

Quantifying sources of black carbon in Western North America using observationally based analysis and an emission tagging technique in the Community Atmosphere Model

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Abstract

1
2 The Community Atmosphere Model (CAM5), equipped with a technique to tag black carbon
3 (BC) emissions by source regions and types, has been employed to establish source-receptor
4 relationships for atmospheric BC and its deposition to snow over Western North America. The
5 CAM5 simulation was conducted with meteorological fields constrained by reanalysis for year
6 2013 when measurements of BC in both near-surface air and snow are available for model
7 evaluation. We find that CAM5 has a significant low bias in predicted mixing ratios of BC in
8 snow but only a small low bias in predicted atmospheric concentrations over the Northwest USA
9 and West Canada. Even with a strong low bias in snow mixing ratios, radiative transfer
10 calculations show that the BC-in-snow darkening effect is substantially larger than the BC
11 dimming effect at the surface by atmospheric BC. Local sources contribute more to near-surface
12 atmospheric BC and to deposition than distant sources, while the latter are more important in the
13 middle and upper troposphere where wet removal is relatively weak. Fossil fuel (FF) is the
14 dominant source type for total column BC burden over the two regions. FF is also the dominant
15 local source type for BC column burden, deposition, and near-surface BC, while for all distant
16 source regions combined the contribution of biomass/biofuel (BB) is larger than FF. An
17 observationally based Positive Matrix Factorization (PMF) analysis of the snow-impurity
18 chemistry is conducted to quantitatively evaluate the CAM5 BC source-type attribution. While
19 CAM5 is qualitatively consistent with the PMF analysis with respect to partitioning of BC
20 originating from BB and FF emissions, it significantly underestimates the relative contribution of
21 BB. In addition to a possible low bias in BB emissions used in the simulation, the model is likely
22 missing a significant source of snow darkening from local soil found in the observations.

23 **1 Introduction**

24 Black carbon (BC) is the most light-absorbing component of anthropogenic aerosols, and it has
25 been assessed to be responsible for a significant fraction of the climate warming in the Northern
26 Hemisphere (Bond et al., 2013). BC-containing particles impact the radiative balance of the
27 Earth-atmosphere system in several ways, including their “dimming effect” of reducing the
28 amount of radiation reaching the surface, heating the atmosphere by absorbing radiation, and a
29 darkening effect when incorporated in snow/ice at the surface, thereby increasing absorbed solar
30 radiation (Flanner et al., 2007, 2009). The latter effect is of special interest due to the strong
31 positive feedbacks it can trigger (e.g. Hansen and Nazarenko, 2004; Flanner et al., 2007; Bond et
32 al., 2013). Largely because of this latter effect, BC may play a key role in causing climate
33 change in the snow and ice covered regions of the globe, which have undergone accelerated
34 change in recent decades (Lubin and Vogelmann, 2006; Lewis et al., 2007; IPCC, 2013). There
35 have been numerous studies, both observational and modeling, attempting to highlight and
36 understand the role of BC in accelerating changes in the cryosphere (e.g., Warren and
37 Wiscombe, 1980; Clarke and Noone, 1985; Hansen and Nazarenko, 2004; Jacobson, 2004;
38 Flanner et al., 2007, 2009; Ming et al., 2008; Xu et al., 2009; Koch et al., 2009; Doherty et al.,
39 2010, 2013; Qian et al., 2011, 2015; Huang et al., 2011; Ye et al., 2012; Wang et al., 2015).
40 However, with a few notable exceptions, the focus of these studies has been either in the Polar
41 Regions or sharply circumscribed mid-latitude mountainous regions. Some recent studies (e.g.,
42 Flanner et al., 2009; Shindell and Faluvegi, 2009; Bond et al., 2013) have pointed out that the
43 climatic effect of BC might be greater at mid-latitudes, a relatively understudied region, from the
44 standpoint of global mean forcing.

45 An important aspect of the BC–climate connection is the source attribution of BC in the
46 Earth system. Such attribution is important for the formulation of mitigation strategies, a
47 particularly acute issue for BC since its relatively short lifetime holds promise for mitigation of
48 near-term climate warming. In addition, the global BC forcing estimate is very uncertain mostly
49 because of large uncertainties in BC emissions (e.g., Bond et al., 2013). Observational and
50 modeling source-attribution studies focusing on specific receptor regions are useful for
51 identifying biases in emissions. Previous source attribution studies have primarily focused on
52 sources of BC to the Arctic (e.g., Law and Stohl, 2007; Shindell et al., 2008; Hirdman et al.,
53 2010a, b; Huang et al., 2010; Jacobson, 2010; Hegg et al., 2009, 2010; Stohl, 2006; Sharma et al.,
54 2006, 2013; Sand et al., 2013; Wang et al., 2014), the Antarctic (e.g., Graf et al, 2010), or
55 various mountain regions (Fagerli et al., 2007; Kopacz et al., 2011; Lu et al., 2012; Zhang et al.,
56 2015; Wang et al., 2015). A number of studies have also suggested the importance of long-range
57 transport of aerosols to North America (e.g., Jaffe et al., 1999; VanCuren, 2003; Park et al., 2005;
58 Heald et al., 2006; Chin et al., 2007; Hadley et al., 2007; Eguchi et al., 2009; Clarke and
59 Kapustin, 2010; Fischer et al., 2010; Yu et al., 2012, 2013). A few of these studies assessed
60 transport of BC to North America from various remote source regions using numerical models.
61 For example, Hadley et al. (2007) found that long-range transport from Asia was a major source
62 of BC in the upper atmosphere over North America.

63 Recently, Wang et al. (2014) introduced an explicit aerosol tagging technique to a global
64 aerosol-climate model to produce a detailed characterization of the fate of BC in receptor regions
65 of interest emitted from various geographical source regions. Compared to other widely-used
66 approaches (e.g., the emissions perturbation approach) that have been previously employed to
67 establish global aerosol source-receptor relationships, the tagging approach neither assumes a

68 linear response to perturbations to get fractional contribution of different sources, nor requires
69 additional simulations for each source perturbation. Thus we believe the tagging technique is
70 more computationally efficient and gives more accurate results. Zhang et al. (2015) extended the
71 Wang et al. (2014) modeling tool so it tags source types/sectors in addition to source regions, and
72 they conducted a BC source attribution analysis over the Himalayas and Tibetan Plateau. This
73 modeling framework provides a powerful tool for looking at source attribution of BC in North
74 America, an understudied mid-latitude region for BC in snow.

75 A key facet of employing any model such as that of Zhang et al. (2015) is an assessment
76 of how well it actually reproduces observed values. Atmospheric observational data from the
77 Interagency Monitoring of Protected Visual Environments (IMPROVE) long-term surface
78 monitoring network permit an assessment of model predictions of near-surface atmospheric
79 concentrations of BC. Observations of BC in snow in the Arctic and North China have been used
80 to evaluate models in several previous studies (e.g., Flanner et al., 2007; Skeie et al., 2011; Wang
81 et al., 2011; Lee et al., 2013; Jiao et al., 2014; Qian et al., 2014; Zhao et al., 2014). A recent
82 study by Doherty et al., (2014) presented a large-area survey of observed BC concentrations in
83 snow in Western North America (Fig. S1), affording an opportunity to make such an assessment
84 for model predictions of BC in snow. For the first time, we use their measurements of BC in
85 snow over North America to evaluate our global aerosol-climate model in terms of the amount
86 and sources of BC in snow. The Doherty et al. (2014) study included a Positive Matrix
87 Factorization (PMF) source attribution analysis of BC in snow, making feasible an additional
88 assessment of the source attribution of BC in snow in the enhanced CAM5 model. Here we
89 assess the CAM5 results against these observations and analyses for two receptor areas defined
90 by the western North American region for which the Doherty et al. (2014) data are available.

91 Additionally, we present radiative transfer calculations in the atmosphere and snow with
92 the evaluated model to assess the impact of the modeled BC as well as dust on the radiative
93 balance for the studied region. This facilitates a comparison of the radiative forcing between this
94 region and other mid-latitude or high-latitude regions.

95

96 **2 Methods**

97 **2.1 Observations**

98 Monthly mean near-surface atmospheric BC concentrations for January, February and March of
99 2013 used in this study are from IMROVE non-urban background sites within the United States
100 (Malm et al., 1994). Fine particles (PM_{2.5}, particles with aerodynamic diameters < 2.5 μm) are
101 captured on filters, which are weighed and then subjected to BC concentration analysis using the
102 thermal-optical measurement technique in a laboratory (Chow et al. 1993, 2007).

103 While previous observation/model comparisons of BC in snow have typically compared
104 BC mixing ratios in the surface snow, here we compare the average snow column BC mixing
105 ratio (calculated as the sum of all BC in the snow column divided by the column equivalent
106 water mass, hereafter BCC) over a specified period of time. This is likely a better metric for
107 model comparison than the BC concentration in the top snow layer only, since surface snow
108 mixing ratios at a given point in time can be strongly affected by, e.g., how recently new snow
109 fell, accurate representation of BC mixing ratios in the most recent snowfall and other processes
110 that can vary on the timescale of days. In particular, melting of surface snow can strongly
111 enhance surface snow mixing ratios but melting followed by percolation and refreezing
112 redistributes BC particles within the snow column, resulting in no change to the total BC mass in
113 the snow column. Indeed, Doherty et al. (2014) found that BCC is more regionally consistent

114 than BC concentrations in top snow layer. Further, they showed that while there were vertical
115 variations in the mixing ratio of BC in snow at their study sites there is no consistent vertical
116 gradient. This is also the case in the model (Table S1) consistent with the fact that BC emissions
117 during the cold season don't have strong temporal gradient. Hence, in this study, we use the BCC
118 data from Table 6 of Doherty et al. (2014) to evaluate our model.

119 The BCC estimates by Doherty et al. (2014) are based on samples of seasonal snow
120 collected January through March 2013 at 67 sites in the northwest and north-central U.S. and
121 Canada. Snow BC mixing ratios are estimated based on an optical measurement of spectrally-
122 resolved light absorption by all particles in the snow, using an ISSW (Integrating
123 Sphere/Integrating Sandwich) Spectrophotometer (Grenfell et al., 2011). Absorption is
124 apportioned to BC and non-BC particulate components using the measured absorption Ångström
125 exponent 450-600 nm along with assumed absorption Ångström exponents of the BC and non-
126 BC components. Note that the absorption Ångström exponent is the slope of the logarithm of
127 absorption versus the logarithm of wavelength. Absorption attributed to BC is then converted to
128 a BC mass mixing ratio using a set of calibration standards with weighed amounts of synthetic
129 BC. Full details of the analysis are given by Grenfell et al. (2011) and Doherty et al. (2014). Of
130 relevance here is that this is not a direct measure of BC, but an estimate of mass based on
131 measured absorption and the assumed optical properties of these absorbing components.

132 **2.2 Model description and experimental design**

133 An explicit BC source tagging capability was developed in the Community Atmosphere Model
134 version 5 (CAM5) by Wang et al. (2014), and they applied it to establish source-receptor
135 relationships for BC in the Arctic and quantify source contributions from a few major
136 geographical regions. Zhang et al. (2015) extended this tool to quantifying sources of BC in the

137 Himalayas and Tibetan Plateau originating from biomass & biofuel (BB) and fossil fuel (FF)
138 sectors in various regions. In this study, we use CAM5 with this explicit BC tagging technique,
139 including a recently improved representation of convective transport and wet scavenging of
140 aerosols (H. Wang et al., 2013). We conduct a CAM5 simulation at a horizontal resolution of
141 $1.9^\circ \times 2.5^\circ$ and 56 vertical levels in the specified dynamics mode (Ma et al., 2013), in which
142 model meteorology (e.g., wind, temperature, surface pressure, surface stress, and surface fluxes)
143 are constrained to agree with the NASA Modern Era Retrospective-Analysis for Research and
144 Applications (MERRA) 6 hourly reanalysis (Rienecker et al., 2011), while atmospheric
145 constituents such as water vapor, clouds, and aerosols are allowed to evolve according to their
146 prognostic equations in the model. Although land surface processes including those involve BC
147 in snow are not directly nudged to observations, the constrained meteorological fields should
148 make modeled precipitation and BC deposition more accurate. Monthly-mean model fields for
149 January to March 2013 are used for the comparison to observations in the large-area survey of
150 BC in snow in Western North America (Doherty et al., 2014) and in the comparison to the
151 IMPROVE surface network measurements, and they are used to establish source-receptor
152 relationships and quantify BC radiative forcing.

153 Accurate BC emissions are critical to accurate modeled distributions of BC in the
154 atmosphere and snow, but BC emissions are highly uncertain (e.g., Bond et al., 2013). Instead of
155 using the Intergovernmental Panel on Climate Change (IPCC) AR5 present-day (year 2000) BC
156 inventory (e.g., Lamarque et al., 2010), we compile a new BC emission dataset of year 2010 for
157 our simulation. The 2010 BC emission dataset consists of three parts: 1) The annually-constant
158 total BC emissions over land surfaces, obtained from the ECLIPSE (Evaluating the Climate and
159 Air Quality Impacts of Short-Lived Pollutants) V4a dataset (Stohl et al., 2015), which was

160 developed within the framework of the ECLIPSE European project (<http://eclipse.nilu.no>) using
161 the Greenhouse gas and Air pollution Interactions and Synergies (GAINS) model (Amann et al.,
162 2011), including BC emissions from gas flaring (Stohl et al., 2013); 2) The 2010 annually-
163 constant BC shipping emissions from the IPCC RCP6 (Representative Concentration Pathways);
164 and 3) The 2010 seasonally-varying biomass burning BC emissions from the Global Fire
165 Emission Database (GFED) version 3 (van der Werf et al., 2010). Emission datasets for all other
166 aerosol species are obtained from the IPCC AR5 emission inventories (Lamarque et al., 2010).

167 To prepare BC emissions for the source-type tagging in the CAM5 simulation, we first
168 divide the total ECLIPSE BC emissions over land surface into two types, fossil fuel and biofuel,
169 using the ratio of biofuel to the total (biofuel plus fossil fuel) in each model grid provided by
170 Dentener et al. (2006). In order to make the model source categories directly comparable to those
171 given by the PMF analysis using the observational data, we then combine the GFED biomass
172 burning emissions and ECLIPSE surface biofuel emissions to form the BB emission sector
173 (biofuel and biomass). This is because, as discussed below, the PMF is unable to distinguish
174 open burning (fires) from biofuel burning. The IPCC RCP6 shipping emissions and ECLIPSE
175 surface fossil fuel emissions are also combined to form the FF emission sector (fossil fuel).
176 Figures S2 shows the geographical distributions of JFM (Jan., Feb. and Mar.) mean BB and FF
177 BC emission rate for year 2010 dataset we compiled.

178 Following the division of source/receptor regions in Work Plan (WP 2.1) of the Task
179 Force on Hemispheric Transport of Air Pollution ([http://iek8wikis.iek.fz-
180 juelich.de/HTAPWiki/WP2.1](http://iek8wikis.iek.fz-juelich.de/HTAPWiki/WP2.1)), we define fifteen geographical source regions (Fig. 1a) for this
181 study, including ARC (Arctic), WCA (West Canada and Alaska), ECA (East Canada), LAM
182 (Latin America), NWU (Northwest USA), NEU (Northeast USA), SWU (Southwest USA), SEU

183 (Southeast USA), EAS (East Asia), SAS (South Asia), SEA (Southeast Asia), ERCA (Europe,
 184 Russia and Central Asia), AFME (Africa and Middle East), PAN (Pacific, Australia and New
 185 Zealand), and ROW (Rest of World).

186 Figure 1b summarizes the fractional contributions to global total BC emissions by
 187 different source regions and sectors. The JFM mean global total BC emission rate is 7.69 Tg yr⁻¹
 188 with 53.5% (sum of the red bars) from the BB sector and 46.5% (sum of the blue bars) from the
 189 FF sector. Emissions from source regions in North America (i.e., WCA, ECA, NWU, NEU,
 190 SWU and SEU) are quite low compared to the emissions from the major source regions in Asia,
 191 Europe and Africa.

192 2.3 Metrics

193 Here we define two metrics, following Lee et al. (2013), to quantify the deviation of the
 194 simulated values from the observations.

195 (1) Log-mean normalized bias (LMNB) is defined as

$$196 \quad \text{LMNB} = \frac{\sum_{i=1}^N \log_{10}\left(\frac{C_{mod}^i}{C_{obs}^i}\right)}{N} \quad (1)$$

197 (2) Log-mean normalized error (LMNE) is defined as

$$198 \quad \text{LMNE} = \frac{\sum_{i=1}^N \left| \log_{10}\left(\frac{C_{mod}^i}{C_{obs}^i}\right) \right|}{N} \quad (2)$$

199 N is the total number of data points in a given region for model evaluation. At each point
 200 i , the modeled value (C_{mod}^i) represents the grid mean, while the observed value (C_{obs}^i) is the
 201 average of all point measurements taken within the model grid cell.

202

203 We also define metrics to quantify fractional contribution (C_i^{BB} and C_i^{FF}) and emission
 204 source efficiency (S_i^{BB} and S_i^{FF}), following Zhang et al. (2015), as follows:

$$205 \quad C_i^{BB} = \frac{A_i^{BB}}{\sum_{i=1}^N (A_i^{BB} + A_i^{FF})} \quad C_i^{FF} = \frac{A_i^{FF}}{\sum_{i=1}^N (A_i^{BB} + A_i^{FF})} \quad (3)$$

206 where C_i^{BB} and C_i^{FF} are fractional contributions of BB and FF emissions, respectively,
 207 originating from the source region i to a BC property A_i^{BB} and A_i^{FF} (e.g., mass mixing ratio,
 208 column burden, or deposition flux) in a specified receptor region; and

$$209 \quad S_i^{BB} = \frac{C_i^{BB}}{\left[\frac{E_i^{BB}}{\sum_{i=1}^N (E_i^{BB} + E_i^{FF})} \right]} \quad S_i^{FF} = \frac{C_i^{FF}}{\left[\frac{E_i^{FF}}{\sum_{i=1}^N (E_i^{BB} + E_i^{FF})} \right]} \quad (4)$$

210 where S_i^{BB} and S_i^{FF} are the source efficiencies of BB and FF emissions, respectively, originating
 211 from the source region i , in changing BC in a specified receptor region. E_i^{BB} and E_i^{FF} are the BB
 212 and FF emission rates, respectively, in the source region i . The summation $\sum_{i=1}^N (E_i^{BB} +$
 213 $E_i^{FF})$ represents the global total emission rate from all source regions ($N = 15$ in this study). Thus
 214 the denominator terms are the corresponding contribution of BB or FF emissions in source
 215 region i to the global total BC emissions (Fig. 1b), and the efficiencies S_i^{BB} and S_i^{FF} characterize
 216 the sensitivity of BC properties in a specified receptor region to per-unit BB and FF emissions,
 217 respectively, in source region i .

218 2.4 Data preparation for source attribution

219 In addition to BC concentrations in snow, Doherty et al. (2014) also provide a PMF analysis of
 220 the sources of light absorption by all particulates in the snow. In brief, the PMF analysis
 221 determined the set of orthogonal factors, each with an associated chemical “fingerprint”, that are
 222 associated with variations in light absorption by all particulates in snow. Each of the factors are
 223 then associated with specific source types (e.g. biomass burning, fossil fuel burning, soil, mineral

224 dust, etc.) based on their chemical fingerprints. The chemical markers from open biomass
225 burning (e.g., forest fires) and biofuel burning (e.g., woodsmoke from fireplaces and wood
226 stoves) are quite similar, so biomass and biofuel sources cannot be distinguished in the PMF;
227 both sources would be included in the factor identified as “biomass burning (BB)”. In order to do
228 a comparison to CAM5, which tracks the sources of BC only, rather than all light-absorbing
229 species to snow, we re-ran the PMF analysis so it determined the sources that contribute to
230 variations in snow BC only (i.e. C_{BC}^{est} , in Doherty et al., 2014). This PMF analysis of sources of
231 BC in snow (Fig. S3) shows a similar, though not identical, source attribution as that for all light-
232 absorbing particulates in snow (given in Doherty et al., 2014). For both, the main source sectors
233 are pollution (likely mainly fossil fuel combustion), soil, and biomass/biofuel burning. These
234 three categories account for almost all of the light absorption by BC and other particles in the
235 snow samples. The fractional contribution of the fossil fuel/pollution source is higher for BC
236 (Fig. S3) than for total particulate absorption (Doherty et al., 2014), and the fractional
237 contribution by the soil factor is lower for BC than for total particulate absorption. The issue of
238 the nature of a BC component associated with soil, which is not intuitively obvious, is discussed
239 below.

240 The estimated snow BC concentration used in the PMF analysis and the fraction of
241 absorption due to the biomass burning, pollution/fossil-fuel and soil sources (F_{BB} , F_{FF} and F_{SOIL})
242 from the PMF analysis, are given in the Table S2. The PMF analysis allows some factors to
243 contribute negative fractions to absorption, which is of course unphysical. To rationalize the data
244 for comparison with CAM5, we first set all negative fractions F_{BB} , F_{FF} and F_{SOIL} to zero, and
245 then scale the remaining fractions so that they sum to 1.0, yielding adjusted values f_{BB} , f_{FF} and
246 f_{soil} .

247 We next calculate average fractional contributions by the BB and FF sources from the
248 PMF analysis for each of the snow samples sites falling within a given model grid box, using Eq.
249 (A1) in the Appendix. It is important to note that the sum of BB and FF contributions does not
250 necessarily equal to 100%. This is, of course, because of the soil source in the PMF model, a
251 source of BC not present in CAM5. This renders the comparison between the model (i.e.,
252 CAM5) and observed (i.e., PMF) sources of BC imperfect, an issue that will be discussed further
253 below. The CAM5 JFM mean fractional contributions for the BB and FF sectors in each model
254 grid box, where observational/PMF data are available, are calculated using Eq. (A2). Note that
255 the sum of BB and FF contributions equals to 100%.

256 Based on the above procedures, we calculate the regional average of fractional
257 contributions from the BB and FF sectors from the PMF analysis and from the CAM5 simulation
258 using Eqs. (A3) and (A4), respectively. In principle, another fraction corresponding to the soil
259 contribution should also be present in Eq. (A3) for the PMF analysis. By excluding this fraction,
260 we are essentially renormalizing our fractional contributions such that \overline{BB}_{obs} and \overline{FF}_{obs} now
261 represent the fractions of direct combustion emissions (fossil fuel and biomass/biofuel) that can
262 be attributed to the BB and FF sectors. This renders these fractions equivalent to those generated
263 by CAM5 via Eq. (A4).

264

265 **3 Results and discussion**

266 **3.1 Near-surface atmospheric BC concentrations**

267 There are 42 non-urban IMPROVE observation sites available in the northwest of the USA
268 (Figure S4). For comparison with model results, measurements at sites located in the same model
269 grid box are averaged first. As a result, we obtain 30 model/observation comparison pairs. The

270 following analysis is based on the JFM mean modeled and observed values for these 30
271 comparison pairs.

272 Figure 2a shows the scatter plot of simulated versus observed JFM mean near-surface BC
273 concentrations. About 57% of the ratios fall within a factor of 2. The linear correlation
274 coefficient (R) is 0.5. The statistical significance of R is at >99% confidence level ($p = 0.005$, N
275 $= 30$). The LMNB and LMNE are calculated using Eqs. (1) and (2), respectively. The CAM5
276 results over the 30 grid boxes have LMNB of -0.05, which means that the model-predicted BC
277 concentrations are smaller than observations by 11% ($=1-10^{-0.05}$) on average. The model error
278 relative to the observations is, however, more substantial. The LMNE is 0.3, which means that
279 the model predictions are, on average, within a factor of 2 ($=10^{0.3}$) of the observations. Figure 2b
280 shows statistics for the JFM near-surface BC concentrations for the IMPROVE observations and
281 CAM5 results, respectively. The model moderately under-predicts mean and median BC
282 concentrations, as expected. The maximum observed and modeled near-surface BC
283 concentrations among the sites are close, but the modeled minimum and 25th percentile values
284 are higher than observed values. The observed and modeled mean values (\pm standard deviation)
285 are $72.0 \pm 63.3 \text{ ng m}^{-3}$ and $54.8 \pm 42.5 \text{ ng m}^{-3}$, respectively. The strong spatial variation in BC over
286 these sites, indicated by the high coefficient of variation (i.e., the ratio of the standard deviation
287 to the mean – see also the spatial distributions of BC in Fig. S4), renders the comparison of these
288 mid-latitude observations with CAM5 (having a horizontal grid spacing of $1.9^\circ \times 2.5^\circ$)
289 challenging. In this light, we consider the model-observational agreement within a factor of two
290 quite reasonable.

291 **3.2 BC in snow column**

292 In addition to evaluation of BC in the atmosphere, we also evaluate the model
293 performance with respect to BC in snow. Figure 3 shows a comparison between CAM5
294 predictions of BCC and the corresponding observations of BCC from the 49 sampling sites given
295 in Table 6 of Doherty et al. (2014), where total-column snow BC could be calculated. We obtain
296 36 observation/model comparison pairs by averaging measurements made at all sites located in
297 the same model grid box. This results in 20 comparison pairs in the Northwest USA and 16 in
298 West Canada (Fig. 3d; BCC concentrations for individual pairs are summarized in Table S3).
299 Modeled BCC does not differ appreciably between January, February and March for the grid
300 boxes where we made comparisons, so we use the mean BCC across all three months (JFM) in
301 the comparison with the observation.

302 Figure 3a shows the scatter plot of the simulated JFM mean values compared to observed
303 BCC over the 36 observation/model pairs. BCC is substantially lower in the modeled snowpack
304 than in the observations. This model low bias in BCC is substantially larger than in near-surface
305 atmospheric concentrations of BC (hereafter, referred to as BCS) discussed in the previous
306 section. Indeed, the contrast in the model-observational bias of BCC as compared to the bias for
307 BCS is quite interesting and suggestive of the sources of the bias in the BCC model-
308 observational comparison. However, it is important to note here that we are not comparing BCS
309 values with BCC values but rather comparing the model-observational biases of the two
310 variables.

311 The linear correlation coefficient (R) for the modeled versus observed BC mixing ratios
312 in snow is 0.2, significant only at the 70% level ($p = 0.3$, $N = 36$). The CAM5 BCC has a LMNB
313 (Eq. 1) of -0.2 which means that the model-predicted BCC concentrations are lower than the
314 observations by 37% ($=1-10^{-0.2}$) on average. The LMNE (Eq. 2) in the CAM5 BCC is 0.3 which

315 means that the model predictions are, on average, within a factor of 2 ($=10^{0.3}$) of the observations,
316 though as noted above the correlation between the two is poor. The observed and modeled means
317 (\pm standard deviation) for these 36 BCC values are 32.7 ± 24.5 ng g⁻¹ and 19.1 ± 11.5 ng g⁻¹,
318 respectively. As was the case with model comparisons for BCS, BCC has a large coefficient of
319 variation (i.e., the ratio of the standard deviation to the mean), reflecting the strong spatial
320 variation of BCC in this region (Fig. 3d).

321 Figure 3b compares the simulated and observed BCC as a function of latitude. The
322 modeled JFM zonal mean of BCC over the longitude range of 93.75–123.75° W (blue line in Fig.
323 3b) shows an increasing trend with latitude in the Northwest USA and a decreasing trend in West
324 Canada. This trend is also seen in the observations in West Canada, but there is no trend in BCC
325 with latitude in the Northwest USA. The model agrees well with the observations in Canada, but
326 has generally lower concentrations of BC in snow in the U.S. (Fig 3c). The observed values of
327 BCC range between 8 and 110 ng g⁻¹ in the Northwest USA with a mean of 44 ng g⁻¹, and 7 to 39
328 ng g⁻¹ in West Canada with a mean of 19 ng g⁻¹. The correlation coefficient between the observed
329 and modeled BCC is low ($R=0.1$) for the Northwest USA with negligible statistical significance
330 ($p = 0.6$, $N = 20$). However, the correlation coefficient (R) is relatively high (0.7) for West
331 Canada, significant at >99% confidence level ($p = 0.005$, $N = 16$). The good agreement between
332 the model estimates and observations of BCC for Western Canada argues against a systematic
333 problem with model deposition processes.

334 Turning next to the regionally stratified LMNB and LMNE values, for the Northwest
335 USA region, the LMNB and LMNE are -0.39 (59% low bias) and 0.47 (a factor of 3),
336 respectively, while for West Canada, LMNB and LMNE are -0.04 (9% low bias) and 0.17 (a
337 factor of 1.5), respectively. Hence, for West Canada the model bias is essentially the same for the

338 BCC as it is for the BCS (in Northwest USA) while the model error is actually appreciably less.
339 For the Northwest USA, on the other hand, the LMNE is substantially worse for BCC than it was
340 for BCS. Furthermore, most of this error is associated with a model low bias far larger than that was
341 the case for BCS. Note that the measurements of BCS and BCC are from different locations and
342 are not necessarily representative of the whole model grid box, so the comparison of biases in
343 BCS and BCC is not ideal but is nonetheless informative. Although BCC and BCS at a given
344 location depend on different processes that occur on different timescales, averaging over the
345 three-month time period and/or across snow columns at the available sites should be sufficient to
346 make the biases in BCS and BCC comparable.

347 The smaller error (LMNE) in BCC for West Canada than for BCS in the Northwest USA
348 indicates the model might also be doing a better job of predicting BCS in West Canada than in
349 the Northwest USA, but it is not possible to know this since all the BCS observations we have
350 are from sites in the USA. For the Northwest USA sites the substantially larger low bias in BCC
351 versus in BCS is quite interesting. A commonly invoked explanation for a low bias in model
352 predictions of atmospheric BC has been flawed emissions inventories. For example, Mao et al.
353 (2011) indicated that there is a large uncertainty in the emissions of BC from biomass burning in
354 western North America. However, the larger low bias in BCC compared to BCS suggests that
355 deficiencies in emissions inventories are not likely the primary explanation for the model under-
356 prediction of BCC in this instance, since a source-based bias should show up in both BCS and
357 BCC (similar source attribution of BCS and BC deposition shown in Fig. 4), assuming the model
358 representation of deposition/scavenging processes is not flawed. In fact, the small bias in model-
359 predicted BCC in West Canada indicates that the model representation of BC deposition is less
360 likely to be the primary cause of the large low bias in BCC in Northwest USA.

361 In addition to emissions or model processes errors, another possibility for the difference
362 in modeled and observed BCC is a bias¹ in the observational estimates. In a recent comparison,
363 Schwarz et al. (2012) found that estimates of the mixing ratio of BC in snow using the ISSW
364 (used in the Doherty et al., 2014 study to estimate BCC) were biased high by up to a factor of
365 three when BC is mixed with dust. While this artifact could possibly explain a portion of the
366 observed discrepancy between the model predictions and the observations, it is not fully
367 consistent with the contrast in model-observational comparisons between the Northwest USA
368 and West Canada regions. Although there is significantly less dust in the Canadian samples
369 (based on both ISSW analysis of BC/non-BC partitioning of absorption and the PMF analysis)
370 than for the Northwest USA, the amount of dust present at the West Canada sites is still
371 substantial: The PMF analysis suggests that ~17% of the light absorption is associated with dust
372 for the Canadian sites on average, and much more at some sites, whereas it's ~36% at the U.S.
373 sites. Given this, we would expect to also find a model low bias in BCC for Canada on the order
374 of half that in the Northwest U.S., e.g. LMNB of about -0.2, rather than the actual near-zero bias
375 (LMNB=-0.04). Hence, the relatively good model-observational agreement for the Canadian
376 sites makes it unlikely that measurement bias in BCC is the sole source of the discrepancy
377 between the CAM5 predications and the field observations.

378 Another possible cause of lower BCC in the model versus the observations is a missing
379 source of BC to snow in the model. The sources of BC in CAM5 are biofuel burning, biomass
380 burning and fossil fuel combustion. In the model, emissions of BC from these sources are
381 incorporated in surface snow either in snowfall (wet deposition) or by settling directly to the
382 surface snow (dry deposition). In contrast to this, the PMF analysis suggests that a significant

¹ For simplicity and consistency we use “model bias” below to describe the difference between model results and observations, although the measurements might have a significant bias or error.

383 source of BC in snow is soil. At first glance this seems counter-intuitive, since soil itself does not
384 produce BC. However, in mid-latitude regions the snow is often patchy, and intermixed with
385 large areas of exposed soil. This soil can mix with the snow mechanically (e.g. by livestock; X.
386 Wang et al., 2013) or by winds, which loft the soil and deposit it to snow on scales of tens to
387 hundreds of meters (Doherty et al., 2014). These exposed soil areas are subject to BC deposition
388 throughout the year and likely accumulate a substantial reservoir of BC from a multitude of
389 sources (e.g., Schmidt and Noack, 2000; Hegarty et al., 2011). This deposited BC is then subject
390 to re-suspension via saltation and deposition on the surrounding snow, along with the soil. As
391 mentioned above, the contribution of the soil/dust source to light absorption by snow impurities
392 for the Canadian sites is $17 \pm 5\%$. In contrast, for the U.S. sites it is $36 \pm 4\%$, consistent with the
393 thinner and more variable snow cover in the U.S. region (snow cover fraction derived from
394 satellite measurements shown in Fig. S5). While the magnitude of this source of BC to snow is
395 unknown, the PMF analysis suggests this mechanism for getting BC into snow is not
396 insignificant in some locations. Thus, soil as a source of BC to snow at the USA sites likely
397 explains a substantial portion of the low bias in modeled snow BC for sites in this region with
398 patchy snow cover, and is also likely the explanation for much of the low bias over the entire
399 data set. We turn next to an assessment of the source attribution of BC in CAM5, including a
400 comparison with the results of a PMF analysis of the North American observations of BC in
401 snow.

402

403 **3.3 Source attribution and emission source efficiency**

404 **3.3.1 Modeled source-receptor relationships using CAM5**

405 The direct source tagging method in CAM5 provides a straightforward means of quantifying
406 source-receptor relationships for BC reaching the receptor regions in North America originating
407 from the various source regions and types. Figures 4a and 4b show relative contributions (as
408 defined in Sect. 2.3, Eq. 3) to the JFM mean BC atmospheric column burden, deposition flux,
409 and near-surface atmospheric concentrations for two receptor regions, the Northwest USA and
410 West Canada (as outlined by white boxes in Fig. 3d). The contributions are shown explicitly for
411 all major source regions and both source types (solid bar for BB and stippled bar for FF). The
412 contributions of BB and FF from minor source regions are lumped together (black bar in Figs. 4a
413 and b). Clearly, FF sources play a primary role in determining atmospheric concentrations and
414 deposition fluxes of BC. Contributions of BB and FF from the North American sources
415 (hereafter, for brevity, we use USA to denote four source regions NWU, NEU, SWU and SEU;
416 see Figure 1a for region definitions) increase in importance moving from total column
417 atmospheric burden to deposition fluxes and then to near-surface atmospheric concentrations of
418 BC. North American sources, especially FF sources, are definitely the major sources of BC in the
419 near-surface atmosphere and of BC deposited to the surface – i.e. to snow – as they are within or
420 close to the receptor regions. Long-range transport of BC from distant sources in Asia and Africa
421 (e.g., EAS, SAS, SEA and AFME) to North America takes place mainly in the middle and upper
422 troposphere (shown in Fig. S8); BC in this part of the atmosphere is less prone to wet removal,
423 and thus contributes more to column burden than to near-surface BC or deposition. The spatial
424 distributions of JFM mean BC column burden and deposition along with BC transport pathways
425 from various distant and domestic source regions and sectors to North America are shown in Fig.
426 S6–S11.

427 Contributions to BC atmospheric column burden from all source regions are 38% BB and
428 62% FF for the Northwest USA receptor region, and 37% BB and 63% FF for the West Canada
429 receptor region. Contributions to BC column burden from the overseas combination of EAS
430 (East Asia), SAS (South Asia), SEA (Southeast Asia) and AFME (Africa and Middle East) to the
431 Northwest USA and West Canada receptor regions are 57% (32% BB and 25% FF) and 63% (32%
432 BB and 31% FF), respectively, among which BB from SAS and FF from EAS are the two main
433 overseas sources. Contributions to BC column burden in the receptor regions from the North
434 American source regions (USA and WCA) are 41% (5% BB and 36% FF) for the Northwest
435 USA and 34% (5% BB and 29% FF) for West Canada.

436 Relative to that for total column burden, the contribution from FF increases for deposition
437 and is even greater for near-surface atmospheric BC. Contributions from the combined source
438 regions of USA and WCA to BC deposition over two receptor regions, Northwest USA and West
439 Canada, are 77% (10% BB and 67% FF) and 81% (11% BB and 70% FF), respectively. For
440 near-surface atmospheric BC, the total FF contributions from the USA and WCA (West Canada
441 and Alaska) increase to 82% (76% from USA) and 83% (75% from WCA) over Northwest USA
442 and West Canada, respectively.

443 Figures 4c and 4d show emission source efficiency (as defined in Sect. 2.3, Eq. 4) in
444 affecting the three JFM mean BC properties in both receptor regions. We use this efficiency
445 (assuming a global mean efficiency of 1) as an index to quantify the sensitivity of BC in a
446 receptor region to a fixed mass perturbation in emissions in different source regions and sectors.
447 It is not surprising that BC in a given receptor region is most sensitive to local emissions (i.e.,
448 NWU for the Northwest USA receptor and WCA for the West Canada receptor). As was the case
449 for source attributions in Figure 4a and 4b, the emission source efficiency (Fig. 4c & 4d) of more

450 local sources is lowest for total atmospheric column burden, then increases for deposition and
451 near-surface atmospheric BC. The distant emission sources have quite low efficiencies, with
452 significant non-local contributions only for the total column burden.

453 Differences in the vertical distribution of contributions to atmospheric BC are shown in
454 more detail in Fig. 5a and 5b. Modeled vertical profiles of area-averaged BC mixing ratio and
455 liquid cloud fraction over both receptor regions are also shown, in Fig. 5c and 5d, to indicate the
456 altitude where wet scavenging of aerosols in clouds is most likely to occur. Clearly, the
457 contribution of local sources significantly decreases above 800 hPa, while distant sources
458 become progressively more important at higher altitudes (Fig. 5a & 5b). BC from distant sources
459 contribute less to wet scavenging of BC mass than they do to column burden in the two receptor
460 regions. Liquid clouds are at a maximum in the 600–800 hPa layer. Here, the BC profiles also
461 show a minimum, possibly associated with cloud scavenging of BC in the model. This layer
462 (600–800 hPa) has an intermediate local source contribution between those in the higher layers
463 and the bottom layer (800–1000 hPa). Above 400 hPa, liquid clouds and thus wet removal are
464 minimal. Below 800 hPa, below-cloud scavenging by precipitation removes BC from the air and
465 in this altitude range BC sources are mostly local. This would increase the local source
466 contribution to the total deposition flux.

467 **3.3.2 Comparison of source sector attribution between CAM5 and PMF**

468 Using the procedures described in Sect. 2.4, our PMF source attribution results are compared
469 with the corresponding CAM5 source attributions (Table 1). Comparisons are done for each
470 model grid box where we have a model/observation comparison pair. We reiterate that for both
471 data sets BB includes emissions from both open biomass burning and biofuel burning.

472 As discussed in Sect. 2.4, the BB and FF fractions for the PMF analysis are not precisely
473 comparable to those from CAM5 since the PMF analysis has identified an additional BC source,
474 soil, which is not included in the CAM5 simulation. This is reflected in the fact that, while the
475 sum of CAM5 BB and FF contributions equals 1, the sum of BB and FF contributions from the
476 PMF analysis are commonly less than 1. Due to the lack of soil source in CAM5 and
477 uncertainties in both measurements and emissions (e.g., spatial distribution of sources and the
478 partitioning between BB and FF sectors), it is not surprising that there are quite large
479 discrepancies between the CAM5 and PMF values for some individual comparison pairs. When
480 compared to the PMF values (which included contributions from FF, BB and soil), CAM5
481 underestimates the BB contribution for 80% of the comparison pairs (modeled mean and
482 standard deviation of $18\% \pm 5\%$ vs. PMF values of $28\% \pm 22\%$) and overestimates the FF
483 contribution for all comparison pairs ($82\% \pm 5\%$ vs. $47\% \pm 21\%$).

484 For a better quantitative PMF/CAM5 comparison, relative contributions to BC were also
485 calculated for a PMF analysis allowing for BC only from direct combustion sources, i.e., the BB
486 and FF sources of BC considered in the CAM5 simulation. Average contributions of BC from
487 combustion sources only are compared for our two receptor regions in Figure 6. The two regions
488 differ little in the partitioning of the BC between BB and FF sources, but in both regions the
489 PMF indicates a larger role by BB than does the model. The PMF model attributes 32% of the
490 BC to BB for the Northwest USA region, while for West Canada the fraction is 28%. CAM5
491 attributes 16% of BC in the Northwest USA to BB and 15% to BB in West Canada. Averaging
492 over both regions, the PMF model attributes 30% of the BC to BB while CAM5 allocates 16% to
493 this source. Compared to the PMF results, CAM5 over-predicts the ratio of FF to BB for the
494 North American receptor region.

495 While certainly significant, the difference in source attribution between CAM5 and the
496 factor analysis is not surprising. The factors that possibly cause the substantial model low bias in
497 BCC could potentially generate biases in the source-type attribution. In addition, uncertainties in
498 BC emission data and model treatment of BC aging/deposition processes can also be a source of
499 bias in the attribution, including but not limited to 1) the partitioning of BC emissions into fossil
500 fuel and biofuel based on the ratio provided by Dentener et al. (2006); 2) initial injection heights
501 (up to 6 km) of biomass burning emissions that directly affect BC interaction with clouds and its
502 wet deposition in CAM5; 3) treatment of the mixing of hydrophobic BC particles with
503 hygroscopic components (e.g., sulfate and organics) that is important for BC aging and wet
504 removal but does not differentiate BB or FF origin in the model. These factors, among many
505 others, along with the possible measurement bias for samples with large soil dust concentrations,
506 could explain the difference in source-type attribution between CAM5 and the PMF analysis.
507 The data we have are not sufficient to distinguish between these possible sources of bias.

508 **3.4 Radiative forcing**

509 Figure 7 shows the CAM5 modeled JFM mean atmospheric BC all-sky shortwave direct
510 radiative forcing (DRF) at the surface (dimming effect), at the top of the atmosphere (TOA) and
511 in the atmosphere (heating effect), and it also shows the radiative forcing due to BC and mineral
512 dust in snow (darkening effect), as a function of latitude (zonally averaged over the longitude
513 band 93.75–123.75° W). The forcing due to BC is separated out from other aerosol components
514 using the radiation diagnostic calculations recently implemented in CAM5 by Ghan et al. (2012),
515 while the BC- and dust-in-snow forcing are calculated in the SNICAR (SNow, ICe, and Aerosol
516 Radiative) model (Flanner et al., 2007), which is coupled to CAM5. The CAM5/SNICAR
517 models do include the light-absorbing effect of mineral dust particles (in addition to BC). Note

518 that the surface radiative forcing due to BC and dust in snow shown here is the total-area mean
519 forcing (i.e., zero values enter the calculation for snow-free grids during the model integration),
520 so this represents the true climate forcing (Flanner et al., 2007).

521 The DRF by BC in the atmosphere (in-atmosphere heating) decreases with latitude, as
522 does DRF at the surface (cooling). The DRF of BC at the TOA maximizes around 50° N, where
523 BC- and dust-in-snow radiative forcings also reach their maxima. To explain these variations
524 with latitude, we plot the zonal mean of JFM mean BC total column burden in Figure 7, and we
525 also plot BC and dust deposition scaled by the snow cover fraction (SCF) to weigh the
526 contribution by each grid box to the area mean forcing by BC and dust in snow. The model
527 estimate of surface SCF was first assessed and found to be in reasonable agreement with the
528 satellite retrievals (shown in Fig. S5). Clearly, the total column burden shows the same trend as
529 the DRF in the atmosphere, and the BC- and dust-in-snow radiative forcing follow the respective
530 latitudinal variations of deposition flux. This suggests that the source attribution for BC DRF in
531 the atmosphere and forcing by BC in snow could be by approximated using the source-receptor
532 relationships for BC total column burden (Fig. 4) and BC deposition (Table S4), respectively, if
533 one assumes a linear relationship between radiative forcing and BC concentrations. Note that we
534 did not use such an assumption in the radiative forcing calculation.

535 The color-coded numbers in Fig. 7 correspond to the various JFM mean radiative
536 forcings averaged over the entire receptor regions, Northwest USA and West Canada. The BC
537 darkening effect on snow is significant and comparable to its DRF in the atmosphere, especially
538 in West Canada where snow covers almost the entire area (Fig. S5). It's interesting to note that
539 the BC darkening effect outweighs the BC dimming effect (i.e., cooling at the surface) and
540 warming effect on the Earth-atmosphere system (i.e., DRF at the TOA) over both of the two

541 regions. The modeled surface radiative forcing due to dust in snow is very small in these regions.
542 However, Doherty et al. (2014) found that local soil dust, which is not considered in the CAM5
543 simulation, is a significant contributor to light absorption in snow over the U.S. Northern Plains,
544 as well as at some sites in Canada. Intra-regionally transported desert dust has also been shown
545 to have a significant impact on snow in the San Juan Mountains of Colorado (e.g., Painter et al.,
546 2010, 2012) and in northwest China (X. Wang et al., 2013; Zhang et al., 2013). This suggests
547 that CAM5 and other climate models that ignore the surface radiative forcing induced by soil
548 and/or desert dust in snow may significantly underestimate the impact of light-absorbing
549 impurities on snowmelt and climate.

550

551 **4 Summary and conclusions**

552 In this study, the CAM5 global model, implemented with an explicit BC source tagging
553 technique, has been employed to establish source-receptor relationships for atmospheric BC and
554 its deposition to snow over a large receptor area encompassing a substantial portion of the Great
555 Plains of North America. The model meteorological fields are constrained to agree with the
556 MERRA reanalysis data sets for year 2013. Model-predicted near-surface atmospheric BC
557 concentrations and BC-in-snow concentrations in January, February and March (JFM) were
558 evaluated against atmospheric observations from the IMPROVE network and field
559 measurements from a recent large-area survey of BC (and other light-absorbing particles) in
560 snow over land (Doherty et al., 2014), respectively. We found that CAM5 had a small low bias
561 (11%) but a substantial random error (about a factor of 2) in the estimates of monthly mean near-
562 surface atmospheric BC concentrations. However, the model had a substantial error (a factor of 2)
563 and a larger negative bias (37%) in the prediction of BC-in-snow concentrations at all the snow

564 sampling sites. A common explanation for a low bias in model predictions of atmospheric BC
565 has been an underestimate of BC emissions and/or an overestimate of removal during the
566 transport. However, systematic biases in emissions and/or model processes should show up
567 consistently in both atmospheric BC and BC in snow, and/or in adjacent geographic regions of
568 sampling sites. Analysis of the geographic variation in the bias and error in modeled BC in snow
569 versus that observed, along with the model-observational comparison of the atmospheric near-
570 surface BC at the U.S. sites, suggests that the negative model bias is more likely due to the lack
571 of a soil source for BC in patchy snow rather than an underestimate of direct combustion
572 emissions in the model simulation. Patchy snow at the U.S. sites is prone to contamination of soil
573 dust originating from the exposed soil areas. The soil dust may contain BC deposited from the
574 atmosphere, which was not included in the emission inventory for the CAM5 simulation.
575 Although our analysis supports this plausible explanation for the larger BC-in-snow model bias
576 at the U.S. sites, an underestimate of regional BC emissions still cannot be excluded as a cause of
577 the model-observational difference. It is also possible that some of the difference between model
578 and observation is due to a high bias in the measurements when BC is mixed with significant
579 amounts of light-absorbing soil dust. However, the relatively good model-observational
580 agreement for the Canadian sites makes it unlikely that BC-in-snow measurement bias is the sole
581 source of the discrepancy between the CAM5 predications and the field observations.

582 The explicit direct source tagging technique in CAM5 permits a quantitative attribution
583 of BC in receptor regions (Northwest USA and West Canada) to source regions (North American
584 or more distant emissions) and source types (fossil fuel, FF, versus biomass/biofuel, BB). In the
585 model, local sources generally contribute more to near-surface BC and deposition than distant
586 sources. However, distant sources contribute significantly to the column BC burden, especially

587 to BC in the middle and upper troposphere. At these altitudes wet removal is relatively weak, so
588 little of this BC likely reaches the surface snowpack. In the model, FF is the dominant source
589 type for total column BC over the two receptor regions. FF is also the dominant local source type
590 for BC column burden, deposition, and near-surface BC. However, for all distant source regions
591 combined the contribution of BB is larger than FF.

592 An observationally-based PMF analysis of the sources of BC to snow, based on snow
593 chemistry, is compared to the CAM5 source attribution based on source tagging. While the
594 CAM5 source attribution was biased high for the FF sector and low for the BB sector compared
595 to PMF, they both show that the contribution of the FF sector is much larger than that of the BB
596 sector. For the two receptor regions examined in this study (Northwest US and Northwest
597 Canada), the relative contribution of the BB sector was underestimated by about a factor of two
598 in CAM5 relative to that given by the PMF analysis. The quantitative difference in the source-
599 type attribution between CAM5 and PMF analysis could be due to an underestimation of North
600 American BB emissions, the lack of a soil source of BC with a high BB/FF ratio in the model,
601 model treatment of aerosol aging/deposition processes such that the wet removal rate of BC from
602 the BB sector is overestimated, and/or biases in the measurements.

603 Based on the CAM5 predictions of BC concentrations in both the air and snow, and of
604 dust in snow, radiative forcing calculations were carried out for our two North American
605 receptor regions (Figure 3d). The darkening effect of BC in surface snow (i.e., snow albedo
606 reduction due to the presence of BC) is substantially larger than the BC dimming effect (i.e.,
607 reduction in surface radiative flux due to BC in the atmosphere) but is comparable to BC heating
608 in the atmosphere. The modeled surface radiative forcing due to dust in snow is small in the two
609 regions. However, Doherty et al. (2014) found that local soil, which is not considered in the

610 CAM5 simulation, is a significant contributor to light absorption in snow, suggesting that CAM5
 611 and other climate models that ignore the local soil contributions to snow may significantly
 612 underestimate the impact of light-absorbing impurities on snowmelt and climate.

613 **Appendix:**

614 The average fractional contributions by the BB and FF sources from the PMF analysis for
 615 each of the snow samples sites (k) falling within a given model grid box are calculated using Eq.
 616 (A1).

$$617 \quad BB_{obs}^i = \frac{\sum_{k=1}^S C_{obs}^k \times f_{BB}^k}{\sum_{k=1}^S C_{obs}^k \times (f_{BB}^k + f_{FF}^k + f_{soil}^k)} \quad FF_{obs}^i = \frac{\sum_{k=1}^S C_{obs}^k \times f_{FF}^k}{\sum_{k=1}^S C_{obs}^k \times (f_{BB}^k + f_{FF}^k + f_{soil}^k)} \quad (A1)$$

618 where $f_{BB}^k + f_{FF}^k + f_{soil}^k = 1$. C_{obs}^k is the estimated snow BC concentrations used in the PMF
 619 analysis for the snow sampling site k (Table S2). S is the total number of sampling sites within
 620 the same model grid box.

621 The CAM5 JFM mean fractional contributions for the BB and FF sectors in each model
 622 grid box, where observational/PMF data are available, are calculated using Eq. (A2).

$$623 \quad BB_{mod}^i = \frac{\sum_{j=1}^M C_{mod}^j \times D_{BB}^j}{\sum_{j=1}^M C_{mod}^j \times (D_{BB}^j + D_{FF}^j)} \quad FF_{mod}^i = \frac{\sum_{j=1}^M C_{mod}^j \times D_{FF}^j}{\sum_{j=1}^M C_{mod}^j \times (D_{BB}^j + D_{FF}^j)} \quad (A2)$$

624 where C_{mod}^j are the modeled snow BC concentrations in month j for the model grid box i .
 625 D_{BB}^j and D_{FF}^j are fractional contributions of BB and FF deposition, respectively, to total BC
 626 deposition in month j , and $D_{BB}^j + D_{FF}^j = 1$. M is 3 (total number of months).

627 The regional average of fractional contributions from the BB and FF sectors from the
 628 PMF analysis and from the CAM5 simulation is calculated using Eqs. (A3) and (A4),
 629 respectively.

$$\overline{BB}_{obs} = \frac{\sum_{n=1}^N \overline{C}_{obs}^n \times BB_{obs}^n}{\sum_{n=1}^N \overline{C}_{obs}^n \times (BB_{obs}^n + FF_{obs}^n)} \quad \overline{FF}_{obs} = \frac{\sum_{n=1}^N \overline{C}_{obs}^n \times FF_{obs}^n}{\sum_{n=1}^N \overline{C}_{obs}^n \times (BB_{obs}^n + FF_{obs}^n)} \quad (A3)$$

631

$$\overline{BB}_{mod} = \frac{\sum_{n=1}^N \overline{C}_{mod}^n \times BB_{mod}^n}{\sum_{n=1}^N \overline{C}_{mod}^n \times (BB_{mod}^n + FF_{mod}^n)} \quad \overline{FF}_{mod} = \frac{\sum_{n=1}^N \overline{C}_{mod}^n \times FF_{mod}^n}{\sum_{n=1}^N \overline{C}_{mod}^n \times (BB_{mod}^n + FF_{mod}^n)} \quad (A4)$$

633 where N is the total number of observation/model comparison pairs (n) in a given region.

634

635

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Table 1. BB and FF fractional contributions based on the PMF and CAM5 source attribution results for BC in snow for each model/observation comparison pair (i). $\overline{C_{obs}^i}$ is the mean of the estimated BC concentrations used in the PMF analysis when more than one sampling sites reside in the same model grid box. $\overline{C_{mod}^i}$ is the JFM mean of CAM5 modeled BC concentrations in snow column. The contributions are calculated as given in Eqs. (A1) (observations) and (A2) (model).

| Comparison pair i | $\overline{C_{obs}^i}$ (ng g ⁻¹) | BB_{obs}^i (%) | FF_{obs}^i (%) | $\overline{C_{mod}^i}$ (ng g ⁻¹) | BB_{mod}^i (%) | FF_{mod}^i (%) |
|------------------------|---|---------------------|---------------------|---|---------------------|---------------------|
| 1 | 15.5 | 62 | 38 | 0.8 | 21 | 79 |
| 2 | 5.8 | 100 | 0 | 9.5 | 28 | 72 |
| 3 | 13.3 | 51 | 49 | 14.7 | 28 | 72 |
| 4 | 14.2 | 70 | 26 | 9.8 | 25 | 75 |
| 5 | 13.7 | 47 | 21 | 15.3 | 26 | 74 |
| 6 | 29.3 | 27 | 47 | 14.3 | 26 | 74 |
| 7 | 24.2 | 27 | 71 | 14.2 | 25 | 75 |
| 8 | 22.0 | 20 | 51 | 12.6 | 23 | 77 |
| 9 | 90.1 | 0 | 0 | 5.4 | 19 | 81 |
| 10 | 28.4 | 16 | 42 | 11.3 | 26 | 74 |
| 11 | 50.6 | 7 | 11 | 10.1 | 16 | 84 |
| 12 | 40.7 | 11 | 26 | 37.1 | 11 | 89 |
| 13 | 17.9 | 34 | 44 | 24.0 | 12 | 88 |
| 14 | 49.5 | 23 | 53 | 52.5 | 13 | 87 |
| 15 | 5.9 | 46 | 52 | 51.8 | 12 | 88 |
| 16 | 25.8 | 16 | 31 | 46.6 | 11 | 89 |
| 17 | 110.6 | 3 | 31 | 30.5 | 14 | 86 |
| 18 | 61.4 | 8 | 61 | 23.3 | 14 | 86 |
| 19 | 24.8 | 13 | 76 | 27.6 | 11 | 89 |
| 20 | 26.9 | 17 | 33 | 39.9 | 12 | 88 |
| 21 | 22.2 | 26 | 56 | 44.5 | 15 | 85 |
| 22 | 17.8 | 31 | 61 | 18.2 | 15 | 85 |
| 23 | 27.5 | 23 | 28 | 12.6 | 15 | 85 |
| 24 | 15.8 | 22 | 63 | 7.2 | 19 | 81 |
| 25 | 14.4 | 32 | 68 | 5.6 | 19 | 81 |
| 26 | 26.0 | 0 | 77 | 12.6 | 15 | 85 |
| 27 | 15.1 | 15 | 48 | 16.0 | 15 | 85 |
| 28 | 18.4 | 16 | 69 | 22.0 | 13 | 87 |
| 29 | 8.4 | 66 | 34 | 29.2 | 15 | 85 |
| 30 | 17.0 | 18 | 75 | 24.8 | 15 | 85 |
| 31 | 8.4 | 45 | 55 | 9.1 | 18 | 82 |
| 32 | 14.7 | 30 | 68 | 20.2 | 15 | 85 |
| 33 | 21.5 | 24 | 61 | 27.3 | 16 | 84 |
| 34 | 17.5 | 18 | 61 | 29.8 | 17 | 83 |
| 35 | 25.0 | 22 | 66 | 38.9 | 16 | 84 |

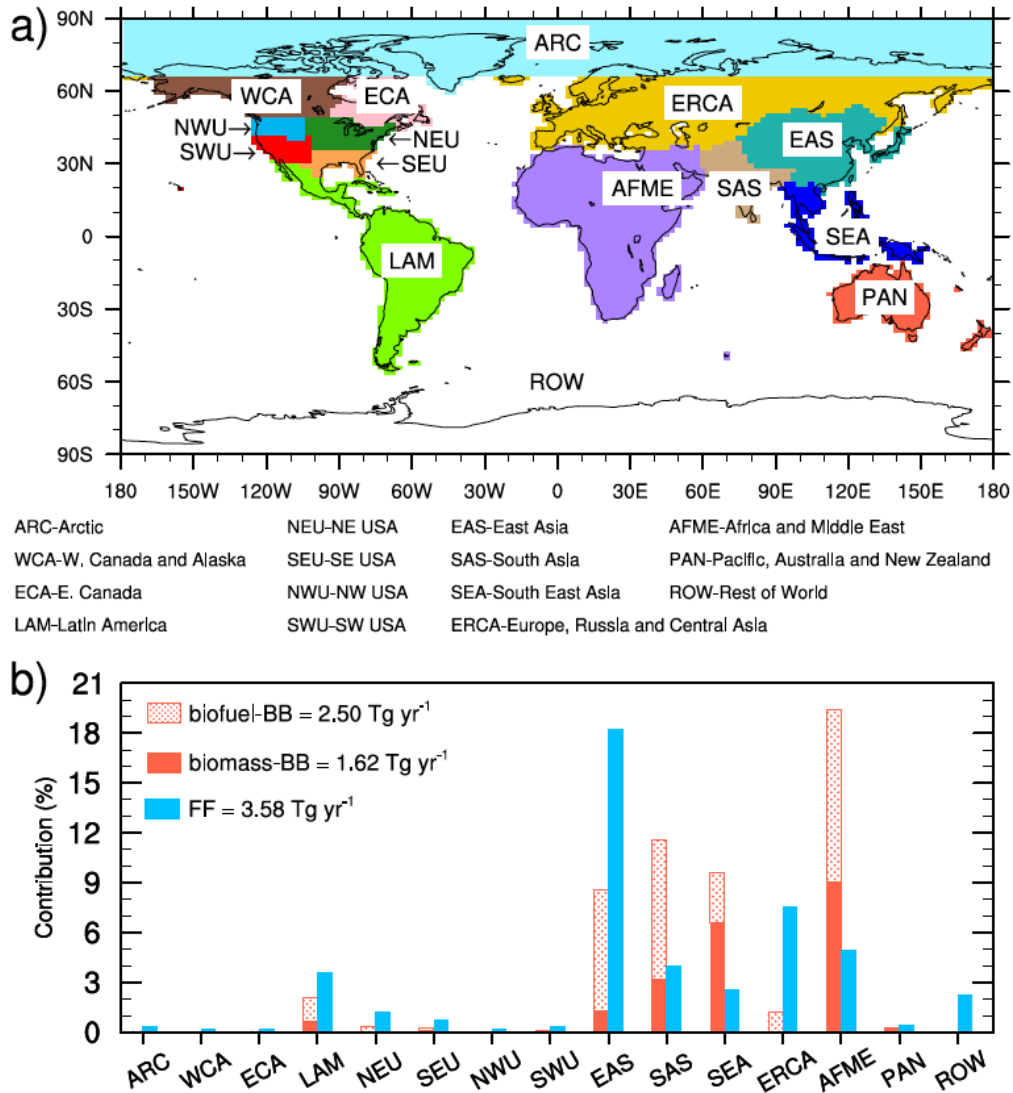


Fig. 1. (a) Tagged source regions and (b) the contributions (%) to the global mean BC emissions (7.69 Tg yr^{-1}) for January, February and March from the individual source regions (marked on the horizontal axis) and sectors (FF in blue, biomass-BB in solid red, and biofuel-BB in dotted red).

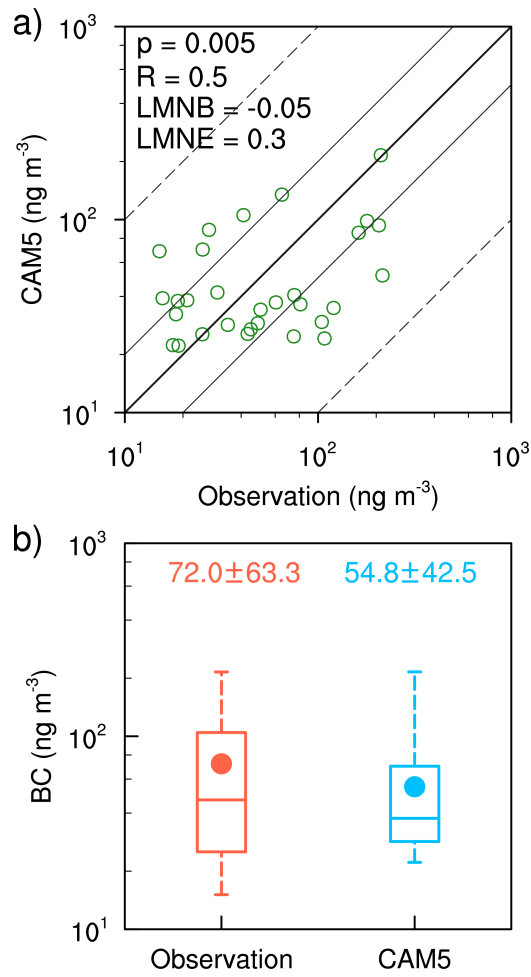


Fig. 2. (a) Scatter plot of CAM5 simulated versus observed JFM mean near-surface atmospheric BC concentrations (ng m^{-3}) in 2013 at the IMPROVE network sites. The observations are averages across sites falling into the same model grid box. The correlation coefficient (R), the statistical significance of R (p), the log-mean normalized bias ($LMNB$), and the log-mean normalized error ($LMNE$) are shown in numbers in the top-left corner; the 1:1 (thick solid), 2:1 (thin solid) and 10:1 (dashed) lines are also plotted for reference. (b) Box and whisker plot of observed (red color) and simulated (blue color) JFM mean of near-surface BC concentrations (ng m^{-3}) for all comparison pairs. The 25th, 50th, and 75th percentiles are marked with a box, the mean value with a dot, and the minimum and maximum values with whiskers; the colored numbers give the mean and standard deviation for the observed (red) and modeled values (blue).

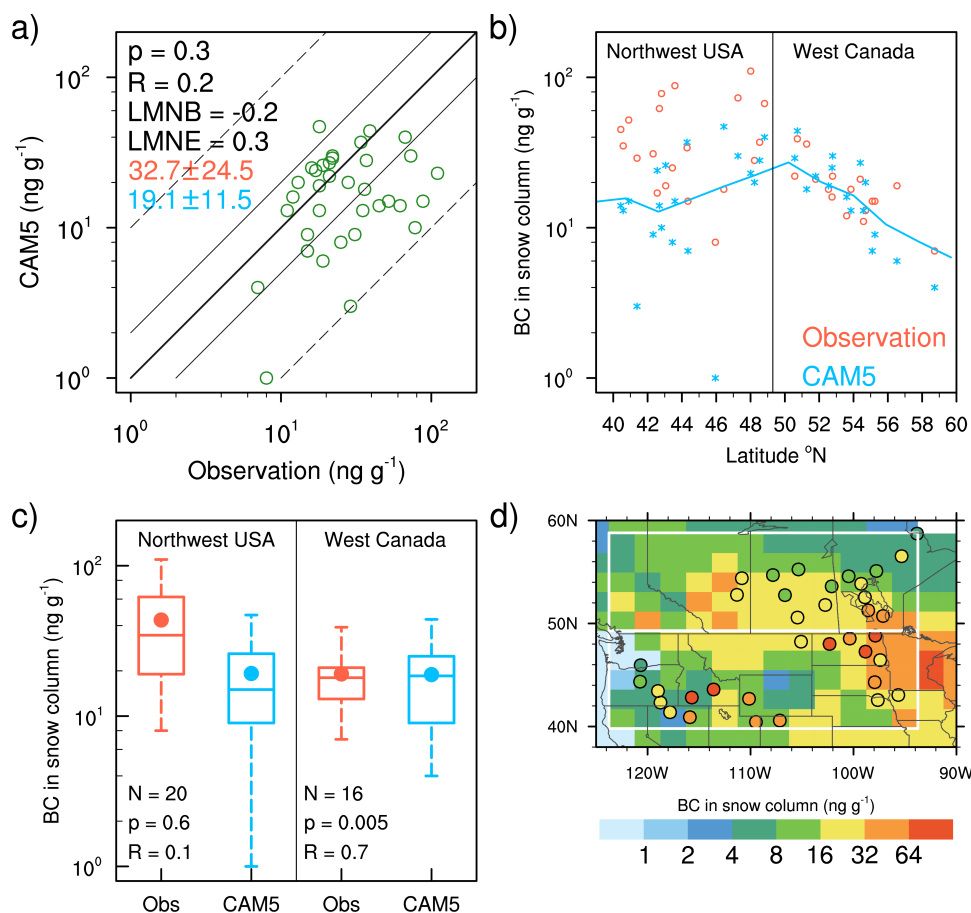


Fig. 3. (a) Scatter plot of simulated versus observed BC concentrations (ng g^{-1}) in the snow column (BCC). As in Figure 2, R , p , LMNB, and LMNE are shown in numbers on the top-left corner; the color numbers show the mean and standard deviation for observations (red) and modeled values (blue). (b) Observed (red circle) and simulated (blue asterisk) BCC versus latitude for the 36 comparison pairs in Northwest USA and West Canada. The modeled values are the JFM mean. The blue line indicates the modeled JFM zonal-mean values over the longitude band $93.75\text{--}123.75^\circ \text{W}$ (white outlines in panel d) for BCC. (c) Box and whisker plot of observed (red color) and simulated (blue color) BCC in the two regions. The 25th, 50th, and 75th percentiles are marked with a box, the mean value with a dot, and the minimum and maximum values with whiskers; the number of samples (N), R , and p for each region are shown at the bottom. (d) Spatial distributions of modeled JFM mean BCC with the observed BCC (color circles with black outlines) superimposed. In d) the observed values are averages across the sampling sites of Doherty et al. (2014), when more than one sampling site fell within a model grid box. The white boxes in d) outline the two receptor regions, Northwest USA ($39.8\text{--}49.3^\circ \text{N}$, $93.75\text{--}123.75^\circ \text{W}$) and West Canada ($49.3\text{--}58.8^\circ \text{N}$, $93.75\text{--}123.75^\circ \text{W}$).

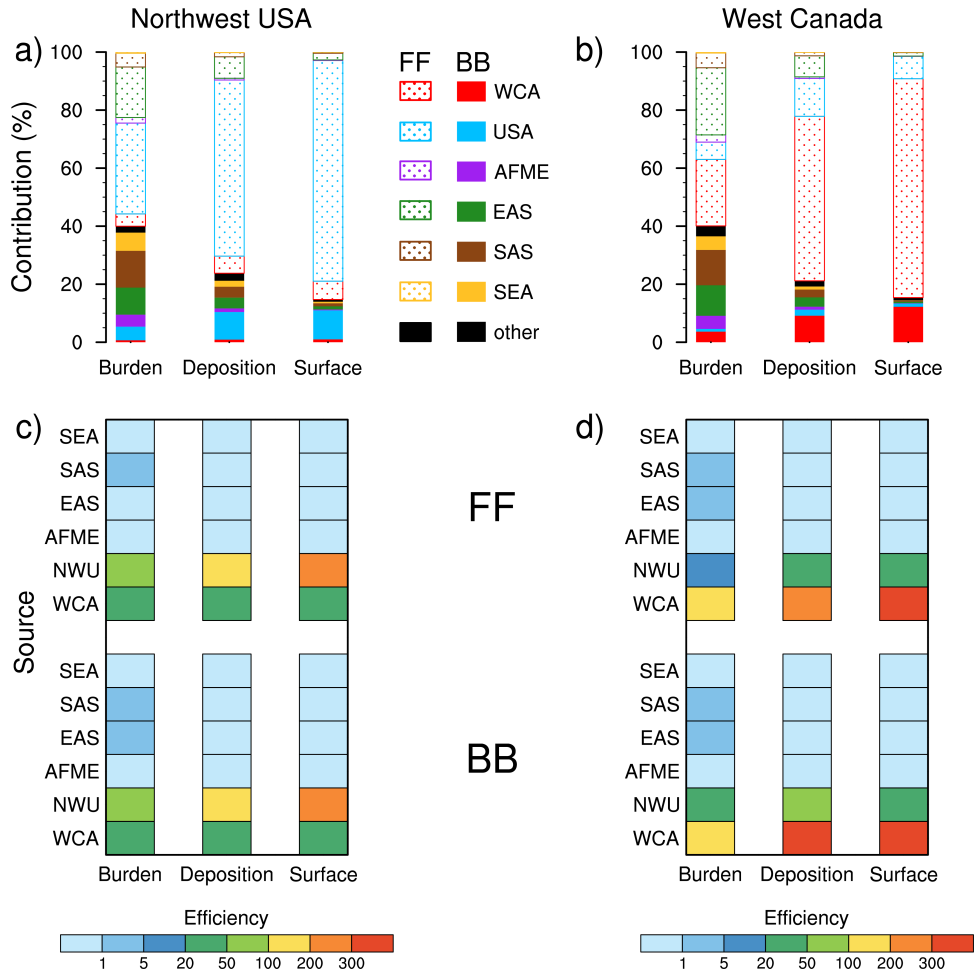


Fig. 4. Fractional contributions to JFM mean BC total column burden, deposition and near-surface concentrations over (a) Northwest USA and (b) West Canada (as defined in Fig. 3d), from six major tagged source regions (colors) and sectors (solid color and stippled bar for BB and FF, respectively); the black bar in each column represents the combined contribution from all of the other tagged source regions and sectors. Panels (c) and (d) show efficiency of FF (top) and BB (bottom) emissions from six major tagged source regions (marked on the y-axis) in changing JFM mean BC total column burden, deposition and near-surface concentrations over Northwest USA (c) and West Canada (d).

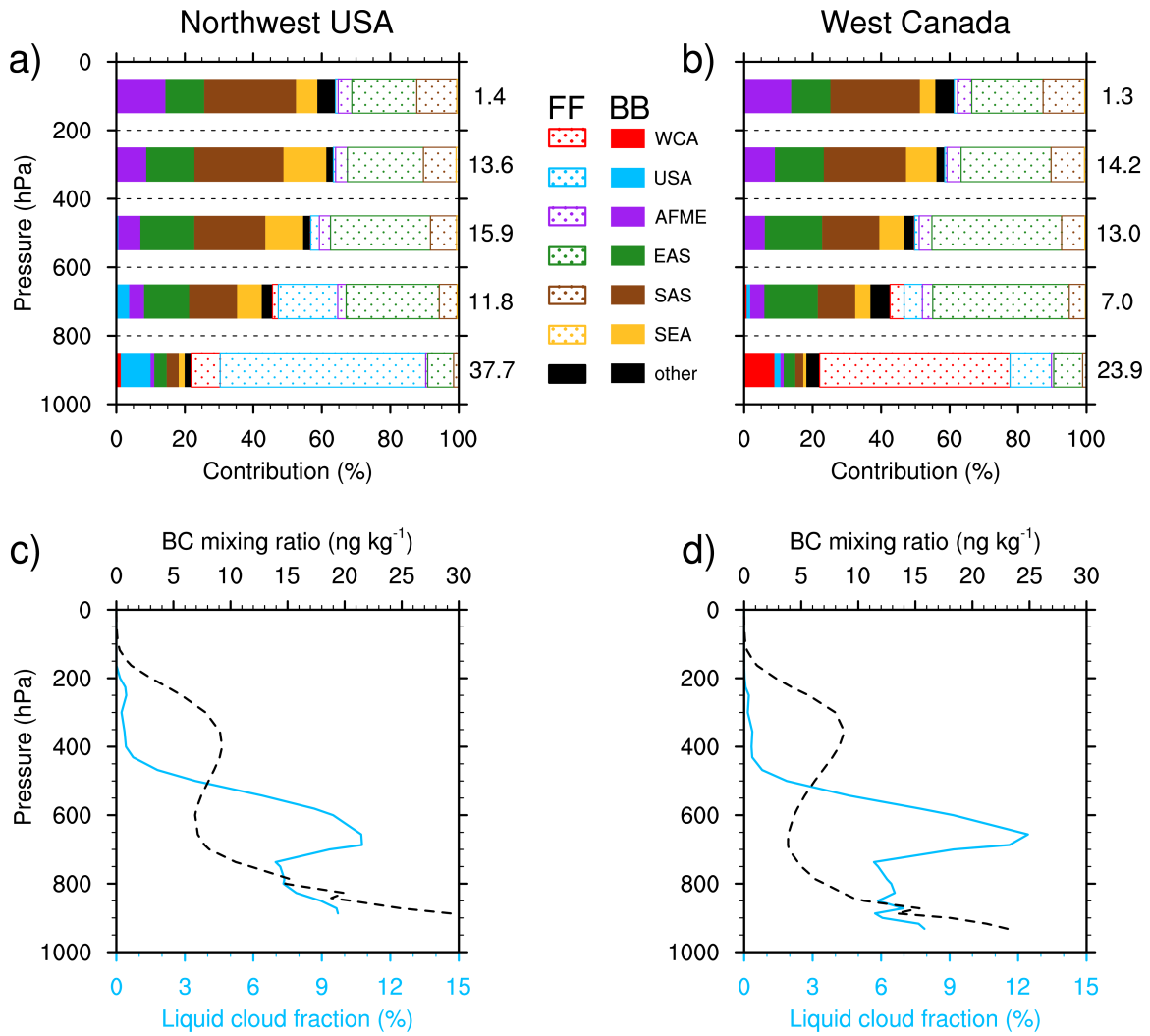


Fig. 5. Panels (a) and (b) are similar to Fig. 4a and b, respectively, but for fractional contributions to BC column burden in five separate vertical layers: 0–200, 200–400, 400–600, 600–800 and 800–1000 hPa. Panels (c) and (d) show the vertical profiles of area-averaged BC mixing ratio (in black) and liquid cloud fraction (in blue) over Northwest USA and West Canada, respectively. All fields are from the CAM5 model run.

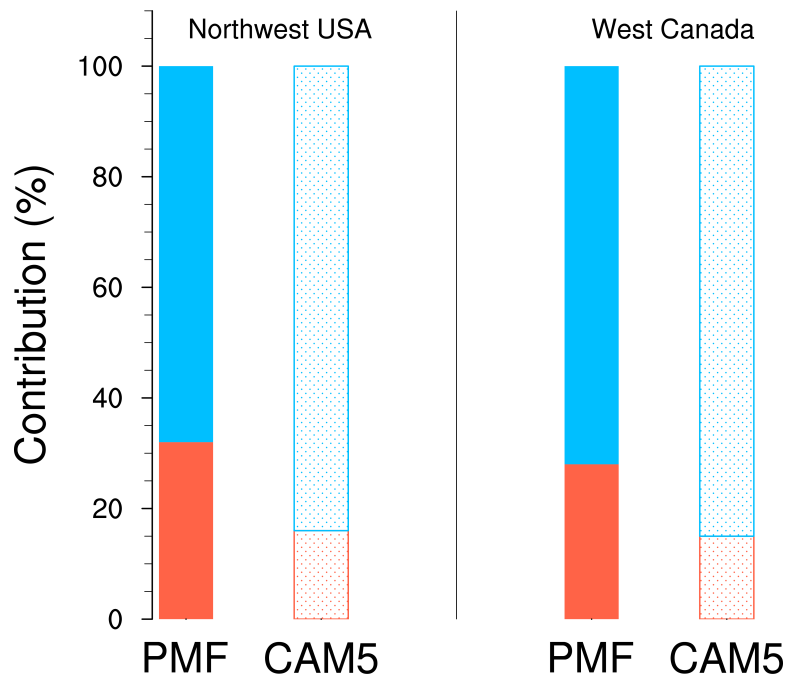


Fig. 6. Regional average contributions from BB (red color) and FF (blue color) sector to combustion-sourced BC in snow in Northwest USA and West Canada based on the PMF analysis (solid bar) and CAM5 simulation (stippled bar). The contributions are calculated as in Eqs. (A3) (observed values) and (A4) (modeled values).

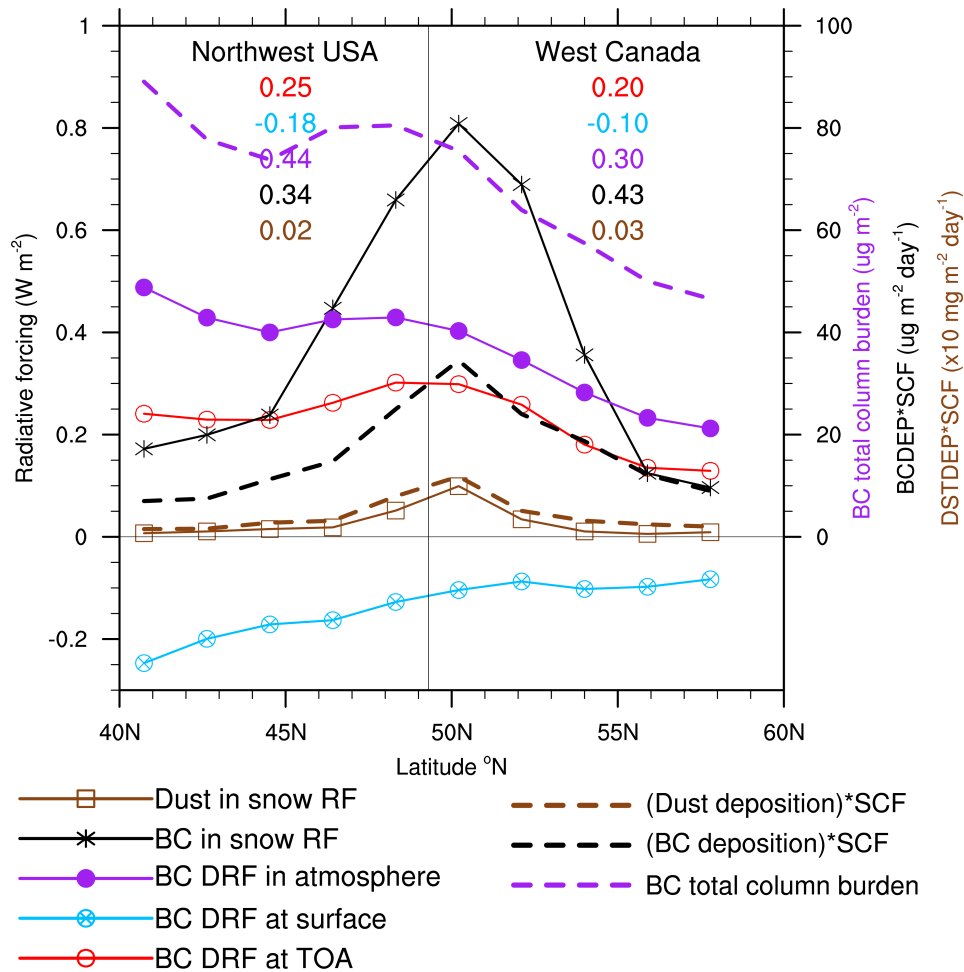


Fig. 7. Modeled JFM and zonal mean radiative forcing (RF) values (in $W m^{-2}$, using y-axis on the left) induced by the various BC effects and the dust-in-snow effect (indicated by the different colors and symbols in the legend) over the longitude band $93.75-123.75^{\circ} W$ (white outlines in Fig. 3d). The corresponding area-average RF values are shown in colored numbers for Northwest USA and West Canada, respectively. Modeled JFM and zonal mean values of BC total column burden (in $\mu g m^{-2}$), BC deposition (in $\mu g m^{-2} day^{-1}$) and dust deposition (in $10 mg m^{-2} day^{-1}$) multiplied by SCF (snow cover fraction) are shown in colored dashed lines (using y-axis on the right).