

Quantifying sources, transport, deposition and radiative forcing of black carbon over the Himalayas and Tibetan Plateau

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Abstract

1
2 Black carbon (BC) particles over the Himalayas and Tibetan Plateau (HTP), both airborne and
3 those deposited on snow, have been shown to affect snowmelt and glacier retreat. Since BC over
4 the HTP may originate from a variety of geographical regions and emission sectors, it is essential
5 to quantify the source-receptor relationships of BC in order to understand the contributions of
6 natural and anthropogenic emissions and provide guidance for potential mitigation actions. In
7 this study, we use the Community Atmosphere Model version 5 (CAM5) with a newly
8 developed source tagging technique, nudged towards the MERRA meteorological reanalysis, to
9 characterize the fate of BC particles emitted from various geographical regions and sectors.
10 Evaluated against observations over the HTP and surrounding regions, the model simulation
11 shows a good agreement in the seasonal variation of the near-surface airborne BC concentrations,
12 providing confidence to use this modeling framework for characterizing BC source-receptor
13 relationships. Our analysis shows that the relative contributions from different geographical
14 regions and source sectors depend on season and location in the HTP. The largest contribution to
15 annual mean BC burden and surface deposition in the entire HTP region is from biofuel and
16 biomass (BB) emissions in South Asia, followed by fossil fuel (FF) emissions from South Asia,
17 then FF from East Asia. The same roles hold for all the seasonal means except for the summer
18 when East Asia FF becomes more important. For finer receptor regions of interest, South Asia
19 BB and FF have the largest impact on BC in Himalayas and Central Tibetan Plateau, while East
20 Asia FF and BB contribute the most to Northeast Plateau in all seasons and Southeast Plateau in
21 the summer. Central Asia and Middle East FF emissions have relatively more important
22 contributions to BC reaching Northwest Plateau, especially in the summer. Although local
23 emissions only contribute about 10% of BC in the HTP, this contribution is extremely sensitive

24 to local emission changes. Lastly, we show that the annual mean radiative forcing (0.42 W m^{-2})
25 due to BC in snow outweighs the BC dimming effect (-0.3 W m^{-2}) at the surface over the HTP.
26 We also find strong seasonal and spatial variation with a peak value of 5 W m^{-2} in the spring
27 over Northwest Plateau. Such a large forcing of BC in snow is sufficient to cause earlier snow
28 melting and potentially contribute to the acceleration of glacier retreat.

29

30 **1 Introduction**

31 Black carbon (BC) is a distinct type of carbonaceous particulate matter mainly emitted from the
32 incomplete combustion of fossil fuels, biofuels and biomass burning. It is the dominant insoluble
33 light-absorbing particulate species, both in the atmosphere and after deposition on snow and ice.
34 In addition to its impact on air quality, BC plays a unique and important role in the climate
35 system through its effect on radiation, clouds and snow albedo, and associated feedbacks that
36 modify atmospheric circulation patterns and/or accelerate the snowmelt and glacier retreat in the
37 Arctic and across the mid-latitudes of the northern hemisphere (Bond et al., 2013).

38 Modeling studies (e.g., Hansen et al., 2005; Qian et al., 2011) indicate that the climate
39 efficacy of BC in snow is much greater than efficacy of carbon dioxide or other anthropogenic
40 forcings owing to a sequence of positive feedback mechanisms (Warren and Wiscombe, 1980,
41 1985; Conway et al., 1996; Hansen and Nazarenko, 2004; Jacobson, 2004; Flanner et al., 2007;
42 Ye et al., 2012; Hadley and Kirchstetter, 2012; Doherty et al., 2014). Flanner et al. (2009)
43 demonstrated that the global annual BC snow-albedo effect (darkening) outweighs the aerosol
44 (BC and organic matter) dimming effect (i.e., reduced the downwelling irradiance reaching the
45 surface) by a factor of about 6. The snow/ice-covered Himalayas and Tibetan Plateau (HTP)
46 region is more prone to these BC effects than other regions because of the surrounding two major

47 BC source regions, East Asia and South Asia, at present and likely in the future (e.g., Bond et al.,
48 2007; Ohara et al., 2007; Xu et al., 2009; Lamarque et al., 2010; Menon et al., 2010).

49 The HTP, often referred to as the Third Pole, has received much less scientific attention
50 than the Polar Regions (Qiu, 2008), although it is the highest and largest plateau that stores one
51 of the largest ice masses of the Earth system. The HTP also has a large area of seasonal and
52 permanent snow cover and represents the most sensitive and visible indicator of climate change
53 with its unique location for complex interactions among the atmosphere, hydrosphere and
54 cryosphere (e.g., Pu et al., 2007; Xu et al., 2009; Yao et al., 2012). The glaciers and the
55 associated snowmelt over the HTP have a great potential to modify the regional hydrology and to
56 trigger natural hazards that impact a large portion of the population in and around the region (e.g.,
57 Barnett et al., 2005; Singh and Bengtsson, 2004; Xu et al., 2008; Kaser et al., 2010; Immerzeel et
58 al., 2010; Yao et al., 2012; Bolch et al., 2012). The HTP also exerts profound influences on
59 atmospheric circulation patterns and climate through mechanical and thermal effects due to its
60 large area, highly elevated topography and geographical location in the Earth system (Yeh et al.,
61 1957; Manabe and Terpstra, 1974; Ye and Gao, 1979; Yanai et al., 1992; Ye and Wu, 1998; Wu
62 et al., 2012). The HTP acts as a giant wall across the Eurasian continent that blocks cold
63 outbreaks from high latitudes in winter and confines the winter monsoon to eastern and southern
64 Asia, while in summer the HTP serves as a huge heat source through the strong surface sensible
65 heating and latent heating over central and eastern Plateau (Wu et al., 2012).

66 Under the background of global warming, the climate of the HTP is changing rapidly. For
67 example, the surface sensible heat flux has weakened in recent decades, mainly due to global
68 warming (Duan and Wu, 2008). Observational evidence indicated that the surface air
69 temperatures on the HTP have increased about 1.8°C over the past 50 years (Wang et al., 2008),

70 while the large area at elevations above 4000 m has warmed at 0.3°C per decade in the past three
71 decades (Xu et al., 2009). A number of recent studies reported that glaciers on the HTP have
72 undergone widespread losses at an increasing rate in past decades (e.g., Qin et al., 2006; Li et al.,
73 2008; Kang et al., 2010; Bolch et al., 2012) and have undergone accelerated retreat in recent
74 years (Yao et al., 2007). The rapid warming and the accelerated glacier retreat have been
75 primarily attributed to increasing greenhouse gases (e.g., Duan et al., 2006; Ren et al., 2006), but
76 other factors may be partly responsible for the accelerated warming over the HTP, such as
77 atmospheric heating by absorbing aerosols, land use changes, and reduction of snow albedo
78 induced by light-absorbing impurities in snow (Kang et al., 2000; Prasad and Singh 2007;
79 Ramanathan et al., 2007; Flanner et al., 2007, 2009; Yasunari et al., 2010; Xu et al., 2009; Qian
80 et al., 2011, 2015). Lau et al. (2006, 2010) proposed and demonstrated the Elevated Heat Pump
81 mechanism, whereby heating induced by airborne BC and dust absorption can strengthen local
82 circulations and lead to a northward shift of the monsoon rain belt, widespread enhanced
83 warming over the HTP, and accelerated snowmelt and glacier retreat. Previous observational and
84 modeling studies have indicated that BC deposition on snow and ice, which has a rapidly
85 increasing trend in recent years, has been a significant contributor to the early snowmelt and
86 rapid glacier retreat over the HTP (e.g., Flanner et al., 2007, 2009; Ming et al., 2008; Xu et al.,
87 2009; Kaspari et al., 2011; Menon, et al., 2010; Qian et al., 2011, 2015; Wang et al., 2015).
88 Flanner et al. (2007) found that the largest regional annual mean forcing due to BC in snow is
89 located in the HTP. Xu et al. (2009) and Lau et al. (2010) suggested that the BC in snow/ice may
90 be partly responsible for the observed acceleration of glacier retreat in the HTP.

91 Understanding the role of BC in accelerating snow-cover reduction and glacier retreat is
92 becoming increasingly important. Over 60% of BC in the present-day atmosphere originates

93 from anthropogenic activities (e.g., Bond et al., 2007; Lamarque et al., 2010). Reduction of
94 emissions from BC-rich sources represents a potential mitigation strategy to slow down present-
95 day climate change because BC has a positive radiative forcing but a short atmospheric lifetime
96 (Bond et al., 2013). Since BC over the HTP may originate from a variety of geographical regions
97 and emission sectors, it is essential to quantify the source-receptor relationships of BC in order to
98 understand the contributions of open fire and anthropogenic emission sectors to BC over the
99 HTP. This exercise is also essential to provide guidance for potential mitigation actions.

100 Some studies have used the conventional back-trajectory approach to identify possible
101 source regions for both airborne BC and that deposited on snow and ice, by tracking air mass
102 reaching sampling sites over the HTP (e.g., Ming et al., 2008, 2009; Cao et al., 2009; Bonasoni
103 et al., 2010; Zhao et al., 2013; Zhang et al., 2013). Lu et al. (2012) developed a novel back-
104 trajectory approach to analyze the origin of BC transported to the HTP during 1996-2010. They
105 derived the overall transport characteristics of BC to the HTP and showed the spatial distribution
106 of sources for BC reaching the HTP region based on a large set of seven-day back trajectories
107 arriving at the given height (i.e., 500 m) and receptor locations, BC emissions and transport
108 efficiencies. The statistical analysis of trajectories has good accuracy on short time scales for
109 source regions with close proximity to the receptor, but this approach has limitations in
110 determining contributions from distant sources to BC in the mid- and upper-troposphere that
111 could contribute significantly to the total column burden but less to BC deposition and boundary-
112 layer concentrations. Using the adjoint of the GEOS-Chem global chemical transport model,
113 Kopacz et al. (2011) attempted to identify the originating locations of BC arriving at five glacier
114 sites (i.e., five model grid-cells as the receptors) in the HTP for year 2001. This method can
115 provide a global distribution of emissions that directly contribute to BC concentrations at

116 receptor locations. Note that the adjoint model results are not source attributions but rather the
117 source-receptor sensitivities, which can be interpreted as the effectiveness of incremental
118 changes to existing emissions in affecting BC at receptor locations. While the adjoint approach
119 has the advantage of not predefining source regions, it does require performing separate
120 simulations for each of the defined receptor regions.

121 In this study, we use an aerosol-climate model with a newly developed explicit source
122 tagging approach (Wang et al., 2014) to produce a detailed characterization of the fate of BC
123 emitted from various geographical regions and sectors (e.g., fossil fuel, biofuel and biomass
124 burning emissions) and transport pathways to the HTP. In contrast to the back-trajectory and the
125 adjoint approaches, the direct tagging method has the flexibility to do source attribution of BC
126 mass mixing ratio at any model layer and the surface dry and/or wet deposition within a single
127 simulation for any receptor regions. Section 2 describes the aerosol-climate model and the
128 tagging method used in this study. Section 3 presents an evaluation of modeled BC surface
129 concentrations and seasonal snow cover over the HTP region. The transport pathways and source
130 attribution results are presented in Sect. 4. The radiative effects of BC in the atmosphere and of
131 both BC and mineral dust in snow are compared in Sect. 5, followed by the summary and
132 conclusions in Sect. 6.

133

134 **2 Model Configuration and Experimental Design**

135 **2.1 The CAM5 model and the source-tagging method**

136 We use the Community Atmosphere Model Version 5 (CAM5; Neale et al., 2012), which is the
137 atmospheric component of the Community Earth System Model version 1 (CESM1) (Hurrell et
138 al., 2013). It includes relatively comprehensive representations of aerosols and clouds, and

139 mechanisms for their interactions with each other and with climate (Gettelman et al., 2010; Liu
140 et al., 2012). CAM5 employs a modal aerosol module (MAM) to represent aerosols in multiple
141 log-normally distributed modes, with internal mixing assumed for aerosol species within each
142 individual mode, including a 3-mode standard representation (MAM3) and a more complex 7-
143 mode representation (MAM7). The major difference between MAM3 and MAM7 related to
144 carbonaceous aerosols lies in the treatment of aging. In MAM3, BC and primary organic matter
145 (POM) particles are emitted into the accumulation mode that also contains highly hygroscopic
146 species such as sulfate and sea-salt, while in MAM7 BC and POM are emitted into a primary
147 carbon mode, which contains no other species. BC is hydrophobic upon emission, and thus the
148 hygroscopicity of the primary-carbon-mode particles depends on the assumed hygroscopicity for
149 POM. As more hygroscopic species (e.g., H_2SO_4 and NH_3) condense onto the primary-carbon-
150 mode particles, the particles are become more hygroscopic and are gradually transferred into the
151 MAM7 accumulation mode. The rate of transfer is controlled by uncertain aging parameters, and
152 the availability of gas precursors (Liu et al. 2012). In the accumulation mode of both MAM3 and
153 MAM7, BC is internally mixed with other more hygroscopic species and is thus subject to wet
154 scavenging and removal processes. During the transport from sources to remote regions, aerosols
155 are removed too efficiently in the default CAM5 (Liu et al., 2012). Recently, H. Wang et al.
156 (2013) revised some key processes associated with aerosol wet removal and convective transport,
157 which significantly improved the vertical distribution of aerosols and their transport to remote
158 regions such as the Arctic.

159 To better characterize the sensitivity of BC spatial distributions to emission uncertainties,
160 Wang et al. (2014) implemented a direct source tagging method in CAM5, whereby BC emitted
161 from a number of independent source regions and/or sectors can be tagged and explicitly tracked

162 within a single model simulation. This approach provides the quantitative characterization of
163 source-receptor relationships for BC in any receptor region without perturbing emissions from
164 individual BC source regions or sectors. In this study, we apply the BC tagging technique to the
165 accumulation-mode BC in the MAM3 treatment. BC particles emitted from sixteen geographical
166 BC source regions and two emissions sectors (i.e., biomass burning & biofuel emissions and
167 fossil fuel emissions) in each of the regions are tagged and explicitly tracked. Instead of using
168 the global emissions from all sectors for the original one BC mass mixing ratio variable, the
169 thirty two regional/sectoral emissions provide sources to the respective tagged BC mass mixing
170 ratio variables that are all added to the accumulation mode, including both interstitial and cloud-
171 borne states. All physical and dynamic tendencies (e.g., transport, dry and wet removal) are
172 calculated explicitly for the tagged BC mass mixing ratio variables in the same way as the
173 original single BC mass mixing ratio. Also, when aerosol optical properties are calculated, all of
174 the tagged BC mass mixing ratios contribute to the volume-mean refractive index of the
175 accumulation mode that is used in the radiation calculation.

176 In addition to the free-running mode, CAM5 can also be configured in an offline mode,
177 in which temperature, wind, surface fluxes (heat, moisture, and momentum), and pressure are
178 constrained to agree closely with observations, while clouds and aerosol are allowed to evolve
179 freely (Rasch et al., 1997; Lamarque et al., 2012; Ma et al., 2013). In this study, we run the
180 CAM5 model in the offline mode with the direct BC source tagging capability, including the
181 improved representation of convective transport and wet removal of aerosols. We use the NASA
182 Modern Era Retrospective-Analysis for Research and Applications (MERRA) reanalysis dataset
183 (Rienecker et al., 2011), using a horizontal resolution of $1.9^\circ \times 2.5^\circ$ and 56 vertical levels. The
184 goal is to characterize the fate of BC emitted from various geographical regions and sectors, their

185 transport pathways to the HTP, and their radiative forcing with seasonal variations. The
186 simulation is performed for year 2001 with prescribed sea surface temperatures.

187 **2.2 BC source regions and sectors**

188 BC emission datasets have large uncertainties (e.g., Bond et al., 2013), and there are different
189 inventories available for climate modeling. We use the present-day (i.e., year 2000) monthly
190 mean emission inventories for BC provided by Lamarque et al. (2010). They were built for the
191 climate model simulations in the Coupled Model Intercomparison Project Phase 5 (CMIP5)
192 (Taylor et al., 2012) performed for the fifth assessment report (AR5) of the Intergovernmental
193 Panel on Climate Change (IPCC). The AR5 BC emissions being used in our CAM5 simulation
194 include monthly varying elevated open fire emissions (injection altitude up to 6 km), and yearly
195 constant surface emissions from shipping and from six sectors over land: agricultural waste
196 burning, domestic, energy, industry, transportation, and waste treatment. These surface BC
197 emissions sectors do not distinguished between biofuel and fossil fuel combustion. To prepare
198 for the BC source sector tagging, we divide the total surface emissions into two broader sectors,
199 biofuel and fossil fuel, by using the ratio of biofuel to biofuel plus fossil fuel at each model grid
200 provided by Dentener et al. (2006). We then combine the biomass burning (open fire) emissions
201 and surface biofuel emissions, hereafter, referred to as BB (biofuel and biomass) sector. The
202 shipping emissions are combined with the fossil fuel emissions over land to form the FF (fossil
203 fuel) sector. Note that emissions in the BB sector have seasonal variations (associated with the
204 open fire emissions) but the FF sector emissions used in this study have no seasonal variation at
205 all.

206 The sixteen geographical BC source regions (Fig. 1a) are defined using the definition of
207 source/receptor regions by Work Plan (WP 2.1) of the Task Force on Hemispheric Transport of

208 Air Pollution (<http://iek8wikis.iek.fz-juelich.de/HTAPWiki/WP2.1>). They are ARC (Arctic),
209 NAM (North America), CAM (Central America), SAM (South America), EUR (Europe), NAF
210 (North Africa), SAF (South Africa), MDE (Middle East), CAS (Central Asia), SAS (South Asia),
211 EAS (East Asia), SEA (South East Asia), PAN (Pacific, Australia and New Zealand), RBU
212 (Russia, Belarus and Ukraine), HTP (Himalayas and Tibetan Plateau) and ROW (Rest of World).

213 Figure 1b and Table S1 in the Supplement summarize the fractional contributions of BC
214 emissions from the different source regions and sectors. The global annual mean BC emission
215 rate is 7.78 Tg yr^{-1} , with 56.2% (sum of the red bars) from BB emissions (33.6% from fires and
216 22.6% from biofuel) and 43.8% (sum of the blue bars) from FF emissions. The two largest
217 contributors are BB emissions from SAF (about 20%) and FF emissions from EAS (about 15%),
218 followed by BB emissions from SEA (7.7%), EAS (6.4%), SAS (6.2%) and SAM (5.7%), and
219 EUR FF (6.4%) emissions. The geographical distributions of BC annual mean emission fluxes
220 from BB and FF sectors for year 2000 are shown in Fig. S1 (in Supplement). The global annual
221 and seasonal mean lifetime of BC emitted from the tagged source regions and sectors are
222 summarized in Table S2. On the globe average, BB BC has a longer lifetime than FF BC in all
223 seasons, especially in boreal winter (6.9 vs. 3.1 day), due in part to higher open-fire emissions (in
224 the BB sector) during local dry seasons. Another reason is that open-fire emissions have initial
225 injection heights of up to 6 km, resulting in less removal below 6 km. The availability of co-
226 emitted hygroscopic species that are internally mixed with BC in the accumulation mode of the
227 MAM3 aerosol treatment also impacts the scavenging and wet removal rate of BC. This also in
228 part explains the variability of BC lifetime among the different source regions and sectors.
229 Regarding the seasonal cycle, BC emitted from the major source regions (e.g., SAF, EAS, SEA,

230 SAS) has substantially lower lifetime in summer (JJA) than in the other seasons, likely due to
 231 relatively strong removal by the summer monsoon precipitation.

232 We use two metrics for quantifying source-receptor relationships and the sensitivity of
 233 BC in a receptor region to various sources following Wang et al. (2014), but we extend them to
 234 treat BB and FF sectors separately.

235 1) The fractional contribution of BB and FF emissions from source region i to a BC property in
 236 receptor region j (the entire HTP or a subset of it), C_i^{BB} or C_i^{FF} , is defined as

$$237 \quad C_i^{BB} = \frac{A_i^{BB}}{A_j}, C_i^{FF} = \frac{A_i^{FF}}{A_j} \quad (1)$$

238 Where A_i^{BB} and A_i^{FF} are a BC property (e.g., mass mixing ratio, column burden, or deposition
 239 flux) in/over receptor region j resulting from BB and FF emissions, respectively, in source region
 240 i , and $A_j = \sum_{i=1}^N (A_i^{BB} + A_i^{FF})$ represents the total BC property in the receptor region from all
 241 source regions ($N = 16$ in this study) and sectors (BB and FF). Note that for BC properties such
 242 as column burden, surface mixing ratio, and deposition flux, the tagging method in CAM5
 243 explicitly calculates how much is due to emissions from each source region and sector.

244 2) The efficiency of BB and FF emissions from source region i in changing BC in a receptor
 245 region j is defined as

$$246 \quad S_i^{BB} = \frac{C_i^{BB}}{E_i^{BB}/E_{tot}}, S_i^{FF} = \frac{C_i^{FF}}{E_i^{FF}/E_{tot}} \quad (2)$$

247 Where C_i^{BB} and C_i^{FF} are the fractional contribution defined in Eq. (1), and E_i^{BB} and E_i^{FF} are the
 248 total BB and FF emission rates, respectively, in source region i , and $E_{tot} = \sum_{i=1}^N (E_i^{BB} + E_i^{FF})$ in
 249 Eq. (2) represents the global total emission rate. The efficiency metric S_i^{BB} or S_i^{FF} characterizes
 250 the sensitivity of aerosol properties in a receptor region to per unit (BB or FF) emissions in
 251 source region i . As noted in Wang et al. (2014), the efficiency is proportional to the metric of

252 relative contribution per unit source region emission used by Shindell et al. (2008). Physically,
253 the efficiency metric can be viewed as the efficiency of transport from a source region to the
254 receptor region. This metric is perhaps of more interest to policy makers for the purpose of
255 mitigation action, which is not the focus of this study but is worth mentioning.

256

257 **3 Model evaluation against available observations**

258 The CAM5 model has been evaluated in detail from different perspectives with available
259 observations such as aerosol mass concentration, aerosol number concentration and size
260 distribution, aerosol optical properties, cloud properties, aerosol deposition and BC in snow over
261 various regions in previous studies (Liu et al., 2012; H. Wang et al., 2013; Ma et al., 2013; Jiao
262 et al., 2014; Lee et al., 2013; Qian et al., 2014). Because of the complex topography and
263 meteorology of the HTP and the relatively coarse resolution of global model, further model
264 evaluation focusing on the HTP region is critical. Here we use near-surface atmospheric BC
265 concentrations measured at a few HTP sites and the snow cover fraction retrieved from satellite
266 to evaluate the CAM5 performance in the HTP.

267 **3.1 Atmospheric BC surface concentration**

268 There are seven remote sites that have surface measurements of seasonal BC aerosol
269 concentrations available. The locations and elevations of the sites and the sampling time periods
270 and observation methods are described in Table 1. Figure 2 shows the comparison of seasonal
271 mean BC concentrations between observations and CAM5 results. Note that model results
272 represent mean concentrations in the grid box that the sampling sites reside in and at the grid-
273 mean elevation, which could deviate significantly from the sampling point near complex terrain.
274 All sites have non-negligible amounts of BC in the near-surface air. The error bars indicate the

275 intra-seasonal and inter-annual variations if multi-year data were used for given season and site.
276 However, the uncertainties of observed BC surface concentrations mainly originate from the
277 large discrepancies between different measurement methods, the mixing of BC with other
278 components (e.g., organic carbon and mineral dust) in the aerosol samples, and the sampling
279 time and location (Bond et al., 2013; Petzold et al., 2013). BC surface concentrations over the
280 various sites show strong seasonal variations, which are reasonably captured by the model. The
281 modeled magnitude of BC concentrations has a good agreement with observations at some sites
282 (e.g., Fig. 2b, d, g), but the model clearly overestimates BC at the Muztagh Ata site (Fig. 2a) and
283 underestimates at the Lulang site (Fig. 2e). The large underestimation (about 1000 m; see Table
284 1) of the Muztagh Ata site elevation in the model, determined by the model grid resolution, could
285 largely explain the overestimation of BC since BC concentrations have sharp decreases with
286 height in this region. At the sites over southern HTP (i.e., Hanle, Manora Peak, NCO-P, Lulang
287 and NCOS), the BC surface concentrations in the summer (JJA) are lower, mainly due to wet
288 scavenging by more frequent precipitation and partly due to the minimal emissions from
289 domestic heating and wildfires over Himalaya foothills and Indo-Gangetic Plains (IGP) during
290 the Indian summer monsoon season (Marinoni et al., 2010, 2013). Among all these sites, the
291 largest BC surface concentrations occur at the Manora Peak site that is closer to the major
292 sources in South Asia, especially in the winter (DJF) when the model underestimates the
293 concentrations by about 50%. The high concentrations in winter at Manora Peak is mainly due to
294 the dry winter monsoon conditions and increased transport of emissions from regional biomass
295 burning, agricultural waste and wood fuel burning from the IGP (e.g., Ram et al., 2010; Moorthy
296 et al., 2013). The BC surface concentrations peak in the springtime (MAM) at Hanle, NCO-P,
297 Lulang and NCOS sites. This might be related to an increase in BB and/or FF emissions in the

308 Indian Subcontinent, along with the higher regional boundary-layer top over the IGP during the
309 springtime that may favor the transport of particles from the surface up to higher altitudes (e.g.,
300 Marinoni et al., 2010, 2013). Moreover, a long-range transport of pollution emitted from distant
301 regions like the Middle East, North Africa or Europe (Marinoni et al., 2010) could further
302 contribute to BC variability over the South Himalayas, which will also be examined in this study.
303 Part of the discrepancies between observations and model results can be attributed to the inherent
304 difficulty in simulating the cloud/precipitation fields over the complex topography and
305 subsequent wet removal of aerosols during the transport, but emission uncertainties (e.g., Bond
306 et al., 2013) might play a primary role.

307 **3.2 Snow cover fraction**

308 It is important to evaluate the performance of model in simulating seasonal snow over this region
309 in order to assess the importance of BC-in-snow effect. Figure 3 shows the CAM5 simulated
310 seasonal and annual mean snow cover fraction (SCF) during year 2001, in comparison to
311 observed mean SCF, derived from the Moderate Resolution Imaging Spectrometer (MODIS)
312 (Hall et al., 2006) monthly mean of daily products at 0.05 degree resolution. For a better
313 comparison, the MODIS monthly mean SCFs are mapped to the CAM5 grid. The summer (JJA)
314 season only includes July and August for both CAM5 and MODIS due to missing MODIS data
315 in June 2001. To illustrate whether year 2001 can represent the average condition in terms of
316 SCF, the MODIS SCF climatology (2000-2013) is also plotted. The overall SCF in HTP has very
317 small difference between climatology and year 2001 in all seasons except for JJA, when SCF is
318 notably higher over northwest Plateau for the climatology that included June SCF in the average.
319 On average, SCF is about 5% (absolute amount) higher in June than in July and August. Over the
320 52 HTP grid cells, the CAM5 SCF is highly correlated spatially with that of MODIS (for both

321 2001 and 2000-2013) with the statistical confidence level greater than 99%, except for summer
322 (JJA) when the linear correlation is significant only at 80% level.

323 There are strong spatial and seasonal variations in SCF due to the complex terrain and
324 seasonal variation in snowfall and melting. The SCF over the entire HTP reaches the maximum
325 in the winter (DJF), while decreases to almost none (less than 5%) in July and August. Snow
326 covers the western and southeastern Plateau during the transition seasons (MAM and SON). The
327 CAM5 simulation shows a good agreement with MODIS in the annual mean (ANN) SCF and the
328 strong seasonality. The most persistent snow cover at the southern and western edges of the HTP
329 and the relatively less persistent in the HTP interior are captured by the CAM5 model. The
330 performance of the CAM5 has been improved, in comparison to its earlier version (CAM3) that
331 remarkably overestimated the SCF especially over the HTP interior (Qian et al., 2011), although
332 the CAM5 still significantly overestimates the SCF in the western Plateau in DJF and MAM and
333 underestimates it in JJA. The CAM3 model used by Qian et al. (2011) overestimates SCF by up
334 to a factor of 2 during the cold season (November to April). The CAM3 spring (MAM) mean
335 SCF is greater than 35%, while the CAM5 spring mean (21%) in the present study is in good
336 agreement with the MODIS spring SCF ($18\pm 5\%$).

337 Although we believe that the CAM5 SCF biases are qualitatively robust, it is worth
338 noting that the MODIS products have uncertainties as well. Pu et al. (2007) evaluated the
339 MODIS SCF products over the HTP against ground-based snow observations and showed that
340 total error in MODIS SCF products over the HTP is about 10%. However, their analysis based
341 on MODIS eight-day snow-cover composite gave a significantly higher SCF (more than 10%)
342 than the one we show here using daily products, especially, in winter and early spring.
343 Interestingly, based on a different source of observation, Qin et al. (2006) found that snow covers

344 about 59% of the Tibetan Plateau in winter, which is comparable to the mean SCF (50%) in our
345 CAM5 simulation. Nonetheless, we keep this discrepancy in mind when interpreting the
346 wintertime BC-in-snow radiative forcing that suffers the most from such potential SCF bias.

347

348 **4 Modeled transport pathways and source attribution of BC in the HTP**

349 **4.1 Transport pathways**

350 The direct source tagging method can clearly characterize the three-dimensional transport
351 pathways of BC emitted from various source regions and sectors to the HTP receptor region.
352 General circulation patterns over the HTP and surroundings are typically affected by mid-latitude
353 westerlies in the winter and Asian monsoon in the summer, including the South Asian summer
354 monsoon and East Asian summer monsoon (Xu et al., 2009; Yao et al., 2012; Wu et al., 2012;
355 also see Fig. S2).

356 Figure 4 illustrates circulation patterns over HTP and BC transport pathways from six
357 major source regions to the HTP in the winter (DJF) and summer (JJA). (See similar plots in
358 Figs. S3–S5 for other tagged source regions.) In the winter, the strong surface cooling over the
359 HTP leads to subsidence/divergence and the formation of an enhanced local circulation cell,
360 while in the summer air converges toward the HTP from the surroundings, particularly from the
361 South Asia, due to the ascending of strongly heated air over the HTP (e.g., Wu et al., 2012), as
362 also indicated by the arrows in the vertical cross-sections in Fig. 4. In the winter, the subtropical
363 westerlies extend to about 10°N in mid-/upper troposphere and 20°N near surface, and the
364 tropical easterlies are weak (see the white contours of latitude-height cross-section panels in Fig.
365 4). The circulation patterns near the HTP change dramatically during the summer monsoon

366 season. The reversal of surface wind regime in the tropics (e.g., Arabian Sea, Bay of Bengal, and
367 South China Sea) is characteristic of the Asian summer monsoon climate (see Fig. S2e, g). The
368 subtropical westerlies recede to north of 30°N and the center of the westerly jet shifts to about
369 40°N in JJA (from about 30°N in DJF). The strong easterlies characterize the upper troposphere
370 of tropical region (south of HTP), while the southwesterly flow prevails in the lower troposphere
371 (white contours of latitude-height cross-section panels in Fig. 4). The prevailing winds during the
372 transition seasons (MAM and SON) between DJF and JJA are still westerlies (Fig. S2b, d).

373 The circulation patterns determine the transport of BC around the HTP region. However,
374 the variations of spatial distributions of BC emitted from the different source regions and in
375 different seasons could be due to the differences in source location and strength, wet removal
376 rate and lifting. Note that although we combined BC emitted from BB and FF sections to
377 characterize transport pathways in Fig. 4, only BC emissions from BB sector have seasonal
378 variations in the emission inventory we use.

379 The HTP region is surrounded by two major BC source regions, SAS and EAS (Fig. 1a),
380 which potentially have great impact on BC in the HTP (e.g., Menon et al., 2010; Bond et al.,
381 2007; Ohara et al., 2007; Xu et al., 2009; Kopacz et al., 2011; Lu et al., 2012). BC emissions
382 from SAS are dominated by the BB sector, and by FF sector from EAS (Fig. 1b). As shown in
383 Fig. 4, in the winter, a significant amount of BC from SAS can be transported to the eastern
384 Plateau by the strong westerlies under the dry winter monsoon conditions. During the South
385 Asian summer monsoon BC from SAS is effectively removed by the local abundant
386 precipitation, as indicated by the low lifetime in summer (Table S2), but can still affect large area
387 in the southwest of the HTP. However, BC from EAS can be uplifted higher and transported
388 more to the Northeast Plateau in the summer monsoon season than in the winter. Along the

389 wintertime westerlies, BC from upwind source regions (e.g., EUR, NAF, SAF, MDE, and CAS;
390 see Fig. S3) can easily move to the HTP, while the HTP local emissions are transported far away
391 (Fig. 4). BC originating from the distant sources such as SAF and MDE reaches up high (to
392 300hPa) in the HTP. In the summer, continental deep convection can loft BC into higher
393 altitudes where it can be transported to the HTP along the relatively weaker westerlies from
394 upwind source regions (e.g., EUR, RBU, MDE, and CAS; see Figs. 4 and S3). However, BC
395 from distant low-latitude source regions such as SAF barely reaches the HTP region due to weak
396 emissions but strong removal along the transport pathways to the HTP during the summer
397 monsoon season.

398 **4.2 Seasonal variation of BC in the HTP**

399 BC concentrations in the HTP have strong dependence on season and location. Figure 5 shows
400 the annual mean and seasonal variations of BC column burden and deposition rate over the HTP
401 and five sub-regions. The seasonal variation of the ratio of wet to total BC deposition is
402 superimposed. The Central Plateau is the cleanest region during all seasons, compared to other
403 sub-regions in the HTP (Fig. 5e). Both BC column burden and deposition rate from the BB
404 sector peak in MAM over the HTP, mostly in the Himalayas and Southeast Plateau region. The
405 FF BC burden in the HTP peaks in the summer mainly due to the seasonal maximum over
406 Northwest and Northeast Plateaus. However, BC wet removal rate over the Northwest Plateau is
407 at minimum in the summer, as opposed to the summer maximum in other sub-regions and the
408 entire HTP region. For Himalayas, Southeast and Central Plateau, the seasonal variation (i.e.,
409 maximum in MAM followed by a sharp decrease to JJA) of BB and FF column burden (Fig. 5c,
410 d and e) is similar to the variation of observed surface concentrations at sites located in these
411 sub-regions (Fig. 2b, d, e and f). In the Himalayas and Southeast Plateau, the ratio of regional

412 mean BC column burden to deposition rate (Fig. 5c and d), indicator of removal time scale or
413 lifetime, is the smallest (less than 1 day) during the Asian summer monsoon (JJA) due to the
414 efficient wet scavenging of BC by abundant precipitation. In Northwest and Northeast Plateau,
415 the BC column burden increases from DJF to JJA and reaches the maximum in JJA, and then
416 decreases in SON (Fig. 5b and f), partly due to the peak contribution of EAS and CAS emissions
417 in JJA. This trend is also similar to that in the observed surface concentrations (Fig. 2a and g).
418 The deposition rate follows the same seasonal variation of column burden over the Northeast
419 Plateau, while the deposition has a minimum in JJA over the Northwest Plateau when the column
420 burden is at maximum likely due to the less efficient wet removal in this region (Fig. 5b and f).

421 The annual mean BC column burden over the HTP has almost the same contributions
422 from BB and FF emission origins, with BB dominating in DJF and MAM and FF in JJA. In the
423 Himalayas, BC is predominantly from BB sector for all seasons (Fig. 5c). In the Southeast and
424 Central Plateaus, the dominant source sector is BB in DJF and MAM, but FF dominates in JJA.
425 The dominant source sector over the Northwest and Northeast Plateaus is always FF, especially
426 in the summer. We need to analyze the source-receptor relationships in order to quantify the
427 roles of BB and FF emissions from the various source regions in determining BC over the HTP
428 and the sub-regions.

429 **4.3 BC source-receptor relationships**

430 Previous studies (e.g., Xu et al., 2009; Kopacz et al., 2011) have shown that BC and its source-
431 receptor relationships vary significantly with season and location in the HTP. We intend to
432 quantify source contributions to BC at different locations of the HTP and in different seasons.
433 Our analysis also shows that the relative contributions to BC from different source regions and

434 sectors depend on season and location in the HTP. As shown in Fig. 6, the largest contribution to
435 the annual mean BC burden and surface deposition for the entire HTP region is from BB
436 emissions from SAS, followed by FF emissions from SAS and then the FF from EAS. The same
437 roles hold for all the seasonal means except for the summer (JJA) when the EAS FF becomes
438 more important for BC column burden in the HTP and, to a lesser extent, for deposition.

439 The SAS emissions account for 50% of the annual mean burden over the HTP, including
440 33% from BB and 17% from FF. The other 50% is mostly from the EAS (5% BB and 14% FF),
441 HTP (6% BB and 6% FF), CAS FF (4%), MDE FF (4%) and SAF BB (3%). The source
442 attribution for annual mean BC deposition for the entire HTP is similar, but SAS contributes
443 even more to BC deposition than to the column burden. Although RBU has a lower contribution
444 to the annual mean BC in the HTP than the six regions shown in Fig. 6, its contribution to the
445 JJA mean, especially at some locations, is quite substantial (included in the black bar) and even
446 more important than some of the six regions in Fig. 6, as discussed in detail below.

447 BC annual mean burden over the HTP has nearly equal contributions from BB and FF
448 emissions. However, contribution by BB emissions, mainly from SAS, is larger than from FF
449 sector in DJF and MAM. In the summer (JJA), the largest contribution (about 29%) to HTP BC
450 is from EAS FF emissions. This is partly due to the change of circulation patterns (Fig. 4) and
451 effective wet removal of SAS BB emissions. Note that EAS FF emissions are much larger than
452 FF emissions from any other of the source regions, and are more than twice the EAS BB
453 emissions.

454 For BC in the five finer receptor regions of interest (as defined in Fig. 5g), SAS BB and
455 FF have the largest contribution to BC in Himalayas and Central Plateau, while EAS FF and BB

456 contribute the most to Northeast Plateau in all seasons and Southeast Plateau in the summer.
457 Central Asia and Middle East FF emissions have relatively more important contribution to BC
458 reaching Northwest Plateau, especially in the summer.

459 For the Northwest Plateau (Fig. 6b), the prevailing winds in this sub-region are westerly
460 throughout the year (Fig. S2; Cao et al., 2009; Xu et al., 2009), so the important source regions
461 ought to locate at the west of HTP (e.g., MDE, EUR, and parts of SAS and RBU in Fig. 1a). SAS
462 emissions are still the dominant source for the annual BC burden in this sub-region (17% from
463 BB and 14% from FF), followed by HTP local emissions (9% from BB and 13% from FF), CAS
464 (2% from BB and 14% from FF), MDE FF (8%) and EAS FF (7%). BC emissions from SAS are
465 the dominant source in DJF, MAM and SON. CAS becomes the dominant source region (5%
466 from BB and 26% from FF) in JJA, even though CAS is not a significant emission source region
467 on a global basis (Fig. 1b). BC emitted from MDE is predominantly in the FF sector throughout
468 the year. Emissions from the rest of the tagged sources (in addition to the top six) become more
469 significant in this sub-region (black color in Fig. 6b), mostly from EUR and RBU through long-
470 range transport (Fig. S3). The source attribution for BC deposition in this sub-region is similar to
471 that of the column burden, but BC emitted from SAS and MDE appears to be more efficient in
472 deposition except for the JJA season when BC from EAS contributes more to deposition than to
473 column burden.

474 The Himalayas sub-region located along the southern edge of the HTP is in close
475 proximity to the SAS. Thus emissions from SAS are absolutely the dominant source for BC in
476 Himalayas throughout the year. This sub-region receives more BC from BB sector than FF
477 because BC emissions in SAS are mainly in the BB sector, especially in MAM season (Fig. 1b
478 and Fig. S6). For the annual mean burden, BC from SAS contributes 81% (54% from BB and

479 27% from FF), followed by HTP local emissions (6% from BB and 3% from FF). It is worth
480 noting that SAF BB emissions contribute about 10% to burden in DJF through a long-range
481 transport. BC deposition in this sub-region also predominantly originates from SAS, which is
482 consistent with previous studies by Ming et al. (2008) and Kopacz et al. (2011).

483 For the Southeast Plateau (Fig. 6d), the BC source contribution profile is similar to that of
484 Himalayas during DJF and MAM season, in which SAS is still the dominant source, especially
485 in MAM (74% contribution to column burden, including 53% from BB and 21% from FF),
486 although the contribution from EAS is larger here than for the Himalayas. As also pointed by
487 Ramanathan et al. (2007), BC over the SAS can be transported to the Southeast Plateau by the
488 southern branch of the westerlies during the winter and spring. However, the BC source
489 contribution profile changes dramatically during the summer when emissions in EAS become the
490 dominant source to this sub-region (68% to column burden, including 23% from BB and 45%
491 from FF). Kopacz et al. (2011) also found that the BC from south-eastern China is the dominant
492 contributor to the Southeast Plateau in July. For the annual mean burden in this sub-region, SAS
493 is still the dominant contributor (40% from BB and 17% from FF), followed by EAS (8% from
494 BB and 15% from FF), HTP (6% from BB and 6% from FF). BC originating from EAS
495 contributes more to deposition than to burden in this sub-region.

496 For the Central Plateau (Fig. 6e), source attribution profiles for annual and seasonal BC
497 are very similar to those of the entire HTP region with SAS being the dominant source region
498 throughout the year except that EAS has comparable contributions in JJA. Ming et al. (2010)
499 pointed out that pollutants from the Indo-Gangetic Basin could be transported to the Central
500 Plateau by both the summer monsoon and the westerlies. Xia et al. (2011) also found that the
501 substantial regional atmospheric brown haze from the nearby regions of SAS is the main source

502 for the background aerosols in the Central Plateau based on sunphotometer and satellite
503 observations.

504 Compared to the other sub-regions, the Northeast Plateau receives the largest contribution
505 of BC from EAS throughout the year (50% to annual mean burden, including 12% from BB and
506 38% from FF; see Fig. 6f), especially in JJA (17% from BB and 49% from FF). The EAS FF
507 sector contribution and the magnitude of burden (Fig. 5f) over the Northeast Plateau have strong
508 seasonal variations, mostly due to variations in meteorology because the FF emissions in our
509 simulation do not vary seasonally. Kopacz et al. (2011) indicate that the primary contribution to
510 BC over the Northeast Plateau is from western China during January and April (transported by
511 mid-tropospheric westerlies), and from central-eastern China during July and October
512 (transported by boundary layer flow). Other main contributions to BC burden over the Northeast
513 Plateau include 13% from SAS and 10% from HTP local emissions. Similar to the Northwest
514 Plateau, some other upwind source regions (e.g., CAS, MDE, RBU and EUR; see Figs. 4 and S3)
515 have a significant contribution to the Northeast sub-region as well.

516 **4.4 Seasonal variation of HTP BC sensitivity**

517 Following Wang et al. (2014), we defined the “efficiency” metric in Sect. 2.2 to quantify the
518 sensitivity of BC response to absolute change (e.g., per unit perturbation) of emissions in
519 different source regions. This metric has the value of 1 if the entire globe is treated as a single
520 source region, so we may assume the global mean efficiency of 1 as a reference to measure the
521 sensitivity to perturbation from different source regions/sectors.

522 Figure 7 shows efficiencies of tagged sources in affecting the BC seasonal and annual
523 mean column burden and deposition in the HTP and five sub-regions (as defined in Fig. 5g). BC

524 in the same receptor regions is generally most sensitive to change in local emissions, regardless
525 of seasons, emission sectors and locations of receptor regions. Among all the source regions,
526 although the HTP local (FF+BB) emissions only contribute about 10%, BC in the HTP is
527 extremely sensitive to changes in the emissions within HTP (not shown in the figure), mainly
528 because the emission rate is very low. In addition to the local emissions, BC in the HTP is also
529 sensitive to emissions in neighboring source regions (e.g., SAS and CAS) and emissions from
530 distant sources such as MDE. Not only does the SAS have large contribution to BC burden and
531 deposition over the HTP, as well as most of the sub-regions except for the Northeast Plateau
532 (receptor V), but also the efficiencies for SAS emissions are high for almost all of the sub-
533 regions especially the Himalayas (receptor II). BC in the Northeast Plateau (receptor V) is quite
534 sensitive to EAS emissions in all seasons, while BC in the Southeast Plateau is sensitive to EAS
535 emissions in JJA and SON. Although BC emissions from MDE and CAS are weak (Fig. 1b) and
536 their contributions to the HTP are relatively low, their efficiencies are high. BC over Northwest
537 Plateau (receptor I) and Central Plateau (receptor IV) is extremely sensitive to emissions from
538 CAS in JJA. These source-receptor relationships of sensitivity will provide useful information
539 for policymakers to improve the effective mitigation road map in order to potentially slow down
540 the glacier retreat in the HTP region.

541

542 **5 Radiative forcing**

543 The BC-in-snow effect can be quantified using the online calculation of radiative forcing in the
544 SNICAR (Snow, Ice, and Aerosol Radiative) model (Flanner et al., 2007) coupled to CAM5, and
545 then compared to airborne BC radiative forcing. Figure 8 shows seasonal and annual mean BC

546 all-sky shortwave direct radiative forcing (DRF) at the surface (dimming) and the top of the
547 atmosphere (TOA), and the BC-in-snow radiative forcing (darkening) averaged over the entire
548 HTP and the five sub-regions (as defined in Fig. 5g). Note that the BC-in-snow forcing is
549 averaged over all model grids in the area (i.e., zero enters the calculation for any grid when snow
550 is not present). The radiative forcing of BC (and dust) in snow is small in JJA and SON due to a
551 lack of snow cover (Fig. 3). The forcing maximum occurs during the spring melt (MAM) when
552 the insolation is rather intense and BC accumulates at the surface of the snowpack as the snow
553 melts (Conway et al., 1996; Flanner et al., 2007, 2009), and when the snow-albedo feedback is
554 strongest (Hall and Qu, 2006). This strong seasonal variation also explains why the coefficient of
555 variation (i.e., the ratio of the SD to the mean) is greater than 1 for the annual mean BC-in-snow
556 forcing over the entire HTP and all sub-regions. The seasonal variations of airborne BC DRF at
557 the TOA and surface are consistent with that of the BC column burden (Fig. 5). For the entire
558 HTP (Fig. 8a1), the annual mean surface radiative forcing due to BC in snow (0.42 W m^{-2})
559 exceeds the BC dimming effect at the surface (-0.3 W m^{-2}). The annual mean BC-in-snow
560 forcing is even higher over the Northwest Plateau (Fig. 8b1) and Himalayas (Fig. 8c1), and far
561 exceeds the other forcings in the same sub-regions, although the BC-in-snow effect may be
562 overestimated due to the potential positive bias in snow cover fraction in our simulation (Fig. 3).
563 The annual mean BC surface dimming exceeds the BC-in-snow effect in the Southeast (Fig. 8d1),
564 Central (Fig. 8e1) and Northeast Plateau (Fig. 8f1). The minima of all BC-related forcings
565 appear in the Central Plateau where the BC burden and deposition are the lowest among all the
566 sub-regions (Fig. 5), and the SCF is very small (Fig. 3).

567 We have also calculated an approximate source attribution for the BC-in-snow radiative
568 forcing over the HTP and its sub-regions, using the tagged-source BC deposition, which is

569 simply assumed to be linearly proportional to BC-in-snow radiative forcing. The SCF is taken
570 into account in the calculation (i.e., the deposition at each model grid is multiplied by SCF when
571 calculating the area-average deposition). Overall, despite small quantitative differences, the
572 source contributions to BC-in-snow forcing are similar to those for BC deposition (Fig. 6). The
573 SAS BB emissions contribute the most to annual mean forcing over the HTP and sub-regions
574 except for the Northeast Plateau that is mostly contributed by EAS FF emissions. During the
575 winter and spring seasons over Northwest Plateau and Himalayas, when and where the forcing is
576 the largest, SAS (especially the BB sector) is the major contributor.

577 Dust is a major contributor to the total aerosol burden over the HTP (e.g., Zhang et al.,
578 2001). Although we don't focus on other snow impurities such as mineral dust, it is worth noting
579 that dust-in-snow radiative forcing has been considered in our model simulation and it could be
580 an important forcing agent. We also plotted dust-in-snow forcing over the HTP and sub-regions
581 in Fig. 8 (along with the BC-induced forcings). The annual mean dust-in-snow forcing (0.33 W
582 m^{-2}) is comparable to all of the other forcings over the HTP, especially in the springtime when
583 dust outbreaks and can be transported to the HTP from the surrounding sources such as
584 Taklimakan and Gobi deserts (Liu et al., 2008). The annual mean dust-in-snow forcing is as large
585 as 0.99 and 0.59 W m^{-2} in the Northwest and Northeast Plateau, respectively (Fig. 8b1 and f1),
586 which is in close proximity to the Taklimakan Desert (Huang et al., 2007; Chen et al., 2013), but
587 negligibly small in the Southeast Plateau and Central Plateau. In the winter, the dominant dust in
588 snow effect over Northeast Plateau is consistent with the recent observations. Huang et al. (2011),
589 X. Wang et al. (2013) and Zhang et al. (2013) found that insoluble light-absorbing particles in
590 snow are dominated by local soil and desert dust in the Qilian Mountain (Northeast Plateau).

591 Both snow cover fraction (SCF) and mass concentration of snow impurities affect the
592 calculation of radiative forcing in snow. We have evaluated the model estimation of SCF in
593 different seasons (Fig. 3). We have also compared BC concentration and deposition flux from
594 our model results to a recent modeling study by Ménégóz et al. (2014) and to observations in the
595 HTP (Ginot et al., 2014) (Table S3). Bond et al. (2013) pointed out that observations of BC in
596 snow pits or ice cores mostly involve snow/ice samples obtained in the summer and early fall,
597 when almost all grid boxes the sample sites located in are snow free in the HTP. They also
598 indicated that the CAM3 global climate model (Flanner et al., 2009) may be overestimating snow
599 BC concentrations in the HTP, especially in the spring. Our comparison shows that despite a
600 smaller bias than in Ménégóz et al. (2014) the CAM5 model still largely overestimates BC
601 concentrations in snow but underestimates dust concentrations in snow over the HTP. Ménégóz
602 et al. (2014) provided a few possible reasons for the differences between model simulations and
603 observations. Factors such as measurement uncertainties (due to sample treatment and analysis
604 methodology), temporal (inter-annual and seasonal) and spatial variations of BC deposition, and
605 vertical variations of BC in snowpack, can strongly affect the accuracy and representativeness of
606 BC-in-snow measurements for the purpose of evaluating global models. Ming et al. (2013) and
607 Qian et al. (2015) pointed out that BC concentrations in snow and ice samples over HTP tend to
608 decrease with increasing glacier elevations, while global models with coarse grid resolution
609 cannot accurately represent elevation of sampling sites. Often times the difference is significant.
610 Nonetheless, it is likely that positive biases exist in the modeled concentration and radiative
611 forcing of BC and dust in snow.

612 **6 Summary and conclusions**

613 In this study, we employed the CAM5 model with a newly developed source tagging technique,
614 nudged towards the MERRA meteorological reanalysis, to characterize the fate of BC particles
615 emitted from various geographical regions and sectors to the HTP region. In addition, we
616 compare the radiative forcing induced by BC in the atmosphere and in snow over the HTP, as
617 well as forcing induced by dust in snow. Although there are biases in the simulated BC, partly
618 due to the inherent difficulty for coarse-resolution global models to accurately represent transport
619 and wet deposition in this topographically complex region, the CAM5 model simulation shows a
620 reasonable agreement in the seasonal variation of the near-surface airborne BC concentrations
621 with observations over the HTP and surrounding regions. This provides us the confidence to use
622 this modeling framework to characterize BC source-receptor relationships in the HTP. Using
623 very different approaches, Kopacz et al. (2011), Lu et al. (2012) and the present study all show
624 that South Asia and East Asia are the main source regions for BC transported to the HTP, while
625 the magnitude of contributions from each of the source regions varies with season and receptor
626 location. Although all of the three studies can provide quantitative source attributions, a
627 quantitative inter-comparison of the findings is quite difficult, given the differences in the
628 definition of geographical source and receptor regions, emission inventories, time periods for
629 model simulation, and analysis methods. Nevertheless, in addition to quantifying the
630 contributions of source regions, our direct source tagging approach allows us to further break
631 down regional contributions by sectors (i.e., fossil fuel vs. biomass & biofuel) and to characterize
632 the transport pathways of individual regional/sectoral emissions.

633 The explicit source tagging technique enables the characterization of three-dimensional
634 transport pathways of BC to the HTP from different geographical regions and source sectors,
635 which also depends on seasons and the location of the receptor in the HTP. With the IPCC AR5

636 present-day emission inventories, the annual mean BC column burden and surface deposition in
637 the entire HTP region is contributed the most by biomass and biofuel (BB) emissions from South
638 Asia (SAS) (33% and 40%, respectively), followed by fossil fuel (FF) emissions from SAS (17%
639 and 20%, respectively), and then the FF from East Asia (EAS) (14% and 14%, respectively). The
640 same roles hold for all the seasonal means except for the summer when the EAS FF becomes
641 more important. Although BC emissions from the entire EAS source region are much stronger
642 than those from SAS, the concentrated FF BC emissions in central-eastern China are only
643 transported towards the HTP during the East Asian summer monsoon. Thus seasonal prevailing
644 winds are important in determining the seasonal variations in BC transport and source-receptor
645 relationships.

646 Both the annual and seasonal mean BC properties and their source-receptor relationships
647 vary significantly with location in the HTP. For the multiple finer receptor regions of interest,
648 SAS BB and FF have the largest impact on BC in Himalayas and Central Plateau, while EAS FF
649 and BB contribute the most to Northeast Plateau in all seasons and Southeast Plateau in the
650 summer. The Central Asia (CAS) and Middle East (MDE) FF emissions make important
651 contributions to BC over the Northwest Plateau, especially from CAS in JJA.

652 The HTP BC is most sensitive by far to per unit changes in the local emissions, although
653 they only contribute about 10% to the BC burden in the HTP. The SAS region makes large
654 contributions to BC burden and deposition over the HTP and the BC sensitivities to SAS
655 emissions are also high for almost all of the sub-regions of HTP, especially the Himalayas. BC
656 over the Northeast Plateau is quite sensitive to EAS emissions in all seasons, and Southeast
657 Plateau BC is also sensitive to EAS emissions in JJA. Although BC emissions from MDE and
658 CAS are weak and their contribution to the HTP overall is low, their efficiencies are quite high.

659 BC over Northwest Plateau and Central Plateau is extremely sensitive to emissions from CAS in
660 JJA. These source-receptor relationships and sensitivities can be useful to policymakers for
661 improving the effective mitigation road map in order to potentially slow down the glacier retreat
662 in the HTP region.

663 The impact of BC on snow and glacier melting can be characterized by the magnitude of
664 radiative forcing. Our calculations show that the annual mean BC-in-snow radiative forcing (0.42
665 W m^{-2}) outweighs BC dimming effect (-0.3 W m^{-2}) at the surface over the HTP. In the five sub-
666 regions, the annual mean BC-in-snow forcing ranges from 0.04 W m^{-2} in the Central Plateau to
667 1.75 W m^{-2} in the Northwest Plateau. We also showed that the annual mean dust-in-snow
668 induced radiative forcing over the HTP can be quite significant (0.33 W m^{-2} for entire the HTP,
669 and 0.99 W m^{-2} for the Northwest Plateau). More importantly, both BC- and dust-in-snow
670 forcing peaks in the spring melting season when the area-average forcing reaches 1.03 and 0.87
671 W m^{-2} , respectively, over the entire HTP, and their combined forcing is more than 8 W m^{-2} over
672 the Northwest Plateau. Such a large forcing is sufficient to cause earlier snow melting and
673 contribute to the acceleration of glacier retreat, although the model is likely to overestimate BC-
674 in-snow forcing due to the possible positive bias of snow cover fraction in the winter and early
675 spring. According to our estimates of the source attribution, the biomass burning and biofuel
676 emissions in South Asia contribute the most to annual mean forcing over the HTP and its sub-
677 regions except for the Northeast Plateau where the largest contribution is from East Asia fossil
678 fuel emissions. During the winter and spring seasons over Northwest Plateau and Himalayas,
679 when and where the forcing is the largest, South Asia (especially the biomass burning and
680 biofuel sector) is the major contributor.

681

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692

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Table 1. List of sites for the observations of atmospheric BC surface concentrations used in this study to evaluate our model simulation.

Site	Latitude (°N)	Longitude (°E)	Elevation (m)		Sampling time	Observation method	Contributor
			observation	model			
Muztagh Ata	38.3	75.0	4500	3497	2003–2006	Thermal Optical Reflectance (TOR)	Cao et al., 2009
Hanle	32.8	79.0	4250	4862	2009–2010	Aethalometer	Babu et al., 2011
Manora Peak	29.4	79.5	1950	1409	2005–2008	Thermal Optical Transmittance (TOT)	Ram et al., 2010
NCO-P	28.0	86.8	5079	4604	2006–2008	Multi-angle absorption photometer (MAAP)	Marinoni et al., 2010
Lulang	29.5	94.4	3300	3370	2008–2009	TOR	Zhao et al., 2013
NCOS	30.8	91.0	4730	4956	2006–2007	TOR	Ming et al., 2010
QSSGEE	39.5	96.5	4214	2748	2009–2011	Aethalometer	Zhao et al., 2012

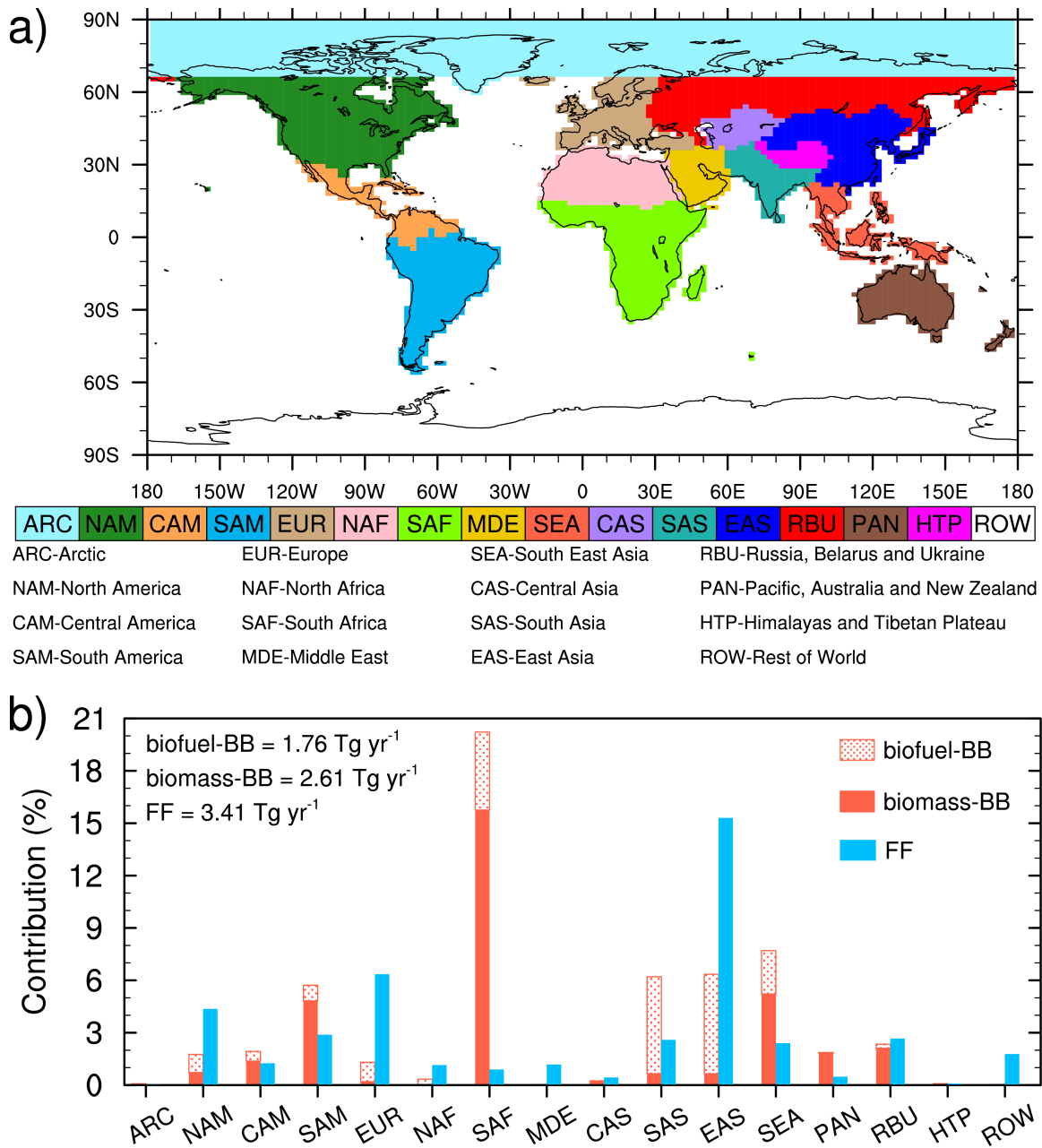


Fig. 1. (a) Tagged source regions and (b) the respective percentage contributions to global annual mean BC emissions from the individual source regions and sectors (including biofuel, biomass burning and fossil fuel). The global annual mean BC emission rate is 7.78 Tg yr^{-1} , which is divided up into the three sectors as indicated by the numbers at the upper-left corner.

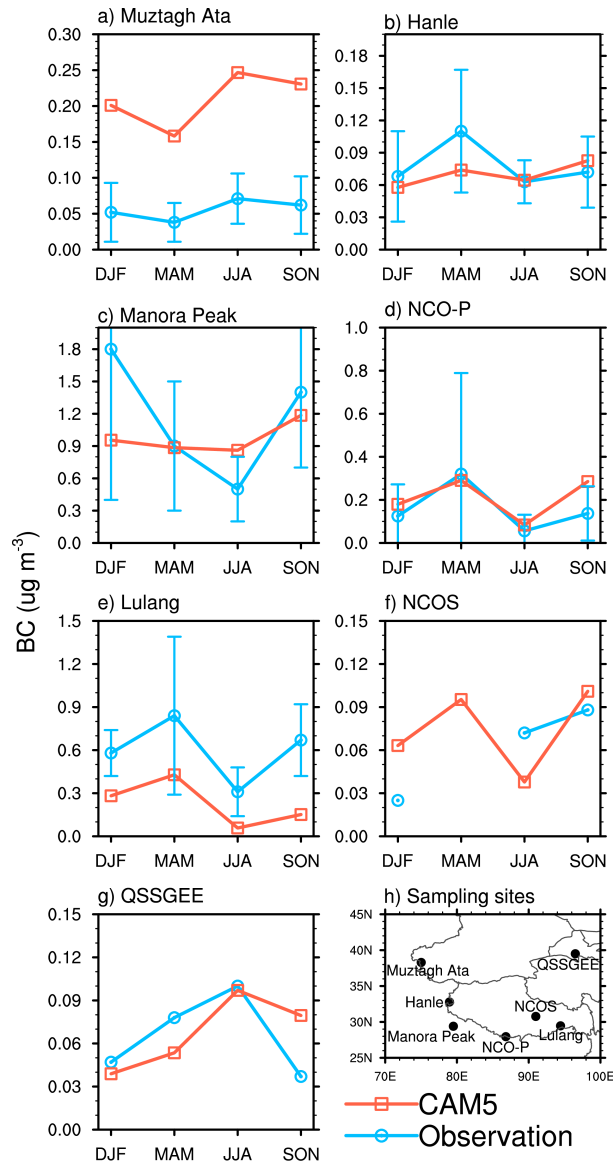


Fig. 2. Seasonal mean surface BC concentration ($\mu\text{g m}^{-3}$) from observations (blue lines with error bars denoting SD) and CAM5 simulation (red lines) at the seven sampling sites listed in Table 1 and marked in map of panel h.

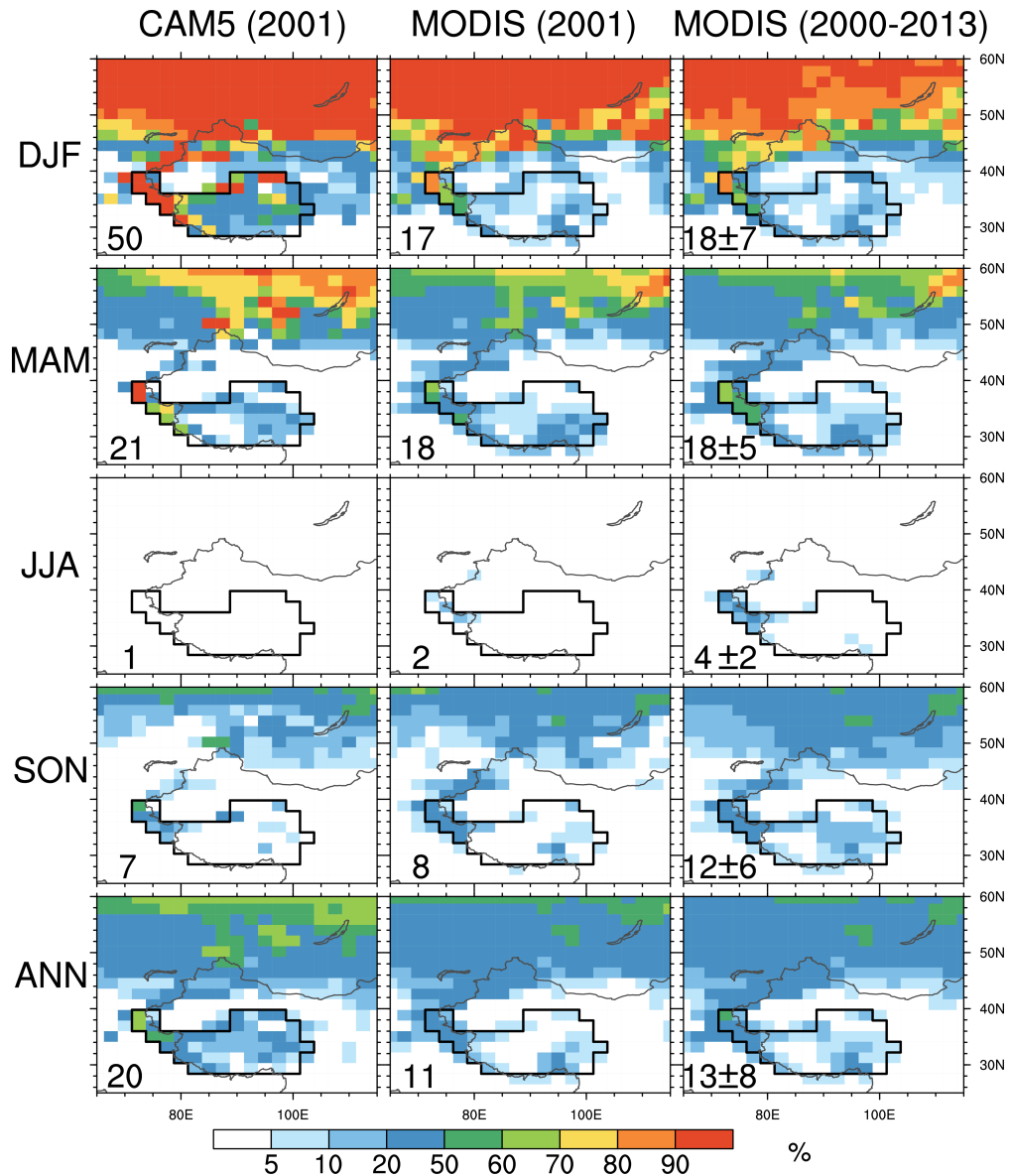


Fig. 3. Seasonal and annual mean snow cover fraction from CAM5 simulation for year 2001 (left), and MODIS retrieval for 2001 (middle) and 2000-2013 (right). The summer (JJA) in 2001 for both CAM5 and MODIS only includes July and August due to missing MODIS data in June. The number in the lower-left corner of each panel is the corresponding spatial mean SCF for the HTP region (which is marked with black outline), and for MODIS the standard deviation calculated from the MODIS multi-year means is also included.

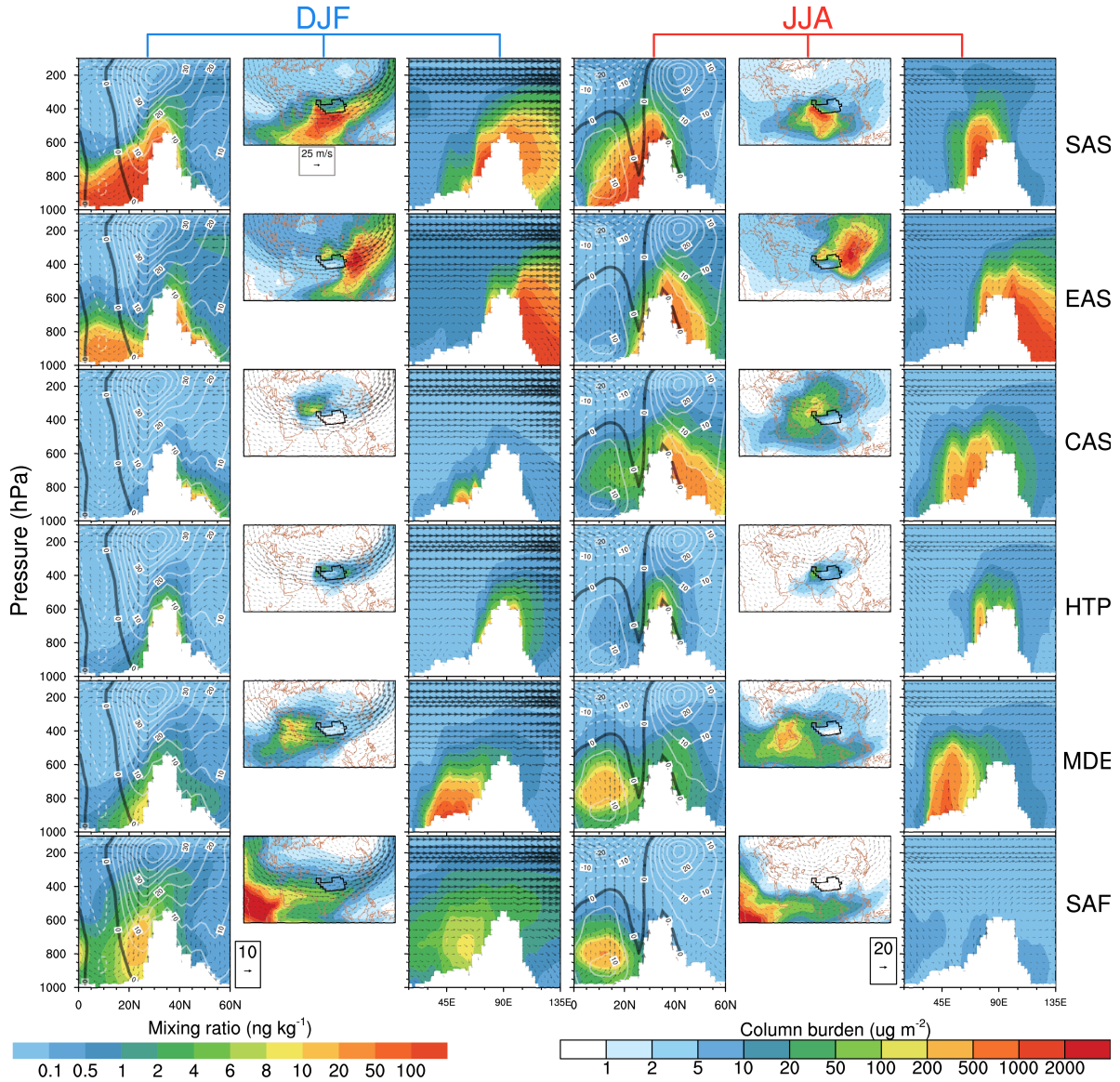


Fig. 4. The first column shows the latitude-height distributions of DJF BC mass mixing ratios (in ng kg^{-1} , colors) averaged over $71.25\text{--}101.25^\circ\text{E}$, originating from BB and FF sectors in the tagged source regions (corresponding to different rows); the white shaded area denotes topography, and the superimposed white contours at intervals of 5 m s^{-1} represent the westerly (solid) and easterly (dashed) DJF mean zonal winds along the cross-section with the thick solid black contour at 0 m s^{-1} ; the wind vectors (consisting of vertical velocity in units of $-10^{-4} \text{ hPa s}^{-1}$ and meridional wind in m s^{-1}) are represented by arrows. Colors in the second column denote spatial distribution of the DJF mean BC column burden (in $\mu\text{g m}^{-2}$), originating from different source regions, and the arrows represent the DJF mean horizontal wind vectors at 500hPa; the HTP is marked with black outline. The third column is similar to the first column except that the quantities are on the longitude-height cross-section averaged over $28\text{--}40^\circ\text{N}$, and thus the horizontal component of the wind vectors is zonal wind (m s^{-1}) instead. The fourth to sixth columns are the same as the first to third columns, respectively, but for JJA means instead.

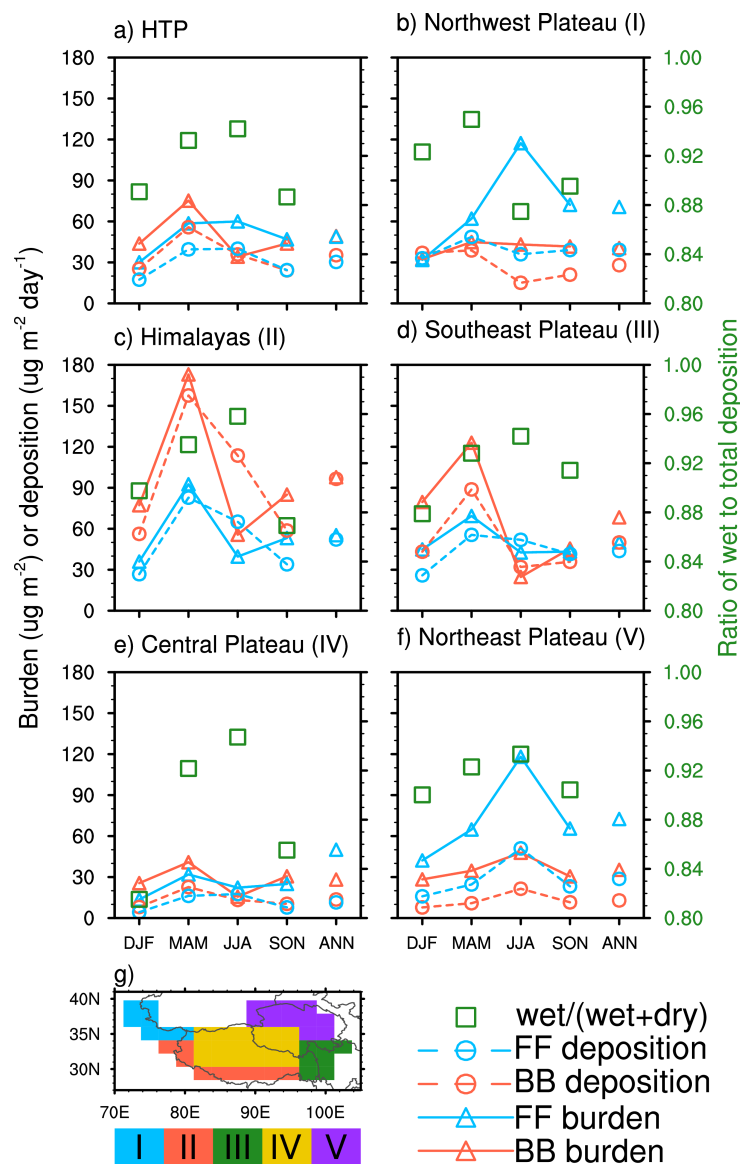


Fig. 5. Seasonal and annual mean BC column burden (solid lines and open triangles, in $\mu\text{g m}^{-2}$) and deposition rate (dashed lines and open circles, in $\mu\text{g m}^{-2} \text{ day}^{-1}$) over (a) the HTP, (b) Northwest Plateau, (c) Himalayas, (d) Southeast Plateau, (e) Central Plateau and (f) Northeast Plateau, emitted from BB (red color) and FF (blue color) source sectors. The green squares denote the ratio of wet to total BC deposition (using y-axis on the right) in four seasons over each receptor region. The geographical locations of five sub-regions of HTP are indicated in panel g.

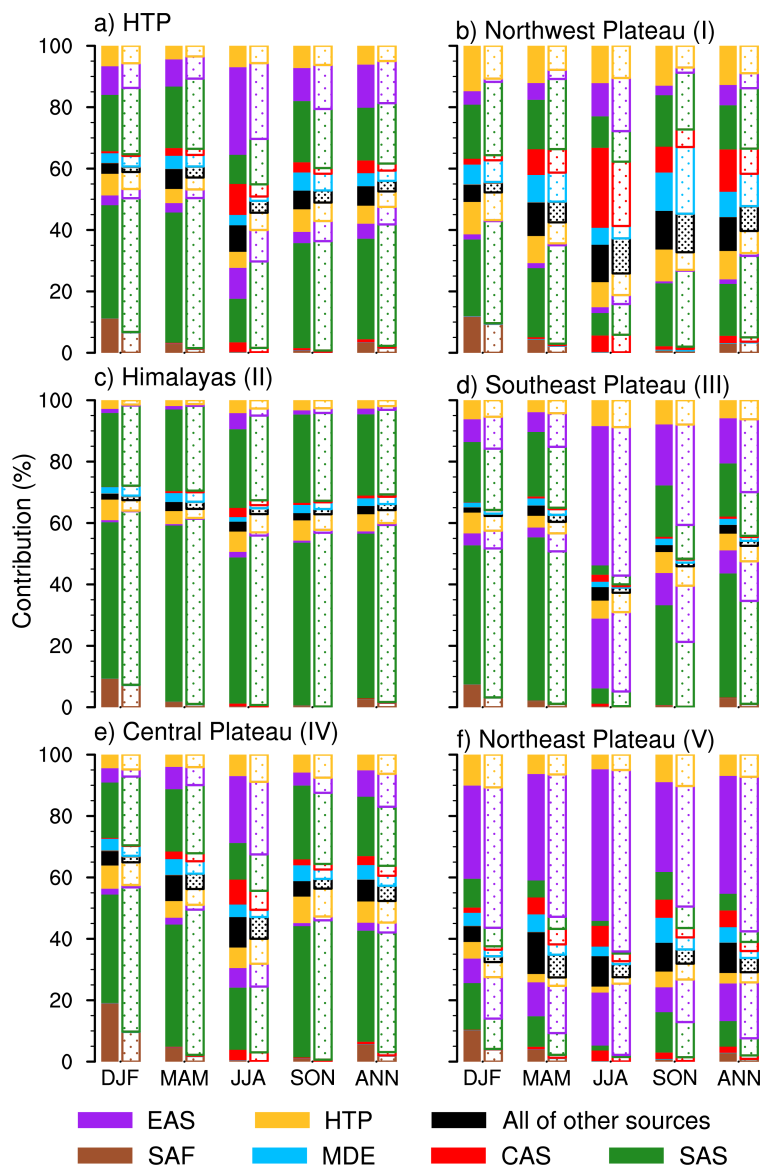


Fig. 6. Fractional contributions (measured by the lengths of color bars) to seasonal and annual mean BC column burden (solid pattern bars) and deposition (dotted pattern bars) over (a) the HTP, (b) Northwest Plateau, (c) Himalayas, (d) Southeast Plateau, (e) Central Plateau, and (f) Northeast Plateau, originating from six major tagged source regions (indicated by colors at the bottom) for BB (colors below the black bar) and FF (colors above the black color) emissions. The black bar in each column represents the contribution from all of the other tagged source regions and sectors.

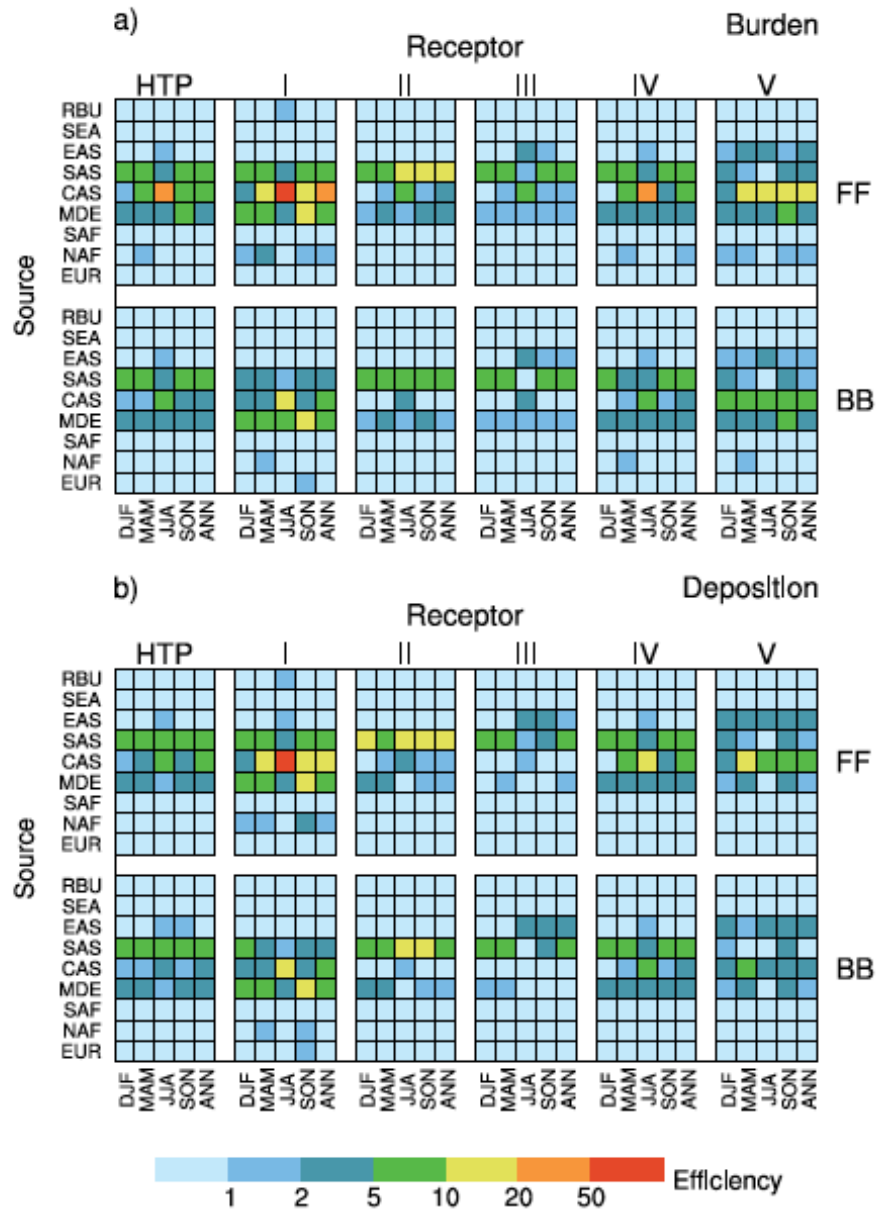


Fig. 7. Efficiency of FF (top) and BB (bottom) emissions from ten source regions (on the y-axis) in changing seasonal and annual mean (a) BC column burden and (b) deposition over the HTP and each of the five sub-regions: Northwest Plateau (I), Himalayas (II), Southeast Plateau (III), Central Plateau (IV) and Northeast Plateau (V).

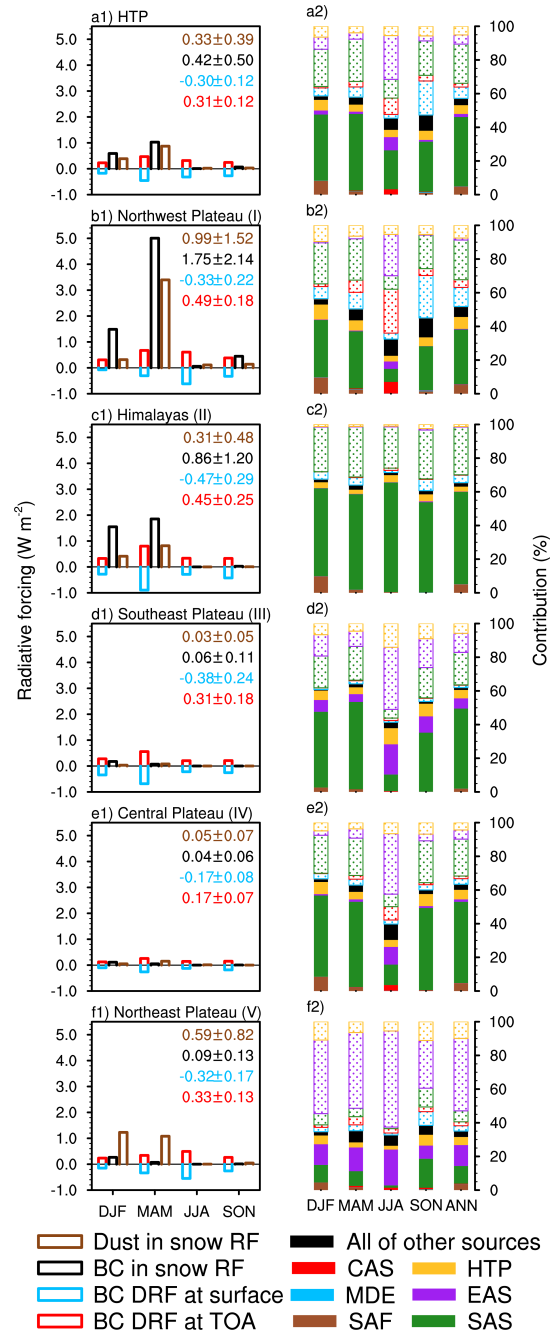


Fig. 8. Seasonal mean radiative forcing (left column) induced by the various BC effects (indicated by the color legend at the bottom) and dust-in-snow effect over (a1) the HTP, (b1) Northwest Plateau, (c1) Himalayas, (d1) Southeast Plateau, (e1) Central Plateau and (f1) Northeast Plateau. The corresponding annual mean forcings and one SD (for 12 monthly means) are shown in numbers on the top-right corner of each panel. The right column (a2-f2) panels represent source contribution to surface BC-in-snow radiative forcing over the corresponding receptors from tagged source regions (colors) and sectors (solid pattern bar and dotted pattern bar for BB and FF, respectively). The black bar in each column represents the contribution from all of the other tagged source regions and sectors.