Editor Decision: Reconsider after minor revisions (Editor review) (05 May 2015) by Xiaobin Xu

Comments to the Author:

Dear authors,

Please study carefully the referee report #2, address the remaining issues and make technical changes as suggested.

Sincerely,
Xiaobin Xu

Dear Editor,

We have addressed the minor issues raised by the referee, but do not agree with the two main suggestions for the reasons detailed below. Regarding the major concern of the referee on our efficiency metric, we want to stress that the metric (which weights source region contributions by their own emissions) and the associated results (Fig. 7) and discussion are neither flawed nor vaguely presented. As we now explain in the manuscript, the efficiency metric is proportional to the sensitivity coefficient (ΔBC_concentration/Δemissions, or ΔBC_deposition/Δemissions), which has been used in other source-receptor studies (e.g., Shindell et al., 2008; Wang et al., 2014; references are provided in the revised manuscript.). Also, it complements the relative contribution metric used in the paper. However, as a compromise, we have removed the local emissions (HTP) efficiency from the figure, as it could potentially cause misinterpretation of our main results.

Thanks,

Hailong Wang (on behalf of all authors)

Referee Report #2

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

Authors: Rudong Zhang, Hailong Wang, Yun Qian, Philip J. Rasch, Richard C. Easter, Po-Lun Ma, Balwinder Singh, Jianping Huang, and Qiang Fu

General Comment

Thanks for the detailed responses and revisions in reply to both my comments and to those from the other referee. However, after reviewing the author response to my

comment about efficacy, I strongly recommend the discussion of efficacy be removed entirely before the manuscript is published. The authors themselves state (line 251): "This metric [efficiency] is of more interest to policy makers for the purpose of mitigation action, which is not the focus of this study but is worth mentioning." This single phrase should be enough to suggest the discussion is not contributing to the basic findings of the study, but I provide a much more extensive critique below.

Specific Comment

Line 94-5: change "slow down the climate change" to "slow down present-day climate change"

Done as suggested.

Section 4.1 and 4.3: I agree that including all the source region graphics from figure S3 in the main text would be unnecessary, but please add cross-referencing to figure S3 throughout these two sections since figures 4 and 6 do not have the information about other regions (like RBU) you are often alluding to. Figure S3 seems really useful to repeatedly bring to the attention of the reader.

Done as suggested.

Figure 6: please make this figure just like was done for figure 7! Figure 6 is not nearly as clear to me as figure 7. A category for "all other source regions" (black in figure 6) could still be included, and the relative contributions of FF and BB could be easily be noted in text within the figure. This would make the message much clearer and draw out the results from figure 6.

We disagree with this suggestion. We would argue that the bar chart (with color-coded elements) is more effective and straightforward than using the color scheme alone (like in Figure 7) for comparing the magnitudes between different contributors/sources and showing seasonal variations of relative contributions by individual sources. It also facilitates the comparison of contributions between burden and deposition by placing them side-by-side. Moreover, the colors make it easy to compare contributions of individual regions to the different receptors across the various panels.

Section 4.4 and Efficiency: Thanks for the explanation, but I still do not think this section or figure 7 are relevant to the paper. It comes down to a metric that can be 1. Easily misinterpreted, and 2. Misinterprets the results. As pointed out by the authors in the response to my original comment, efficiency is a relative metric suggesting the importance of relative changes to emissions from source regions to HTP and sub-regions. Figure 7 shows that HTP is the most important contributor to its own BC. This is not surprising given essentially common knowledge about aerosol lifetimes. The real message is in figure 6 – what regions determine absolute contributions to BC burden and deposition over HTP.

Figure 7 waters down the enormous impact (burden and deposition) of SAS and EAS on HTP and, to varying degrees, on HTP sub-regions, as shown in figure 6. This watering down is evident in the author response: "In other words, the efficiency of local emissions in affecting HTP BC is very high (Figure 8), which means that the impact of per-unit-mass (or equal-magnitude) perturbation in emissions on BC over HTP is much stronger if the perturbation occurs within HTP than in any other source regions including SAS and EAS." The key words are that the authors are viewing the impact of a perturbation in emissions within HTP. My original comment was whether that was relevant to HTP when I said "HTP has practically no emissions and I do not see how anyone could practically expect HTP to develop major emission sources." In other words, what could happen in HTP that would perturb the emissions in any way? Are power plants expected to be built there? Are fires expected to increase? Population?

Look at my point another way using your own numbers, roughly shown in the table below. Numbers I label "Efficiency" are values under "Figure 6a (approx)" divided by "Ei/Etotal (fraction)" under "Combine Fig 1 and S1". The conclusion that could be read is that emissions controls/mitigation in SAS and EAS is not as needed as making sure HTP keeps emissions low since HTP has a higher efficiency (191 vs 19 for SAS and 2.1 for EAS). The higher efficiency is a result of dividing a comparable contribution (0.11 for HTP, vs 0.17 for EAS, vs 0.49 for SAS, estimated from figure 6a) by a very small number – HTP total emissions are 143x less than EAS and 45x less than SAS. Altogether this obfuscates the real problem: absolute contributions from SAS and EAS. If SAS and EAS stopped emitted BC, HTP burden would drop precipitously.

I strongly recommend dropping the discussion of efficiency in this manuscript, and dropping the equation and discussion for efficiency in Section 2.2. This is better left to a dedicated study that talks more deeply about what this actually means for policy makers.

Figure 1 (direct)

BM-BB (Tg/yr)	BF-BB (Tg/yr)	FF (Tg/yr)	total (Tg/yr)
2.61	1.76	3.41	7.78

Table S1 (direct)

	BM-BB (%)	BF-BB (%)	FF (%)
SAS	0.65	5.56	2.58
EAS	0.64	5.7	15.3
HTP	0	0.08	0.09

Combine Fig 1 and S1

	BM-BB (Tg/yr)	BF-BB (Tg/yr)	FF (Tg/yr)	total (Tg/yr)	Ei/Etotal (fraction)
SAS	0.0170	0.0979	0.0880	0.2028	0.0261
EAS	0.0167	0.1003	0.5217	0.6388	0.0821
HTP	0.0000	0.0014	0.0031	0.0045	0.0006

Figure 6a (approx)
Ci to HTP (fraction)

SAS 0.49
EAS 0.17
HTP 0.11

Efficiency (from Fig 6, Fig 1, Table S1)
SAS 18.80
EAS 2.07
HTP 191.15

We appreciate the referee's effort in illustrating the point here, and we agree that the reason for the much higher efficiency of the HTP local emissions is quite obvious. With the significant contributions but very little emissions, the local sources are very effective in contributing to BC in the HTP. Although there is not much can be done to HTP local emissions for any mitigation actions, the quantitative efficiencies indicate that a small uncertainty in local emissions could largely impact the magnitude of local contributions to BC over the HTP. This efficiency metric is of more interest to policy makers, but it is also informative to researchers who prepare emission inventories for models and are interested in knowing the quantitative impact of uncertainties in regional emissions. We disagree with the referee that the efficiency is irrelevant to the focus of this study. However, we agree that the magnitude of efficiency for HTP emissions is relatively large and could potentially cause misinterpretations of the main results of this study. To address the referee's main concern, we have removed the HTP (local sources) efficiency from Figure 7, so the emphasis is now on the major source regions such as SAS and EAS.

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

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Abstract

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

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

1 Introduction

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

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

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

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

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

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

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

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

2 Model Configuration and Experimental Design

2.1 The CAM5 model and the source-tagging method

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

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

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

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

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

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

2.2 BC source regions and sectors

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

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

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

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

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

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

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

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

Where A_i^{BB} *and* A_i^{FF} are a BC property (e.g., mass mixing ratio, column burden, or deposition flux) in/over receptor region *j* resulting from BB and FF emissions, respectively, in source region *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 source regions (N = 16 in this study) and sectors (BB and FF). Note that for BC properties such as column burden, surface mixing ratio, and deposition flux, the tagging method in CAM5 explicitly calculates how much is due to emissions from each source region and sector.

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

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

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 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 Eq. (2) represents the global total emission rate. The efficiency metric S_i^{BB} *or* S_i^{FF} characterizes the sensitivity of aerosol properties in a receptor region to per unit (BB or FF) emissions in source region *i*. As noted in Wang et al. (2014), the efficiency is proportional to the metric of

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

3 Model evaluation against available observations

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

3.1 Atmospheric BC surface concentration

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

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

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

321 **3.2 Snow cover fraction**

322 It is important to evaluate the performance of model in simulating seasonal snow over this region
323 in order to assess the importance of BC-in-snow effect. Figure 3 shows the CAM5 simulated
324 seasonal and annual mean snow cover fraction (SCF) during year 2001, in comparison to
325 observed mean SCF, derived from the Moderate Resolution Imaging Spectrometer (MODIS)
326 (Hall et al., 2006) monthly mean of daily products at 0.05 degree resolution. For a better
327 comparison, the MODIS monthly mean SCFs are mapped to the CAM5 grid. The summer (JJA)
328 season only includes July and August for both CAM5 and MODIS due to missing MODIS data
329 in June 2001. To illustrate whether year 2001 can represent the average condition in terms of
330 SCF, the MODIS SCF climatology (2000-2013) is also plotted. The overall SCF in HTP has very
331 small difference between climatology and year 2001 in all seasons except for JJA, when SCF is
332 notably higher over northwest Plateau for the climatology that included June SCF in the average.
333 On average, SCF is about 5% (absolute amount) higher in June than in July and August. Over the
334 52 HTP grid cells, the CAM5 SCF is highly correlated spatially with that of MODIS (for both

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

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

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

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

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

4.1 Transport pathways

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

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

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

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

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

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

4.2 Seasonal variation of BC in the HTP

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

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

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

4.3 BC source-receptor relationships

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

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

453 The SAS emissions account for 50% of the annual mean burden over the HTP, including
454 33% from BB and 17% from FF. The other 50% is mostly from the EAS (5% BB and 14% FF),
455 HTP (6% BB and 6% FF), CAS FF (4%), MDE FF (4%) and SAF BB (3%). The source
456 attribution for annual mean BC deposition for the entire HTP is similar, but SAS contributes
457 even more to BC deposition than to the column burden. Although RBU has a lower contribution
458 to the annual mean BC in the HTP than the six regions shown in Fig. 6, its contribution to the
459 JJA mean, especially at some locations, is quite substantial (included in the black bar) and even
460 more important than some of the six regions in Fig. 6, as discussed in detail below.

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

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

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

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

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

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

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

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

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

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

4.4 Seasonal variation of HTP BC sensitivity

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

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

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

5 Radiative forcing

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

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

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

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

591 Dust is a major contributor to the total aerosol burden over the HTP (e.g., Zhang et al.,
592 2001). Although we don't focus on other snow impurities such as mineral dust, it is worth noting
593 that dust-in-snow radiative forcing has been considered in our model simulation and it could be
594 an important forcing agent. We also plotted dust-in-snow forcing over the HTP and sub-regions
595 in Fig. 8 (along with the BC-induced forcings). The annual mean dust-in-snow forcing (0.33 W
596 m^{-2}) is comparable to all of the other forcings over the HTP, especially in the springtime when
597 dust outbreaks and can be transported to the HTP from the surrounding sources such as
598 Taklimakan and Gobi deserts (Liu et al., 2008). The annual mean dust-in-snow forcing is as large
599 as 0.99 and 0.59 W m^{-2} in the Northwest and Northeast Plateau, respectively (Fig. 8b1 and f1),
600 which is in close proximity to the Taklimakan Desert (Huang et al., 2007; Chen et al., 2013), but
601 negligibly small in the Southeast Plateau and Central Plateau. In the winter, the dominant dust in
602 snow effect over Northeast Plateau is consistent with the recent observations. Huang et al. (2011),
603 X. Wang et al. (2013) and Zhang et al. (2013) found that insoluble light-absorbing particles in
604 snow are dominated by local soil and desert dust in the Qilian Mountain (Northeast Plateau).

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

6 Summary and conclusions

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

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

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

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

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

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

677 The impact of BC on snow and glacier melting can be characterized by the magnitude of
678 radiative forcing. Our calculations show that the annual mean BC-in-snow radiative forcing (0.42
679 W m^{-2}) outweighs BC dimming effect (-0.3 W m^{-2}) at the surface over the HTP. In the five sub-
680 regions, the annual mean BC-in-snow forcing ranges from 0.04 W m^{-2} in the Central Plateau to
681 1.75 W m^{-2} in the Northwest Plateau. We also showed that the annual mean dust-in-snow
682 induced radiative forcing over the HTP can be quite significant (0.33 W m^{-2} for entire the HTP,
683 and 0.99 W m^{-2} for the Northwest Plateau). More importantly, both BC- and dust-in-snow
684 forcing peaks in the spring melting season when the area-average forcing reaches 1.03 and 0.87
685 W m^{-2}, respectively, over the entire HTP, and their combined forcing is more than 8 W m^{-2} over
686 the Northwest Plateau. Such a large forcing is sufficient to cause earlier snow melting and
687 contribute to the acceleration of glacier retreat, although the model is likely to overestimate BC-
688 in-snow forcing due to the possible positive bias of snow cover fraction in the winter and early
689 spring. According to our estimates of the source attribution, the biomass burning and biofuel
690 emissions in South Asia contribute the most to annual mean forcing over the HTP and its sub-
691 regions except for the Northeast Plateau where the largest contribution is from East Asia fossil
692 fuel emissions. During the winter and spring seasons over Northwest Plateau and Himalayas,
693 when and where the forcing is the largest, South Asia (especially the biomass burning and
694 biofuel sector) is the major contributor.
695

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984

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

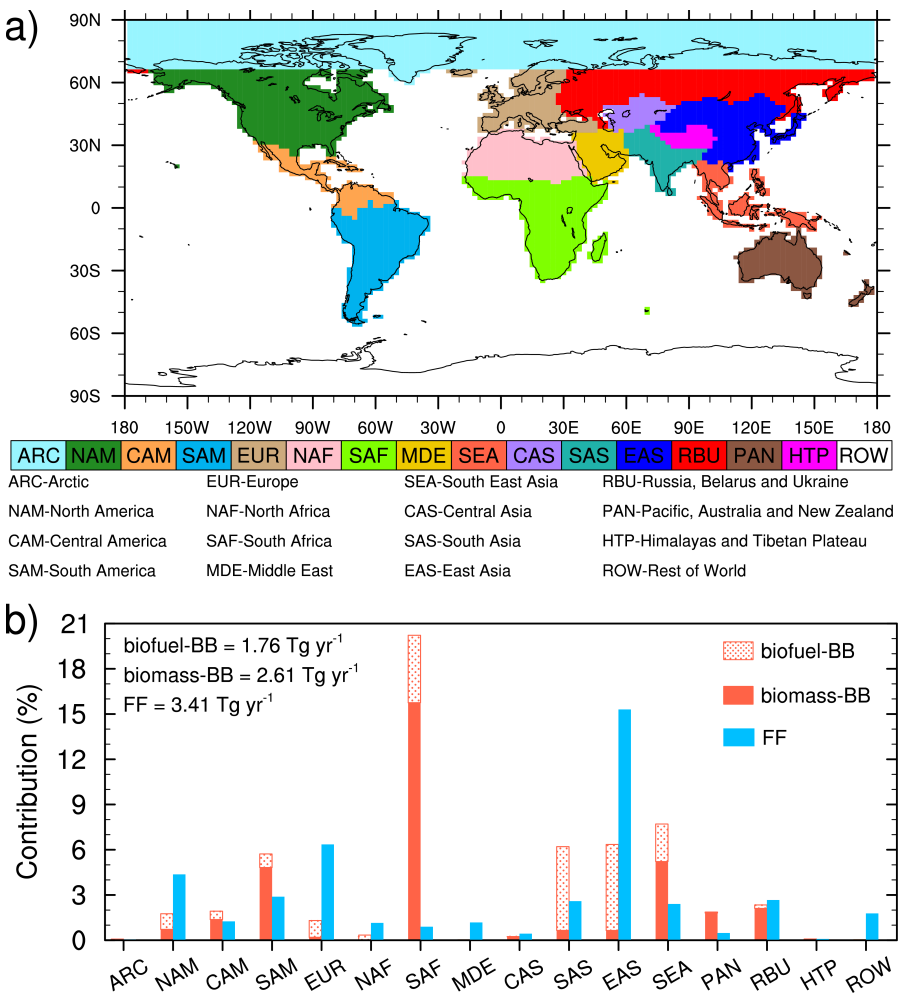


Fig. 1. (a) Tagged source regions and (b) the respective percentage contributions to global annual mean BC emissions from the individual source regions and sectors (including biofuel, biomass burning and fossil fuel). The global annual mean BC emission rate is 7.78 Tg yr^{-1}, which is divided up into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

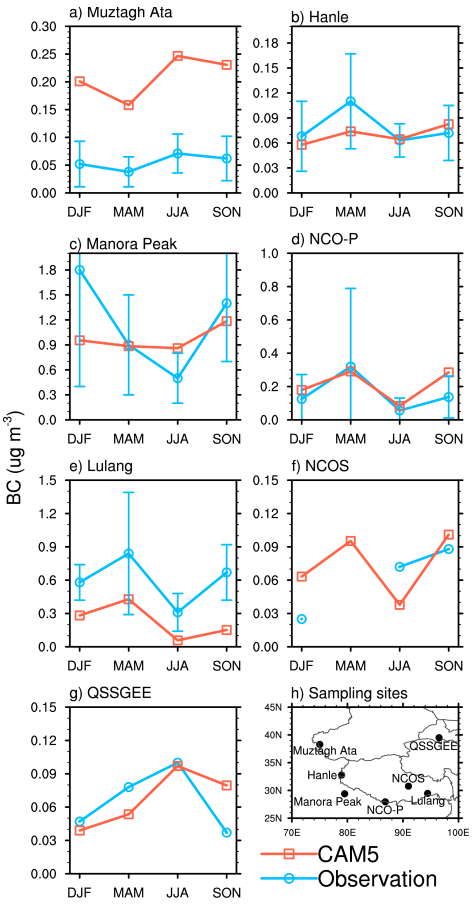


Fig. 2. Seasonal mean surface BC concentration (μg m^{-3}) from observations (blue lines with error bars denoting SD) and CAM5 simulation (red lines) at the seven sampling sites listed in Table 1 and marked in map of panel h.

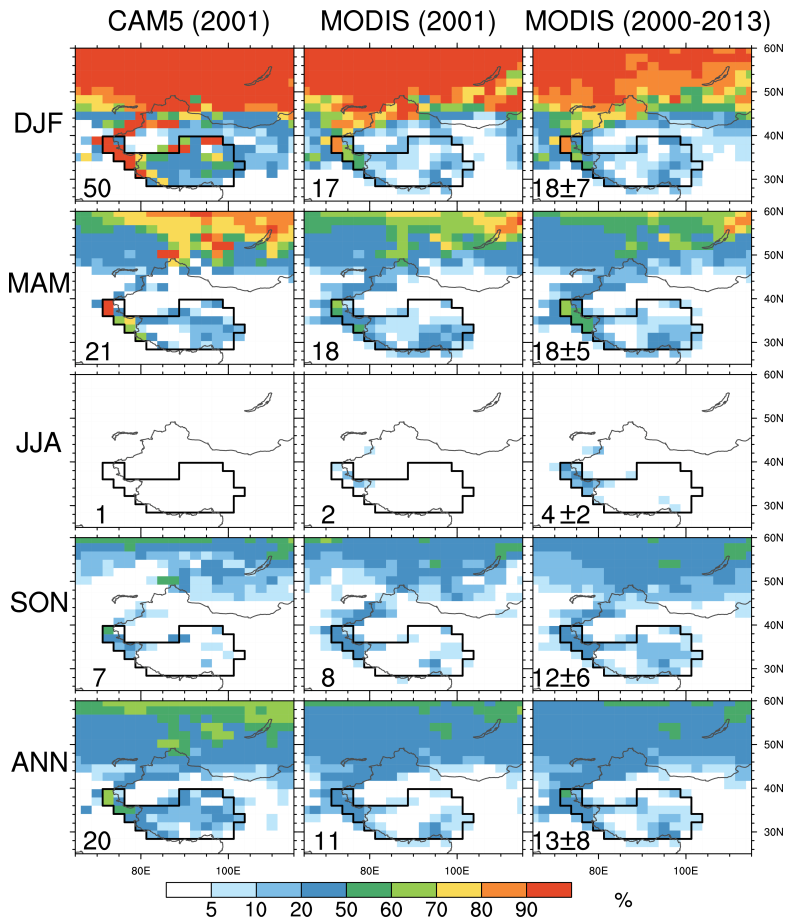


Fig. 3. Seasonal and annual mean snow cover fraction from CAM5 simulation for year 2001 (left), and MODIS retrieval for 2001 (middle) and 2000-2013 (right). The summer (JJA) in 2001 for both CAM5 and MODIS only includes July and August due to missing MODIS data in June. The number in the lower-left corner 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.

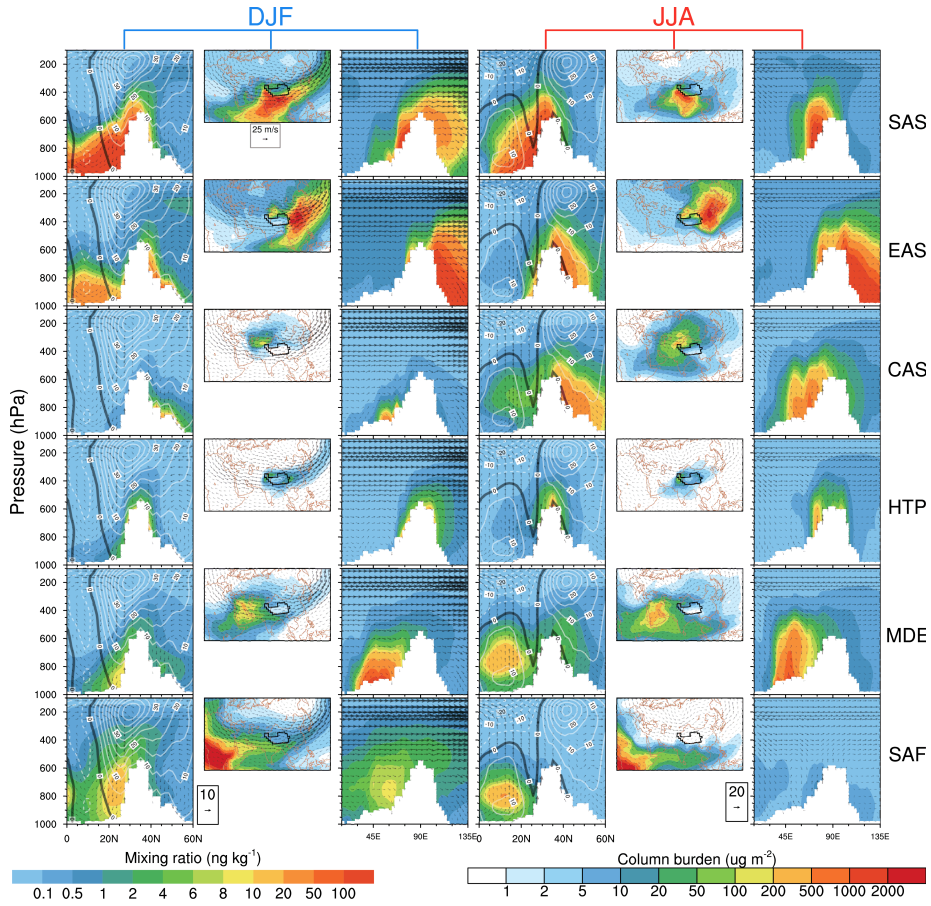


Fig. 4. The first column shows the latitude-height distributions of DJF BC mass mixing ratios (in ng kg^{-1}, colors) averaged over 71.25−101.25°E, originating from BB and FF sectors in the tagged source regions (corresponding to different rows); the white shaded area denotes topography, and the superimposed 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} 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 μg 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 28−40°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.

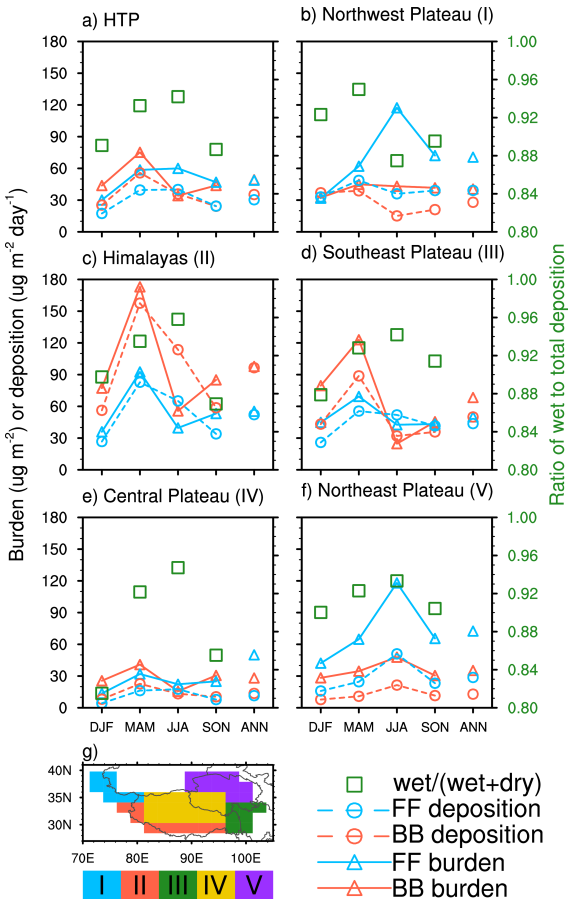


Fig. 5. Seasonal and annual mean BC column burden (solid lines and open triangles, in μg m^{-2}) and deposition rate (dashed lines and open circles, in μg m^{-2} day^{-1}) over (a) the HTP, (b) Northwest Plateau, (c) Himalayas, (d) Southeast Plateau, (e) Central Plateau and (f) Northeast Plateau, emitted 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.

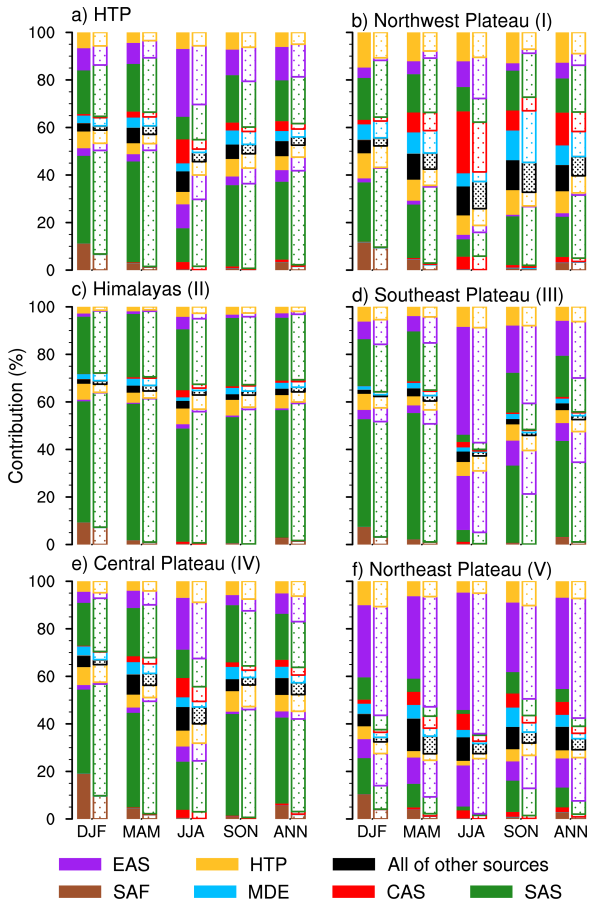
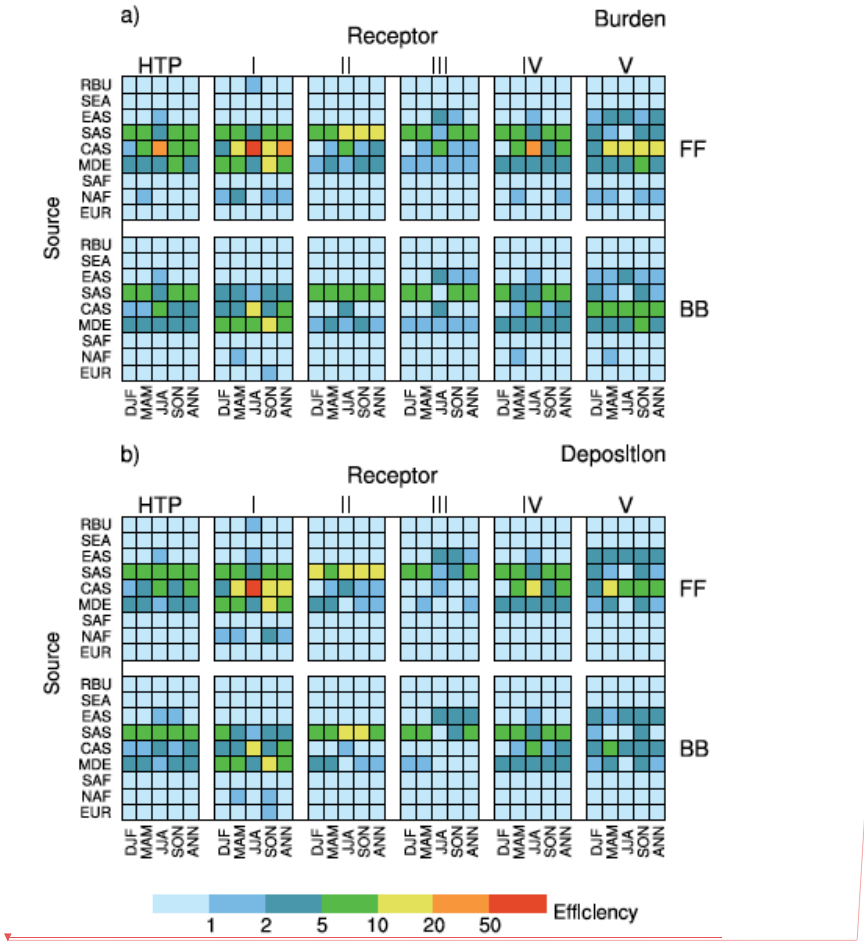


Fig. 6. Fractional contributions (measured by the lengths of color bars) to seasonal and annual mean BC column burden (solid pattern bars) and deposition (dotted pattern bars) over (a) the HTP, (b) Northwest Plateau, (c) Himalayas, (d) Southeast Plateau, (e) Central Plateau, and (f) Northeast Plateau, 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.



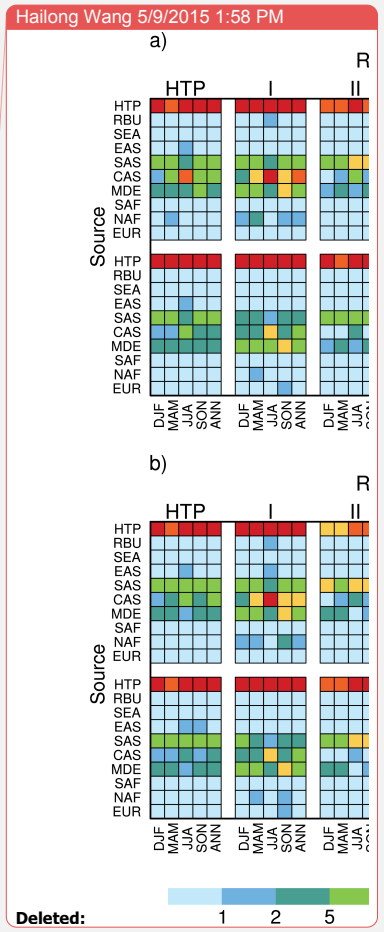


Fig. 7. Efficiency of FF (top) and BB (bottom) emissions from ten source regions (on the y-axis) in changing seasonal and annual mean (a) BC column burden and (b) deposition over the HTP and each of the five sub-regions: Northwest Plateau (I), Himalayas (II), Southeast Plateau (III), Central Plateau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

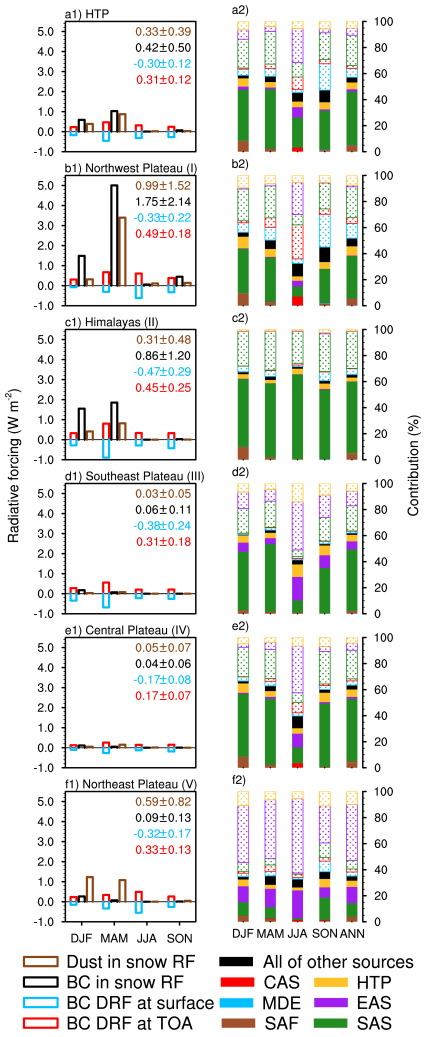


Fig. 8. Seasonal mean radiative forcing (left column) induced by the various BC effects (indicated by the color legend at the bottom) and dust-in-snow effect over (a1) the HTP, (b1) Northwest Plateau, (c1) Himalayas, (d1) Southeast Plateau, (e1) Central Plateau and (f1) Northeast Plateau. The corresponding 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.