et al. (2003) interannual biomass burning emissions.

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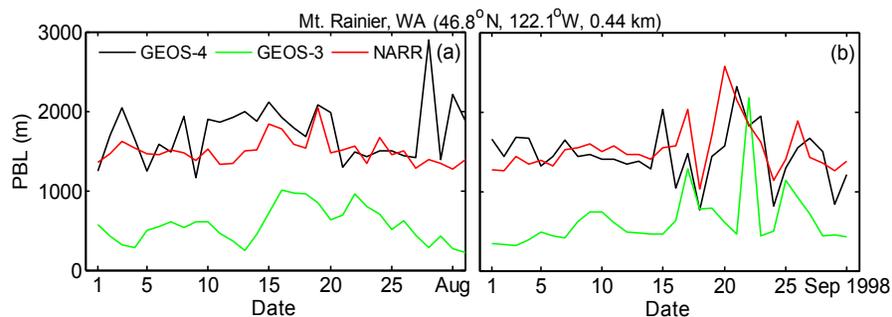


Fig. 7. Planetary boundary layer heights for (left) August and (right) September 1998 at Mt. Rainier, WA (46.8°N , 122.1°W , 0.44 km) (black line, GEOS-4; green line, GEOS-3; red line, North American Regional Reanalysis (NARR; available at <http://www.esrl.noaa.gov/psd/cgi-bin/data/narr/plothour.pl>)). Values are for 13:00 local time.

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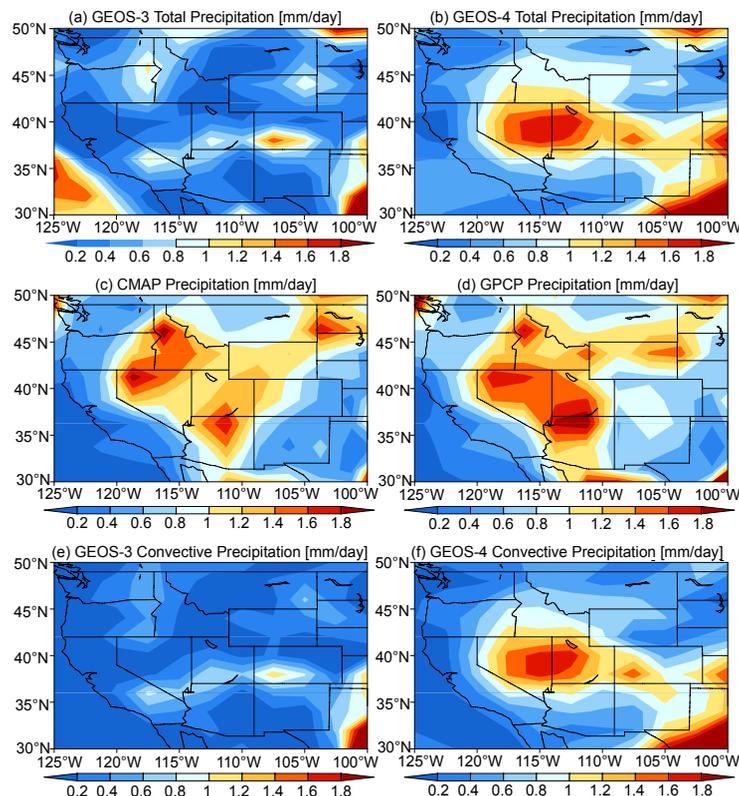


Fig. 8. Monthly mean precipitation for August 1998 in the western US from (a) GEOS-3, (b) GEOS-4, (c) the CPC Merged Analysis of Precipitation (CMAP; available at <http://www.esrl.noaa.gov/psd/data/gridded/data.cmap.html>), and (d) the Global Precipitation Climatology Project (GPCP; available at <http://www.esrl.noaa.gov/psd/data/gridded/data.gpcp.html>). Also shown are monthly convective precipitation from (e) GEOS-3 and (f) GEOS-4.

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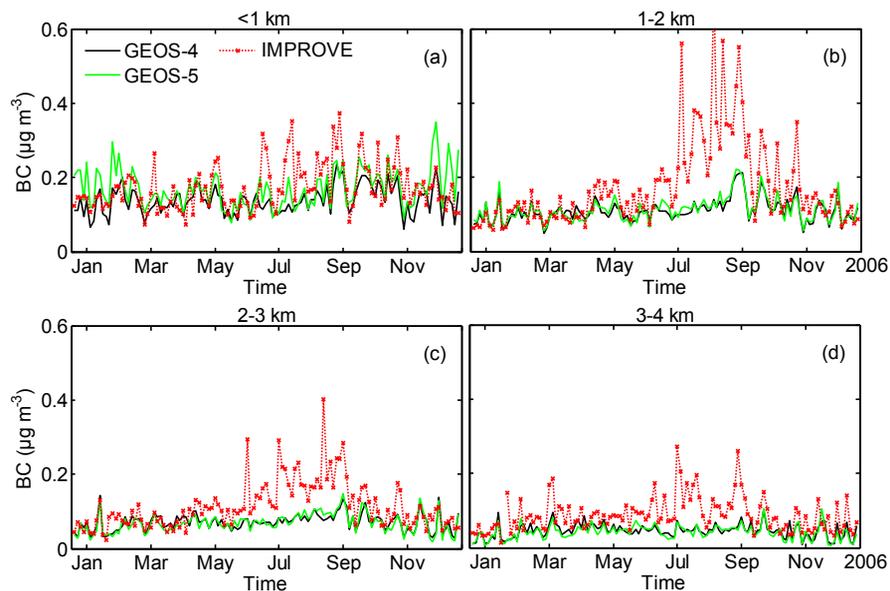


Fig. 9. Same as Fig. 4, but from simulations driven by GEOS-4 (black line) and GEOS-5 (green line) reanalysis data and with 8-day GFEDv2 emissions.

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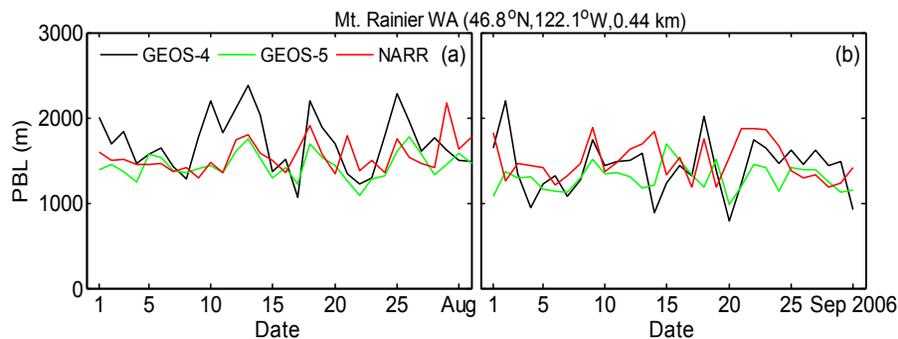


Fig. 10. Same as Fig. 7, but from GEOS-4 (black line), GEOS-5 (green line), and NARR (red line) for 2006.

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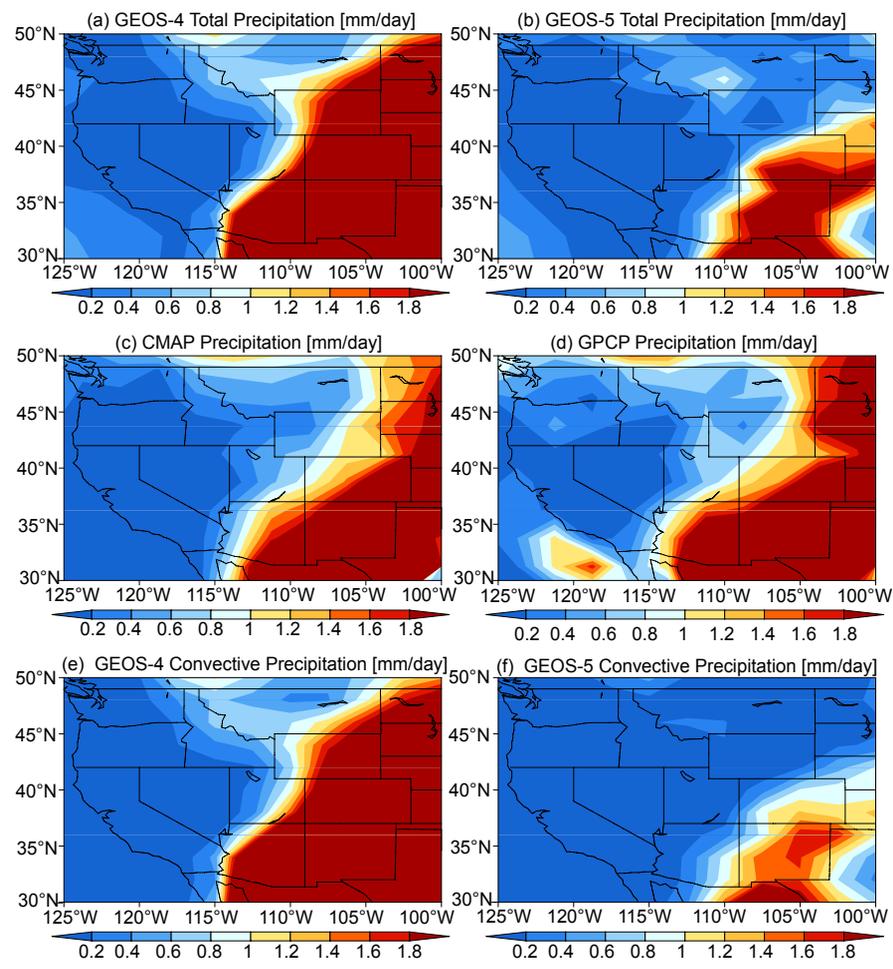


Fig. 11. Same as Fig. 8, but from GEOS-4, GEOS-5, CMAP, and GPCP for August 2006.

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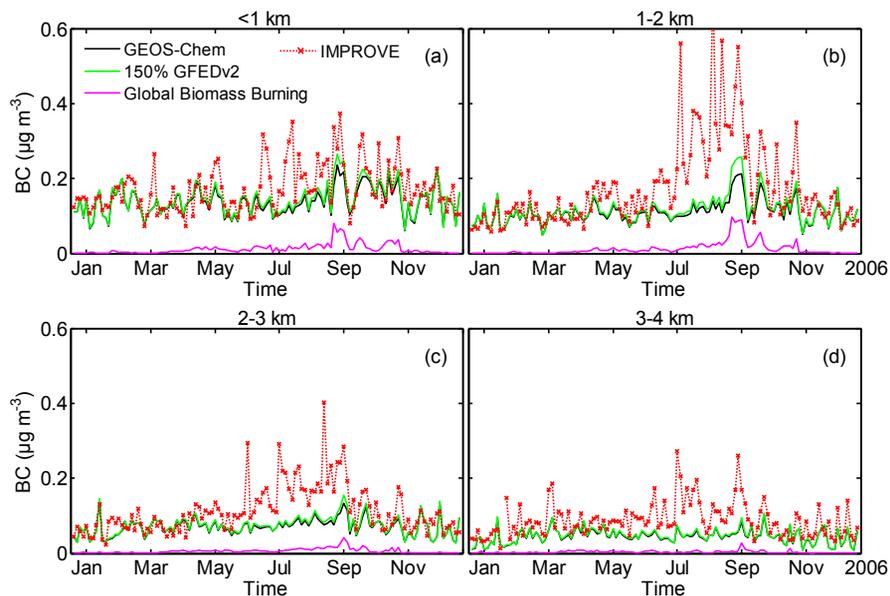


Fig. 12. Same as Fig. 4, but from simulations with standard 8-day GFEDv2 emissions (black line) and with 8-day GFEDv2 emissions increased by 50 % (green line). Also shown are contributions to the surface BC from global biomass burning emissions (pink line).

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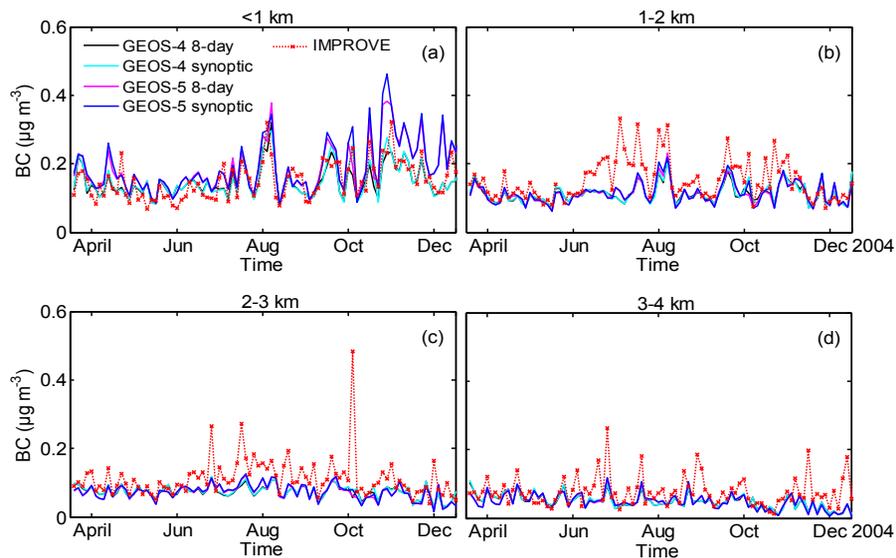


Fig. 13. Same as Fig. 4, but for April–December 2004 and with 8-day GFEDv2 emissions (black line, GEOS-4; pink line, GEOS-5) and synoptic GFEDv2 emissions (green line, GEOS-4; blue line, GEOS-5).

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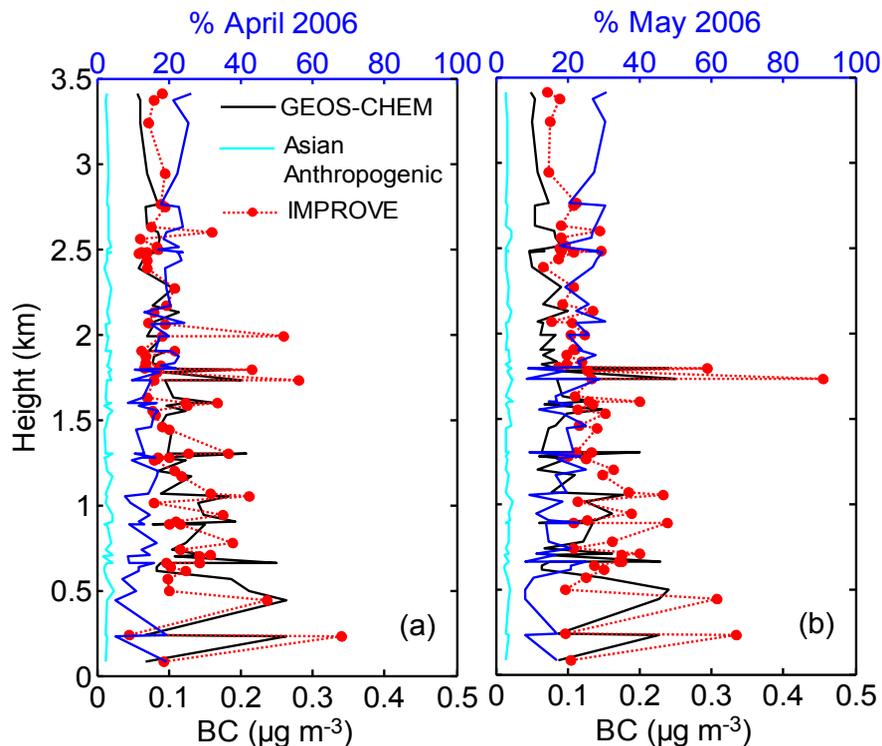


Fig. 14. Monthly mean surface BC concentrations for (a) April and (b) May 2006 at IMPROVE sites (Fig. 1) at different altitudes (red dots, IMPROVE observations; black line, GEOS-Chem simulations driven by GEOS-4 reanalysis data and with 8-day GFEDv2 emissions). Also shown are simulated relative contributions from Asian anthropogenic emissions (blue line).