

We thank the reviewers for their valuable comments and suggestions, which have helped us improve our manuscript in a great deal. We have made a revision accordingly. The point to point responses are provided below in Italic. The comparison of our manuscript between this version and the previous version is also provided.

Anonymous Referee #1

This was an interesting work. The authors used GEOS-Chem model to investigate the origin of transported biomass burning aerosols over the Tibetan Plateau. Their results indicated the 47% of BC in the TP was from South Asia and 46% was from East Asia. Actually, I have suspected these quantitative results. Because the MODEL showed a quite coarse resolution of 2 X 2.5. So, this kind of evaluation probably had great uncertainties while the model in a such coarse grid space was used in a complex terrain (such as TP). A result based on the regional climate model evaluated the contribution of BC from South Asia was nearly 61% in monsoon season and 20% in non-monsoon season (Yang JGR, 2018). And another study that used a high resolution WRF-CHEM also indicated that finer resolution model could represent more reasonable performance (Zhang ACP 2020, Impact of topography on black carbon transport to the southern Tibetan Plateau during the pre-monsoon season and its climatic implication). Their results found the complex topography in the model could generate 50% higher transport flux of BC in Himalayas. That's why I had such doubt need to explain by the authors. The manuscript was generally well written and comprehensive.

Response: Thanks for your comments. Although GEOS-Chem's resolution is relatively coarse, the comparison with observations from previous studies suggests that GEOS-Chem can generally capture the spatial and seasonal variations of surface BC in the TP and its surrounding regions (Table S1 and Figure S1). We compared the seasonal

variations in BC simulations with those in literature over different TP subregions. The comparison shows reasonable agreement between the simulations and observations. Please see the comparison and discussions in the supplement and in lines 170-180.

In this revision, we further ran a set of simulations using the emission perturbation method (see Supplement). The comparisons between the emission perturbation method and the backward-trajectory method developed in this study are reasonably well, suggesting the estimates from the backward-trajectory method can reasonably quantify the relative contributions of different source regions to surface BC in the TP.

We also compared the results from this study with literature (Lu et al., 2012; Zhang et al., 2015; Yang et al., 2018) and we found a general agreement between the two. (1) We estimated that the annual mean total contribution of South Asia and East Asia is 77%, and the estimates by Lu et al. (2012) and Zhang et al. (2015) are respectively 84% and 69%. (2) Our results showed that the contribution of South Asia is stronger in winter and weaker in summer, which was also suggested by Lu et al. (2012), Zhang et al. (2015), and Yang et al. (2018). Yang et al. (2018) modeled that the contribution of South Asia is 61% in non-monsoon season (October-April) and 19% in monsoon season (May-September). (3) The estimate of local contribution in this study and in Zhang et al. (2015) is both around 10%, indicating the dominant role of BC transport from nonlocal regions regarding the origins of the BC in the TP. Our results disagree with some of previous studies in various details. For instance, we estimated that the annual mean contribution of East Asia is approximately 35%, while the estimates by Lu et al. (2012) and Zhang et al. (2015) are respectively 17% and 19%. The discrepancy may be associated with the differences in region definitions and the estimation models. Please see discussions in lines 497-510 and 533-567.

In this study, the simulations are with a horizontal resolution of 2° latitude by 2.5° longitude. Such a resolution may not fully capture processes in the sub-grid scale, such as the mountain-valley wind (Cong et al., 2015). Using a regional model, Zhang et al. (2020) demonstrated that compared with simulations with a coarser resolution, simulations with higher resolution can better resolve the effects of topography and consequently get stronger transport flux of BC from South Asia to the TP. Therefore, our results may somewhat underestimate the contribution of South Asia because of the model resolution. Using regional models at higher resolutions in the future can better describe the terrain effect in the TP. Please see the discussions in lines 575-582.

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Zhang, R., Wang, H., Qian, Y., Rasch, P. J., Easter, R. C., Ma, P.-L., Singh, B., Huang, J., and Fu, Q.: Quantifying sources, transport, deposition, and radiative forcing of black carbon over the Himalayas and Tibetan Plateau, *Atmos. Chem. Phys.*, 15, 6205-6223, <https://doi.org/10.5194/acp-15-6205-2015>, 2015.

I also have minor comments as follows.

a. The definition for the source regions. Xinjiang in the northwestern China should be belong to CAS

Response: Thanks for your suggestion. In this revision, Xinjiang and part of Mongolia are defined as areas in Central Asia (see Figure 1b).

b. For the model performance evaluation, the comparison between in-site and model were not make sense. I did not believe the model output in a such great resolution could represent the surface situation. Probably a supplement if the authors want to show their model was credible.

Response: Thanks for the points. We have moved the comparison to Supplement. It is true that an observation can be compared with the model result in a grid only if the condition in that grid is homogenous. This may be somewhat the case at some remote and rural sites in the TP. The performance of GEOS-Chem in simulating BC over the TP and worldwide has been evaluated by scientists using in situ measurements of BC in surface air, and BC absorption aerosol optical depth (Kopacz et al., 2011; He et al., 2014, Li et al., 2015). He et al. (2014) found that the simulated BC in surface air are compared statistically well with observations at sites away from urban areas and the model can generally capture the seasonality of the observations, whereas the BC concentrations in the TP are likely to be underestimated by the model. We further evaluate the spatial and seasonal variations in GEOS-Chem BC simulations using observational data from literature and found a significant correlation ($r = 0.99$,

$p < 0.05$) between the observations and simulations at the rural and remote sites (Figure S1).

In this revision, we further compared surface BC concentrations, in terms of seasonal and interannual variations, from the GEOS-Chem simulations with those from MERRA2 reanalysis (The Modern-Era Retrospective analysis for Research and Applications version 2, M2TMNXAER, https://cmr.earthdata.nasa.gov/search/concepts/C1276812866-GES_DISC.html). The magnitude and spatial distribution of surface BC concentrations in the TP from GEOS-Chem and MERRA2 are highly consistent, with correlation coefficients over 0.98 in the four seasons (Figure S2). Furthermore, surface BC from GEOS-Chem and MERRA2 shows similar seasonal patterns over the TP as well as over East Asia, South Asia, and Southeast Asia (Table S2). For the interannual variations of surface BC over these regions, the similarity between the simulations and reanalysis is strong in spring, summer, and autumn, while is weak in winter (Table S2). Please see the discussions in Supplement and lines 170-189.

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Impacts of atmospheric transport and biomass burning on the interannual variation in black carbon aerosols over the Tibetan Plateau

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Abstract

Atmospheric black carbon (BC) in the Tibetan Plateau (TP) can largely impact regional and global climate. Still, studies on the interannual variation in atmospheric BC over the TP, and associated variation in BC sources and controlling factors are rather limited. In this study, we characterize the variations in atmospheric BC over the TP surface layer through analysis of 20-year (1995-2014) simulations from a global chemical transport model, GEOS-Chem. The results show that, [surface BC concentrations over the TP vary largely in space and by season, reflecting complicated](#)

interplays of BC sources from different origins. Of all areas in the TP, surface BC concentrations are highest over the eastern and southern TP, where surface BC are susceptible ~~respectively~~ to BC transport from East Asia and South Asia. ~~Combining respectively. Applying a backward-trajectory method that combines~~ the GEOS-Chem simulations and trajectories from the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, we assess the contributions of ~~differentworldwide~~ source regions to surface BC in the TP. ~~On~~We estimate that on the 20-year average, ~~over 9077%~~ surface BC in the TP comes from South Asia (~~4743%~~) and East Asia (~~4635%~~). Regarding seasonal variation in ~~foreignnonlocal~~ influences, South Asia and East Asia are dominant source regions in winter and summer, respectively, in terms of both magnitude and affected areas in the TP. ~~In spring and autumn, the influences from the two source regions are somewhat comparable.~~ Interannually, surface BC over the TP is largely modulated by atmospheric transport of BC from ~~foreignnonlocal~~ regions year-round and by biomass burning in South Asia, mostly in spring. We find that the extremely strong biomass burning in South Asia in the spring of 1999 greatly enhanced surface BC concentrations in the TP (31% relative to the climatology). We find that the strength of the Asian monsoon correlates significantly with the interannual variation in the amount of BC transported to the TP from ~~foreignnonlocal~~ regions. In summer, ~~stronga stronger~~ East Asian summer monsoon and South Asian summer monsoon tend to, respectively, ~~increaselead to more~~ BC transport from central China and ~~northeastnortheastern~~ South Asia to the TP. In winter, BC transport from central China is enhanced in years with a strong East Asia winter monsoon or Siberian High. A ~~strongstronger~~ Siberian High can also ~~increasebring more~~ BC ~~transport~~ from northern South Asia to the TP. This study underscores the impacts of atmospheric transport and biomass burning on the interannual variation in surface BC over the TP. It reveals a close connection between the Asian monsoon and atmospheric transport of BC from ~~foreignnonlocal~~ regions to the TP ~~and the Asian monsoon.~~

1 Introduction

Black carbon (BC) is a carbonaceous aerosol formed from combustion of carbon-based fuels and materials. BC in the atmosphere is a major air pollutant and a strong absorber of solar radiation (Bond et al., 2013). Atmospheric BC can greatly influence regional (Ramanathan and Carmichael, 2008; Zhuang et al., 2018) and global (Allen et al., 2012; Chung et al., 2012) climate through multiple mechanisms. It can cause atmospheric heating (Cappa et al., 2012) and surface dimming (Flanner et al., 2009) and influence cloud formation and development processes (Jacobson et al., 2012). Furthermore, after its deposition on snow or ice, BC reduces the surface albedo and accelerates the melting of glaciers and snow cover (Hansen and Nazarenko, 2004; Flanner et al., 2007).

The Tibetan Plateau (TP) has an average altitude of over 4 km and an area of 2.6 million km², known as the Third Pole. Because of its special geography, the TP can greatly impact regional and global climate through dynamic and thermal processes (Wu et al., 2015; Li et al., 2018). The TP has a large number of glaciers (over 0.1 million km²) and a wide coverage of snow- (59% of the TP in winter) (Qin et al., 2006; Yao et al., 2012). Although atmospheric BC in the TP is among the lowest in the world, BC there can alter the climate (Lau et al., 2010; Jiang et al., 2017), ecosystem (Kang et al., 2019), and hydrology (Barnett et al., 2005) in the TP, consequently influencing the living environment of billions of people in the world. Atmospheric BC is an important factor driving the surface warming in the TP due to its strong absorption of solar radiation (He et al., 2014a). After its deposition to the TP ground, BC in the snow reduces the surface albedo (Ming et al., 2009; Qian et al., 2011; Qu et al., 2014). He et al. (2014a) suggested that the annual mean direct radiative forcing and snow albedo forcing of BC in the TP are respectively ~2.3 and ~2.9 W m⁻² over the snow-covered regions. Because of the effects on radiation, BC

deposited on snow can further reduce the duration of snow cover in the TP (Ménégoz et al., 2014). Zhang et al., (2018) in the TP estimated that such reduction is around 3.1±0.1 days. Furthermore, BC in both atmosphere and cryosphere over the TP is responsible for retreats of the snow cover (Menon et al., 2010; Xu et al., 2016) and glaciers (Xu et al., 2009; Ming et al., 2012; Niu et al., 2020) in the past decades.

Atmospheric BC concentrations in the TP vary with location and season, which was revealed by limited observations over different regions in southern (Marinoni et al., 2010; Putero et al., 2014; Chen et al., 2018), northern (Zhao et al., 2012), and southeastern (Cao et al., 2011; Wang et al., 2018, 2019) TP. Over these regions, seasonal variations in atmospheric BC show different patterns. In Chen et al. (2018) observed that in the Himalayas over the southern TP, surface BC concentrations were observed to reach the highest in spring (over 0.9 $\mu\text{g m}^{-3}$ in April) and the lowest in summer (Marinoni et al., 2010; Chen et al., 2018). In the under 0.1 $\mu\text{g m}^{-3}$ in July) from May 2015 to May 2017. Zhao et al. (2012) reported that in the Qilian Shan over the northern TP, surface BC concentrations were reported less than 0.2 $\mu\text{g m}^{-3}$ from May 2009 to be March 2011 with the highest in summer and lowest in autumn (Zhao et al., 2012). Wang et al. (2016) suggested that surface BC concentrations show a seasonality of winter high and spring low at a site in the southeastern TP, while a pattern of winter low and spring high was at a site in the central TP. Few studies have explored the interannual variation in atmospheric BC concentrations over the TP (Mao and Liao, 2016). Since from November 2012 to June 2013. Because BC observations in the TP are limited due to the harsh environment and sparse sites, we can take advantages of numerical modeling on the variation in simulations to investigate atmospheric BC and its complicated variations over the TP entire TP in space and by season. Furthermore, how these variations fluctuate from year to year over decades is desirable an issue worthy of exploration.

Due to weak anthropogenic activities ~~and biomass burning~~, the contribution of local emissions to atmospheric BC in the TP is low (Zhang et al., 2015). Concentrations of atmospheric BC in the TP are greatly influenced by the long-range transport of BC from ~~foreign nonlocal~~ regions (Kopacz et al., 2011; [Lu et al., 2012](#); Kang et al., 2019). ~~Previous~~ [South Asia and East Asia are suggested to be two main source regions of atmospheric BC in the TP \(Lu et al., 2012\). Zhang et al. \(2015\) estimated that in 2001, local contribution to BC column burden in the TP was only around 10%, while the contributions from South Asia and East Asia are respectively about 50% and 20%. Some studies have also investigated the pathways of BC transport to the TP and derived their characteristics \(Cao et al., 2011\) and some of them suggested that South Asia and East Asia are two main source regions of atmospheric BC in the TP \(Lu et al., 2012\).](#) The Asian summer monsoon system was identified as an important influencing factor for ~~BC~~ transport [of atmospheric species](#) from South Asia to the TP (Chen et al., 2013; Han et al., 2014; Xu et al., 2014; Zhang et al., 2015). In summer, BC from northern India can be transported to the middle and upper troposphere and then crossing the Himalayas to the TP via southwesterly winds (Yang et al., 2018). BC ~~emission~~ [emitted](#) in East Asia can ~~also~~ be uplifted to upper layers by [the](#) summer monsoon circulation and then transported to the northeastern TP (Zhang et al., 2015). The midlatitude westerlies are favorable ~~for~~ [to](#) BC transport from central Asia and northern India to the western TP (Chen et al., 2018) but unfavorable ~~for~~ [to](#) BC transport from eastern China to the TP (Cao et al., 2011). Although previous studies explored the mechanisms of BC transport to the TP, large uncertainties remain in the quantified fractional contributions of BC transport from different source regions to the TP (Yang et al., 2018). More importantly, how BC transport to the TP varies interannually and what are underlying mechanisms for the variation are unclear. Therefore, it is necessary to examine how ~~seasonal~~ BC transport to the TP varies from year to year and ~~whether there is a connection between~~ [how](#) the Asian monsoon ~~and affect~~ the ~~interannual~~ [variation in BC transport to the TP](#).

~~Observations~~Previous observations and simulations showed ~~previously~~ that ~~anthropogenic and fire emissions are~~biomass burning is a major ~~source~~source of atmospheric BC in the TP (Lu et al., 2012; Zhang et al., 2015). Zhang et al. (2015) estimated that biomass burning together with biofuel emissions can contribute to around half of the annual mean BC column burden over the TP. Engling et al. (2011) reported that BC emissions from fire events in Southeast Asia in spring could probably increase the BC concentrations over a mountain site in the southeastern part of the TP. Putero et al. (2014) suggested that over half of the high BC episodes in the southern Himalayas were likely affected by the fire events in South Asia. However, these studies demonstrated the influences of biomass burning in a relative short term or during some fire events, few investigated the influences in a long term over a decade. ~~(Mao and Liao, 2016)~~. The influence of biomass burning on the interannual variation in atmospheric BC over the TP warrants an in-depth study.

In this study, we aim to assess the impacts of atmospheric transport and biomass burning on surface BC concentrations over the TP, especially on the interannual variation in BC during 1995-2014. To estimate BC transport from different source regions to the TP, we adopt ~~a numerical an~~ approach based on a global chemical transport model, GEOS-Chem (Bey et al., 2001), and a trajectory model, the Hybrid Single-Particle Lagrangian Integrated Trajectory model (HYSPLIT) (Draxler and Hess, 1998; Stein et al., 2015). In the following, the method and models are described in section 2. Section 3 discusses the seasonal variations in surface BC over the TP and in BC transport from source regions to the TP based on the mean status of the 20-year simulations. The interannual variation in surface BC over the TP and the impacts of biomass burning and transport on this variation are analyzed in section 4.

~~Conclusions, along with discussion,~~Discussion and conclusions are provided in ~~section~~sections 5 and 6, respectively. In this paper, BC refers to BC aerosols in the

atmosphere. Surface BC refers to atmospheric BC aerosols in the surface layer.

2 Data and methods

2.1 GEOS-Chem simulations

A global chemical transport model, GEOS-Chem (version v9-02, <http://geos-chem.org>) (Bey et al., 2001), is used ~~in this study~~ to simulate global BC concentrations. GEOS-Chem is driven by the NASA Modern-Era Retrospective Analysis for Research and Applications (MERRA) meteorological data (Rienecker et al., 2011). In this study, we focused on how surface BC in the TP responds to interannual variations in natural ~~processes~~influences including biomass burning and meteorology. Therefore, anthropogenic emissions in our ~~simulations~~simulations were allowed to vary seasonally but not interannually, i.e. anthropogenic emissions in 2000 including their seasonality were used for ~~20 simulation years each year of the study period~~. We conducted three GEOS-Chem simulations ~~in this study~~: CTRL, FixBB, and FixMet. The three simulations covered the study period from 1995 to 2014 (using 1994 for spin-up) at 2° latitude by 2.5° longitude horizontal resolution with 47 vertical layers. In CTRL, both biomass burning emissions and meteorological fields varied interannually. In FixBB, interannual meteorology was allowed and fire emissions were fixed in 2005, ~~so~~ to remove the impact of the interannual variation in biomass burning on BC concentrations in the TP. In FixMet, emissions from biomass burning were allowed to vary interannually and meteorology was fixed in 2005, ~~so~~ to remove the impact of interannual meteorology.

In the simulations, global anthropogenic BC emissions were based on Bond et al. (2007), with an annual emission of 4.4 Tg C in 2000 (Leibensperger et al., 2012). Global biomass burning emissions of BC were from the Global Fire Emissions Database version 3 (GFED3) inventory (van der Werf et al., 2010), which covers the period of 1997-2011. BC in GEOS-Chem is represented by two tracers: hydrophobic

and hydrophilic (Park et al., 2003). Freshly emitted BC is mostly (80%) hydrophobic (Cooke et al., 1999). Hydrophobic BC becomes hydrophilic typically in a few days (McMeeking et al., 2011), which is simply assumed as 1.15 days in the model, called an e-folding time (Cooke et al., 1999; Park et al., 2005; He et al., 2014b). Simulations of aerosol dry and wet depositions follows Liu et al. (2001). Dry deposition of aerosols is simulated using a resistance-in-series model (Walcek et al., 1986) dependent on local surface type and meteorological conditions, while wet deposition scheme includes scavenging in convective updrafts, as well as in-cloud and below-cloud scavenging from convective and large-scale precipitation. Dry deposition is generally smaller than wet deposition (He et al., 2014b; [K. Li et al., 2015](#)[2016](#)). Tracer advection is computed every 15 minutes with a flux-form semi-Lagrangian method (Lin and Rood, 1996). The tracer moist convection scheme follows Allen et al. (1996a, b), using GEOS convection, entrainment, and detrainment mass fluxes. The deep convection is parameterized using the relaxed Arakawa-Schubert scheme (Arakawa and Schubert, 1974; Moorthi and Suarez, 1992) and for the shallow convection, the scheme in Hack (1994) is used.

[GEOS-Chem has been widely used for BC simulations in the world \(Park et al., 2003, 2005; K. Li et al., 2016\) and over the TP and its surroundings \(Kopacz et al., 2011; Lu et al., 2012; He et al., 2014b\)](#)

[2.2 GEOS-Chem evaluation](#)
[GEOS-Chem simulations of surface BC concentrations were previously evaluated over the TP and China \(He et al., 2014b;2014a; He et al., 2014b; Mao and Liao, 2016\). The performance of GEOS-Chem in simulating BC over the TP and surroundings was systematically evaluated by He et al. \(2014b\) using in situ measurements of BC in surface air, BC in snow, and BC absorption aerosol optical depth. He et al. \(2014b\) found that the simulated BC in surface air are compared statistically well with observations at sites away from urban areas and the model can generally capture the seasonality of the observations, whereas the BC concentrations](#)

in the TP are likely to be underestimated by the model. In Supplement, we evaluated the spatial and seasonal variations in GEOS-Chem BC simulations using observational data from literature. We found a significant correlation ($r = 0.99$, $p < 0.05$) between the observations and simulations at the rural and remote sites (Figure S1). We further compared surface BC concentrations from the GEOS-Chem simulations with those from MERRA2 reanalysis (The Modern-Era Retrospective analysis for Research and Applications version 2, M2TMNXAER, https://cmr.earthdata.nasa.gov/search/concepts/C1276812866-GES_DISC.html). The magnitude and spatial distribution of surface BC concentrations in the TP from GEOS-Chem and MERRA2 are highly consistent, with correlation coefficients over 0.98 in the four seasons (Figure S2). Furthermore, surface BC from GEOS-Chem and MERRA2 shows similar seasonality over the TP as well as over East Asia, South Asia, and Southeast Asia (Table S2). Regarding the interannual variations of surface BC over these regions, the similarity between the simulations and reanalysis is high in spring, summer, and autumn but low in winter (Table S2).

2.2 Fire and meteorological data

~~Li et al., 2015). Here, we validated the model in the TP and its surrounding regions for enhanced confidence. As surface BC measurements in the TP are rather limited, the observation data collected at 13 sites (Figure 1) from previous literature (Carriero et al., 2003; Qu et al., 2008; Zhang et al., 2008; Beegum et al., 2009; Ganguly et al., 2009; Bonasoni et al., 2010; Ming et al., 2010; Pathak et al., 2010; Ram et al., 2010a, b; Nair et al., 2012) were used in this study, following He et al. (2014b). The 13 sites were grouped into urban, rural, and remote sites (He et al., 2014b). The observational data are available for 2006 at 9 of the 13 sites and available at the other sites for different periods, i.e., 1999–2000, 2004–2005 and 2008–2009. BC observations at a remote site during 2015–2017 from another study (Chen et al., 2018) were also used.~~

The annual mean surface BC concentrations from GEOS-Chem and observations are compared in Table 1. The observed surface BC concentrations are below $2 \mu\text{g m}^{-3}$ at remote sites, about $2\text{--}5 \mu\text{g m}^{-3}$ at rural sites, and as high as $5 \mu\text{g m}^{-3}$ at urban sites (He et al., 2014b). Compared with the observations, GEOS-Chem performs well at the remote sites, moderately at the rural sites, and poorly at the urban sites (Table 1). The simulations substantially underestimate surface BC concentrations at the urban sites, likely due to the coarse horizontal resolution in the model that dilutes the intensity of local emissions in a model grid. Taking the rural and remote sites only (Figure 2a), we found a high consistency between the annual mean simulations and observations, with a significant correlation coefficient of 0.99. The comparison suggests that GEOS-Chem can generally capture the spatial variation in surface BC concentrations over the TP. Moreover, the seasonality of simulated surface BC concentrations was evaluated at three sites (Figures 2b–2d). GEOS-Chem simulates low BC concentrations in summer and high BC concentrations in winter and spring at the sites. The amplitude of the seasonal variation in the simulations is weaker than that in the observations. Moorthy et al. (2013) found that simulated surface BC concentrations by the Goddard Global Ozone Chemistry Aerosol Radiation and Transport (GOCART) model in winter were lower than the observed ones at a TP site and they attributed this to the biases in the atmospheric boundary layer parameterization scheme. Wintertime surface BC concentrations were also underestimated by the Community Atmosphere Model version 5 (CAM5, Zhang et al., 2015), suggesting a common bias in these models.

2.3 Meteorological and fire data

The meteorological data used in this study are the NCEP/NCAR (National Centers for Environmental Prediction/National Center for Atmospheric Research) reanalysis, available from the Physical Sciences Division of NOAA Earth System Research Laboratory

~~(<https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.surface.html>). The data include geopotential height and wind. The horizontal resolution is 2° latitude \times 2.5° longitude.~~

The nighttime fire count product retrieved from ATSR (Along Track Scanning Radiometer) using Algorithm 2, available from European Space Agency (http://due.esrin.esa.int/page_wfa.php), were used to verify the biomass burning emissions in ~~the model~~ [GEOS-Chem](#). ATSR is onboard the Second European Remote-Sensing Satellite (ERS-2). The spatial resolution of the data is 1 km, and the sensor achieves a global coverage every three days. The ATSR satellite data with the period of 1997-2011 were gridded to the GFED3 grids with a resolution of $0.5^{\circ} \times 0.5^{\circ}$ in longitude and latitude.

[2.4 The meteorological data used in this study for analysis and for driving the backward trajectories](#) are the [NCEP/NCAR \(National Centers for Environmental Prediction/National Center for Atmospheric Research\) reanalysis, available from the Physical Sciences Division of NOAA Earth System Research Laboratory](#) (<https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.surface.html>). The data include geopotential height and wind. The horizontal resolution is 2° latitude \times 2.5° longitude.

2.3 Transport estimation

Combining GEOS-Chem simulations and HYSPLIT (version 4, http://www.arl.noaa.gov/HYSPLIT_info.php, Draxler and Hess, 1998; Stein et al., 2015) trajectories, we estimated the contributions of different source regions in the world to surface BC in the TP during 1995-2014. HYSPLIT is an atmospheric transport and dispersion model (Fleming et al., 2012), developed by the Air Resources Laboratory of the National Oceanic and Atmospheric Administration (NOAA).

Meteorological inputs to HYSPLIT are the NCEP/NCAR reanalysis at a resolution of 2.5° latitude \times 2.5° longitude. We evenly divided the TP into 70 GEOS-Chem grids. Considering that the average lifetime of atmospheric BC is about a week, we simulated 7-day backward trajectories originated from each of the 70 grids. The trajectories were initialized four times a day (00, 06, 12 and 18 UTC) during 1995-2014. The starting altitude for the trajectories is 100 m above ground which is within the typical planetary boundary layer in the TP (Ram et al., ~~2010b~~2010). We divided the world into ~~six~~seven regions (Figure 1b), including ~~the TP~~, central Asia, East Asia, South Asia, Southeast Asia, the region of other Asia, Europe, and Africa, and the rest of the world. BC concentrations from CTRL simulation were used in the estimation ~~of~~ BC from different source regions.

Lu et al. (2012) proposed a novel approach that combined BC emissions with ~~the~~ backward trajectories from the TP to quantify the origins of BC in the TP. Modifying Lu et al. (2012)'s approach, we ~~combined~~combine BC concentrations, instead of BC emissions in Lu et al. (2012), with ~~the~~ backward trajectories from the TP for the same purpose. We assume that BC aerosols have a lifetime of D days and the backward trajectories using HYSPLIT are simulated for D days ($D=7$ in this study). To make the estimation stable, the amount of BC transported to a TP surface grid on a day is assumed to be a mean of the BC transport along the backward trajectories originated from that grid in the past D days, i.e.,

$$BC_{imported} = \frac{\sum_{d=1}^D BC_d}{D} \quad (1)$$

where BC_d is the amount of BC that are transported to that TP surface grid along the backward trajectory on a previous day d ($d=1, 2, \dots, D$).

Equation (1) provides a way to estimate the amount of BC that is transported to the TP from any model grid outside the TP during a period of interest. For a grid $g_{i,j,k}$, ~~the total amount ($C_{i,j,k}$) of BC transported from $g_{i,j,k}$ to the TP is estimated~~

outside the TP (by

$$C_{i,j,k} = \frac{\sum_{n=1}^N c \times v}{D \times M} \quad (2)$$

where i, j, k are indices for the model grid in longitude, latitude, and altitude coordinates, respectively), the total amount of BC transported from $g_{i,j,k}$ to the TP during a period of interest ($C_{i,j,k}$) can be estimated by

$$C_{i,j,k} = \frac{\sum_{n=1}^N c \times v}{D \times M} \quad (2)$$

where n is an index for the number of trajectories. N is the total number of trajectories that have passed through the grid $g_{i,j,k}$ during the period of interest, for example, in a month. c is the daily BC concentrations at $g_{i,j,k}$ when trajectory n passing $g_{i,j,k}$, and v is the volume of $g_{i,j,k}$. M is the number of trajectories in a day ($M=4$ in this study).

Therefore, the total amount of BC transported to the TP ($T_{i,j}$) from the entire tropospheric column above a surface grid $g_{i,j,0}$ in a source region outside the TP during the period of interest is can be assessed by

$$T_{i,j} = \sum_{k=1}^K C_{i,j,k} \quad (3)$$

where K is the number of model layers in the troposphere.

Finally, the amount of BC transported from a nonlocal source region to the TP surface can be summed up and the fractional contributions of different source regions to surface BC in the TP can be quantified. This developed method is inspired by Lu et al. (2012) and is robust and stable because it is not sensitive to the number of trajectories taken in a day (M) and the number of days taken for the trajectories (D).

Using this method, the amount of BC transported from a source region nonlocal grid to the TP surface is determined by both BC concentrations over in that region grid and the number of trajectories passing through that region grid within the tropospheric column. An example of the estimation of BC transport to the TP surface in April 2005 is shown in Figure 32. In this example, BC concentrations are high in central China

around 110°E (Figure 3a2a). A large number of trajectories pass through central China and finally arrive at the TP surface (Figure 3b2b). Therefore, the amount of BC transported from central China to the TP is high (Figure 3e2c). In contrast, although trajectories pass through central Asia are also in large numbers, the amount of BC transported from central Asia to the TP is small (Figure 3e2c) because ~~of the low~~ BC concentrations over central Asia are low and the trajectories appear at high altitudes (Figures 3a2a and 3b2b).

3 Seasonal variations in surface black carbon over the Tibetan Plateau and in black carbon transport to the Tibetan Plateau

~~BC transport from each~~In this section, the analysis is based on the mean status of the ~~source regions to the TP surface varies with season. 20-year simulations.~~ Figure 43 shows the amount of BC transported from each of the GEOS-Chem ~~grid~~grids through the tropospheric column to the TP surface in the four seasons, which varies greatly in space and among seasons. Obviously, surface BC in the TP mainly originates from South Asia and East Asia, especially from the regions near the southern and eastern borders of the TP, including central China, northeastern South Asia, and northern South Asia. These spatial distributions of BC_{nonlocal} contributions ~~from different source regions to the BC~~ in the TP in four seasons are similar to those in Lu et al. (20022012). We also found a good agreement ($r=0.72$, $p<0.05$) in the estimation of imported BC between this study and Lu et al. (2012) at several sites in the TP, although our estimates are higher than those from Lu et al. (2012). The ~~back~~backward-trajectory approach modified in this study shows strong performance in identifying the BC-source regions for BC over the TP.

The simulated annual mean surface BC concentrations over the entire TP are shown in Figure 5a4a. The BC concentrations are high along the eastern and southern borders and low in the center of the TP. This spatial variation in surface BC

concentrations dominates in the four seasons (not shown). BC concentrations over the TP show strong spatial gradient, which is likely due to the blocking of BC transport by the mountains with high elevations (Cao et al., 2011; Zhao et al., 2017). Figures ~~5b-5f~~4b-4f show the dominant nonlocal influences to surface BC over the TP. In terms of affected areas, South Asia and East Asia are the most important source regions for the TP, which respectively impact 67% and 25% areas of the TP in the annual mean and by season.BC (Figure 4b). In terms of the annual mean, 93% amount of BC imported, 77% of surface BC in the TP comes from South Asia (47%) and East Asia (46%), which are two dominant source regions for the TP as BC aerosols from South Asia(43%) and East Asia can impact 64% and 34% areas of the TP, respectively (Figure 5b).(35%). Because of the leeward location of East Asia under prevailing westerlies, the influence of East Asia is constrained mainly in northern and eastern TP. In winter, South Asia is the dominant source region for 83% areas of the TP (Figure 5f), while in summer, East Asia is the dominant source region for 54% areas of the TP (Figure 5d).Using a backward-trajectory method, Lu et al. (2012) estimated that the contributions of South Asia and East Asia are respectively 67% and 17%. Using a tracer tagging approach, Zhang et al. (2015) suggested that the contributions of the two regions are respectively 50% and 19%. Our estimate of fractional contribution of both South Asia and East Asia is comparable with the two previous studies, while the estimated fractional contribution of East Asia is higher than them.

Seasonally, the contribution of South Asia to surface BC in the TP is strongest in winter (55%) and weakest in summer (33%). South Asia is identified as the dominant contributor for 83% and 41% areas of the TP in winter and summer, respectively (Figure 4). In contrast, the contribution of East Asia is highest in summer (45%) and lower in winter (23%). East Asia is identified as the dominant contributor for 34% and 11% areas of the TP respectively in summer and winter (Figure 4). These seasonal variations in the contributions of South Asia and East Asia are generally in agreement

with earlier studies (Lu et al., 2012; Zhang et al., 2015; Yang et al., 2018) with various numerical methods. For instance, using the emission perturbation method, Yang et al. (2018) suggested that the contribution of South Asia is 61% in non-monsoon season (October-April) and 19% in monsoon season (May-September).

We further divided the TP into five subregions, namely, eastern TP, southern TP, western TP, northern TP, and central TP (Figure 5a4a). The 20-year means in the subregions show different BC levels, seasonalities, and dominant BC source regions (Figures 5-74-6). Over the eastern TP, the mean surface BC concentrations are the highest among the five subregions (Figure 6b). Over 755b). Above 70% of surface BC in the eastern TP is transported from East Asia (Figure 7b6b).

In the southern TP, the mean surface BC concentrations are the 2nd highest among the five subregions, which are high in spring and low in the other seasons (Figure 6e5c). Such seasonality is likely resulted from the high fire emissions over South Asia in spring, the favorable atmospheric circulation for BC transport to the southern TP in spring, and the strong wet deposition of BC by the monsoon precipitation in summer (Chen et al., 2018). This seasonality is in consistency with observations in previous studies literature (Marinoni et al., 2010; Cong et al., 2015). South Asia is the dominant source region for surface BC in the southern TP year-round, with fractional contributions of over 8573% (Figure 7e6c). The dominant contribution of South Asia to the southern TP was also suggested in previous studies (He et al., 2014a; Zhang et al., 2015; Yang et al., 2018). The second keydominant source region for the southern TP is Southeast Asia, which contributes 13% BC to surface BC in the region in the annual mean.

Over the western TP, the mean BC concentrations are the 3rd lowest among the five subregions, with a seasonality of high BC in winter and spring and low BC in

summer and autumn (Figure 6d5d). The higher values in spring and winter agree with the BC measurements at sites in the western Himalayas (Nair et al., 2013). BC transport from South Asia contributes to 9382% of surface BC in winter and 7670% in summer (Figure 7d6d). Such seasonality with winter high and summer low in the fractional contribution of South Asia to surface BC over the western TP were also suggested by Zhang et al. (2015).

In the northern TP, the mean BC concentrations are the 2nd lowest among the five subregions, which are at maximum in winter and minimum in spring (Figure 6e5e). This seasonality is different from an observational study, which reported that, over the Qilian Shan in the northern TP, surface BC concentrations are highest in summer and lowest in autumn (Zhao et al., 2012). The dominant source region for surface BC over the northern TP is central Asia in spring and autumn, East Asia in summer, and South Asia in winter and East Asia in the other seasons (Figure 7e6e). Influenced by the prevailing westerlies, ~~source regions west of the TP (central Asia, the region of othercentral Asia, Europe, and Africa in Figure 1b)~~ contribute to surface BC in the western (Figure 6d) and northern TP (Figure 6e) more than to other TP subregions (Figure 7e). ~~Source regions west of the TP. In the annual mean, central Asia can contribute to 5-119% and 9-1727% surface BC in the western (Figure 6d) and northern (Figure 6e) TP, respectively (Figures 7d and 7e).~~

Among the five subregions, the central TP is with the lowest BC concentrations (Figure 6f). ~~Source regions other than South Asia and East Asia contribute only 5% or less to surface BC in the central TP (Figure 7f5f)~~. Seasonally, BC concentrations over the central TP is higher in spring and winter than in summer and autumn. This simulated seasonality is different from the observed one at a site in the central TP reported in Wang et al. (2016), who showed that BC concentrations are higher in spring and lower in winter from November 2012 to June 2013. In spring South Asia is

the dominant source region for surface BC in the central TP and its influence is strongest in winter, South Asia respectively contributes contributing to 72% and 91%77% of surface BC in this subregion. In-contrast,In contrast, the influence of East Asia contributes is strongest in summer, contributing to 58%34% of surface BC there in summer.

The local contribution of the TP and the contribution of Southeast Asia are much lower than those of South Asia and East Asia (Figure 6). In the annual mean, local contribution to surface BC in the TP is approximately 11% (Figure 6a). Among all the TP subregions, the fraction in the local contribution is highest in the central TP, being ~14% in the annual mean (Figure 6f). Using a tagging tracer method, Zhang et al. (2015) also suggested that local emissions contribute about 10% of BC in the TP. Influenced by the prevailing westerlies, BC from Southeast Asia cannot be efficiently transported to the TP by atmospheric circulation. The annual mean contribution of Southeast Asia to surface BC in the TP is ~7% (Figure 6a). Seasonally, the contribution of Southeast Asia is highest in spring (10%) (Figure 6a) when fire BC emissions are highest there. Geographically, the contribution of Southeast Asia is highest in the southern TP (13% in the annual mean) (Figure 6c) because of its proximity to Southeast Asia.

4 Interannual variation in surface black carbon over the Tibetan Plateau

4.1 Influences of biomass burning on the interannual variation in surface black carbon over the Tibetan Plateau

Figure 87 shows the anomalies of surface BC concentrations averaged over the TP from the three GEOS-Chem simulations. Note that in these simulations, only natural influences of meteorological and biomass burning in the interannual variation of surface BC in the TP are considered. The simulations from CTRL and FixBB are significantly correlated with each other in all the seasons ($r=0.45$, $p<0.05$ in spring,

$r > 0.8$, $p < 0.05$ in the other three seasons), indicating the important role of meteorology in the interannual variation in surface BC concentrations in the TP. Remarkably, in spring (Figure 8a7a), the correlation coefficient of BC anomalies between CTRL and FixMet simulations reaches 0.87 ($p < 0.05$), indicating the importance of biomass burning to the interannual variation in BC in spring. The largest anomaly of BC concentrations from CTRL simulation is in 1999. The comparison between CTRL and FixMet simulations suggests that this strong anomaly is largely explained by biomass burning. Even if we exclude the extreme year 1999, the correlation ($r = 0.77$, $p < 0.05$) between CTRL and FixMet simulations remains significant, indicating the strong influence of biomass burning on the variation in ~~surface BC from year to year.~~ springtime surface BC from year to year. In Supplement (Figure S3), we applied the mean absolute deviation and absolute percent departure from the mean to quantitatively represent the strength of the interannual variation in surface BC over the TP, similar to Mao and Liao (2016). Figure S3 further confirms that biomass burning is an important driver of the interannual variation in surface BC over the TP in spring, while meteorology is more important than biomass burning in the other seasons.

To further examine the influence of biomass burning in spring, we integrally analyzed data from ATSR satellite fire counts, GFED3 fire emissions, and the GEOS-Chem simulations. Both ATSR and GFED3 data show that fires occur frequently over the Indo-Gangetic Plain, central India, and Southeast Asia (Figures 1 and 98). Fire activities in Asia are well described in the GFED3 inventory that is used in the GEOS-Chem simulations (Figures 9b8b and 9e8c). We found the interannual variation in BC anomalies in the TP from CTRL simulation is significantly correlated to the total number of fire counts in the Indo-Gangetic Plain ($r = 0.76$, $p < 0.05$), and central India ($r = 0.67$, $p < 0.05$). The correlation ~~was~~ is insignificant ~~for~~ to the total number of fire counts in Southeast Asia ($r = 0.19$, $p > 0.05$). In spring of 1999, extreme fire activities

occurred in the Indo-Gangetic Plain and central India (Figure ~~9b~~8b). Driven by the ~~favourable~~favorable atmospheric circulation, the strong BC emissions from the ~~extremely active fires~~extreme fire activities greatly enhanced surface BC concentrations in the TP (Figure ~~9d~~8d). In ~~the~~ CTRL simulation, positive BC anomalies appear over the entire TP, with a regional mean of ~~0.1508~~ $0.0201 \mu\text{g m}^{-3}$ or 31% relative to the 1995-2014 climatology (Figure ~~8a~~7a). ~~Even when 1999 spring is removed from the time series, biomass burning still shows a large impact on interannual surface BC over the TP in spring (Figure S3).~~ Additionally, in winter, biomass burning was extremely strong in 1998 (Figure ~~8d~~7d). ~~The extremely active fires~~7d, which enhanced the regional mean surface BC concentrations in the TP by ~~0.0201~~ $0.0201 \mu\text{g m}^{-3}$ or 5% relative to the climatology.

~~According to Figure 8, In the simulations between CTRL and FixMet are not significantly correlated in summer, autumn, and winter, suggesting that meteorology plays an following, the important role of meteorology in modulating the interannual variation in surface BC inover the TP. Such a role will is to be explored in sections 4.2 and 4.3 from a perspective of the influences of the Asian monsoon on BC transport to the TP in summer and winter, respectively.~~

4.2 Influences of the Asian summer monsoon on the interannual variation in transport of black carbon to the Tibetan Plateau from nonlocal regions in summer

The TP can largely impact the Asian monsoon system through thermal and dynamic processes (Wu et al., 2015). In the meantime, the Asian monsoon can significantly influence the transport of atmospheric species to the TP (Xu et al., 2014). In this section, we show the influences of two Asian monsoon subsystems on the interannual variation in BC transport to the TP in summer. We employed a unified dynamical monsoon index to represent the strength of East Asian summer monsoon (EASM) and

South Asian summer monsoon (SASM). The index was proposed by Li and Zeng (2002) and it has been widely applied to quantify the impact of the Asian monsoon on air pollutants in Asia (Mao et al., 2017; Lu et al., 2018). Using this index, Han et al. (2019) found a close correlation between the EASM and ozone transport from foreign regions to East Asia. The calculation of the index was introduced in Li and Zeng (2002) and Han et al. (2019). The index is respectively termed as EASM index (EASMI) and SASM index (SASMI), when it is applied to represent the strength of EASM and SASM. A higher EASMI indicates a stronger EASM and a higher SASMI indicates a stronger SASM.

Figure 10a9a shows the spatial distribution of the correlation between BC transport to the TP and wind at 850 hPa at each of the grids in summer. As known from Figure 4b3b, central China is a dominant source region for the TP in summer, accounting for 63% of the imported BC from East Asia to the TP surface. In Figure 10a9a, BC from central China correlates significantly with the zonal wind at 850 hPa in central China (Figure 10a9a), with a regional mean correlation coefficient of -0.55 ($p < 0.05$). Westward winds (negative in the zonal component of wind vector) over central China favor BC transport from East Asia to the TP. Furthermore, the EASMI also correlates negatively with the zonal wind at 850 hPa over central China (Figure 10b9b). When the EASM is stronger, the zonal wind in the monsoon circulation weakens over this region (Yang et al., 2014; Han et al., 2019), suggesting that westward winds may occur more frequently or with higher speed. Therefore, BC transport to the TP from central China is enhanced (Figure 10e9c), as a significantly positive correlation is found between the strength of the EASM and BC transport from central China to the TP surface ($r = 0.49$, $p < 0.05$) and between the strength of the EASM and BC transport from central China to the eastern TP surface ($r = 0.48$, $p < 0.05$). This is further confirmed by the differences in BC transport to the TP surface in summers between summers with strong and weak EASM years (Figure 10d9d).

~~How~~Figure 10 shows how the SASM impacts the BC transport from South Asia over the TP surface in summer ~~is also examined (Figure 11).~~ Serving as a heat source in the Asian summer monsoon system, the TP promotes strong convection and modulates the meridional circulation (Xu et al., 2014). Driven by the meridional circulation, BC in South Asia can be transported northward and upward to the TP ~~(Figure 3b)~~. BC transport from northeastern South Asia to the TP accounts for 30% of the total BC transport from South Asia ~~(Figure 4b)~~. Interannually, BC transport from northeastern South Asia is significantly correlated with the meridional wind at 500 hPa ($r=0.65$, $p<0.05$, Figure ~~11a~~10a), which is also closely correlated to the strength of the SASM (Figure ~~11b~~10b). In strong SASM years, an anomalous cyclone locates over the northern South Asia at 500 hPa and correspondingly the meridional wind over the northeastern South Asia is increased (Figure ~~11d~~10d). This well explains why the interannual variation in BC transport from northeastern South Asia correlates positively with the strength of the SASM ($r=0.55$, $p<0.05$ for the TP, $r=0.56$, $p<0.05$ for the STP, Figure ~~11e~~10c). Among all source regions, the differences in BC transport from northeastern South Asia to the TP is largest in summers between ~~summers with~~ strong and weak EASM years (Figure ~~11d~~10d).

4.3 Influences of the Asian winter monsoon on the interannual variation in the transport of black carbon to the Tibetan Plateau from nonlocal region in winter

The Asian winter monsoon is a predominant climate feature in Asia and an important modulator of the distribution and transport of air pollutants (Mao et al., 2017; Zhu et al., 2017). However, the impact of the Asian winter monsoon on the interannual variation in BC transport to the TP is scanty studied. ~~In this section~~Here, we assess such impact with two climate indices. We measure the intensity of East Asian winter monsoon (EAWM) by an index defined by Jhun and Lee (2004). The EAWM index (EAWMI) represents the EAWM intensity by the meridional wind shear associated

with the jet stream in the upper troposphere. It can be calculated by the difference in the regional averaged zonal wind speed at 300 hPa between the areas 27.5-37.5°N, 110-170°E and 50-60°N, 80-140°E. Using the EAWMI, it is found that the EAWM is closely correlated with the interannual variation in pollution transport over East Asia (Q. Li et al., 2016; Han et al., 2019). Furthermore, the Siberian High is a key component of the EAWM system (Wu and Wang, 2002) and its strength can be described using an index defined by Wu and Wang (2002). This Siberian High index (SHI) can be calculated from the regional mean sea level pressure over the area of the Siberian High (40-60°N, 80-120°E). The EAWMI and SHI are highly correlated ($r=0.72$, $p<0.05$).

Figure 12.11 illustrates a connection between the EAWM and BC transport from East Asia to the TP surface. We mainly focused on BC transport from central China, as this area contributes to 54% of the total BC transport from East Asia to the TP (Figure 4.3d). BC transport from central China to the TP surface layer correlates significantly with the zonal wind at 850 hPa over central China ($r=-0.73$, $p<0.05$, Figure 12a.11a). The zonal wind over this region is also correlated with the strength of the EAWM ($r=-0.5$, $p<0.05$) and the strength of the Siberian High ($r=-0.65$, $p<0.05$) (Figure 12b.11b). A significant correlation ($r=0.59$, $p<0.05$) is found between the strength of the EAWM and BC transport from central China to the TP surface (Figure 12e.11c). When the EAWM is stronger, the more frequent or stronger westward winds can enhance BC transport from central China to the TP (Figure 12d.11d).

A connection between BC transport from South Asia to the TP surface and the Siberian High in winter is shown in Figure 13 ~~for winter~~. BC over the northern South Asia can be transported efficiently to the TP by the prevailing subtropical westerlies. Northern South Asia contributes to 70% of BC transported from South

Asia to the TP surface (Figure 4d3d). The contribution of northern South Asia to surface BC in the TP is significantly related ($r=0.72$, $p<0.05$) to the zonal wind at 500 hPa over the TP (Figure 13a12a). The westerlies over the TP are also correlated with the strength of Siberian High (Figure 13b12b). The elevated zonal wind in the middle troposphere over the TP in winters with strong Siberian High can enhance the BC transport from northern South Asia to the TP (Figure 13d12d). Significant correlations are found between the strength of the Siberian High and BC transport from northern South Asia to the TP surface ($r=0.66$, $p<0.05$) and between the strength of the Siberian High and BC transport from northern South Asia to the southern TP surface ($r=0.65$, $p<0.05$) (Figure 13e12c). In addition, the contribution of northern South Asia to surface BC in the western TP increases significantly with the meridional wind at 500 hPa over the western TP ($r=0.64$, $p<0.05$). The differences in BC transport from northern South Asia to the TP is largest in winter between ~~winters with~~ strong and weak SHI years (Figure 13d12d).

5 Discussion and conclusions

~~Using a global chemical transport model, GEOS-Chem, we characterized the variation in surface BC over the TP in 20 years from 1995 to 2014. By comparing with observations available in the literature, GEOS-Chem simulations show good performance in reproducing the spatial distribution, magnitude, and seasonal variation in surface BC over the TP. Applying an approach that combines the BC simulations from GEOS-Chem and backward trajectories from HYSPLIT, we identified the source regions for surface BC in the TP and demonstrated the influences of atmospheric transport and biomass burning on the interannual variation in surface BC over the TP. The major conclusions, along with discussion, are drawn as follows.~~

~~Based on the 20-year mean, surface BC in the TP is mainly influenced by two source regions: East Asia and South Asia. The influence of East Asia is dominant in~~

summer while the influence of South Asia is dominant in winter. We divided the TP into five subregions: eastern, southern, western, northern, and central TP. Surface BC concentrations are higher in the southern and eastern TP than in the other subregions. Surface BC in the southern and eastern TP comes mainly from South Asia and East Asia, respectively. Over the western TP, surface BC comes mainly from South Asia. Over the northern TP, the dominant BC source region is South Asia in winter and East Asia in the other seasons. Over the central TP, the dominant BC source region is East Asia in summer and South Asia in the other seasons.

Interannually, from 1995 to 2014, biomass burning can explain over 75% of the variation in springtime surface BC concentrations over the TP if biomass burning and meteorology are both considered in GEOS-Chem simulations. Indeed, springtime surface BC in the TP is significantly correlated to the total number of fire counts over the Indo-Gangetic Plain in South Asia ($r=0.76$, $p<0.05$), according to ATSR satellite data. In the spring of 1999, the extremely strong biomass burning in South Asia largely elevated surface BC concentrations ($0.15 \mu\text{g m}^{-3}$ or 31% relative to the climatology) over the TP. We noticed that the strong biomass burning in South Asia in the winter of 1998 also enhanced BC concentrations over the TP.

The interannual variation in surface BC over the TP are greatly influenced by meteorology. Specifically, the Asian monsoon system alters the long-range transport of BC to the TP by modulating the atmospheric circulation. In summer, when the EASM is stronger, the more frequent or stronger westward wind in the lower troposphere can enhance BC transport from central China to the TP. When the SASM is stronger, the increased meridional wind over the northeastern South Asia in the middle troposphere can enhance BC transport from northeastern South Asia to the TP. In winter, when the EAWM is stronger, the reduced zonal wind in the lower troposphere tends to increase BC transport from central China to the TP. A stronger

~~Siberian High can enhance the zonal wind in the middle troposphere over the TP and consequently increases BC transport from northern South Asia to the TP.~~

The findings in this study provide an enhanced understanding of the long-range transport of BC to the TP. We comprehensively assessed the BC transport from worldwide source regions to the TP. Our results reveal the source regions of surface BC over the entire TP in the four seasons, which was investigated by limited studies (Lu et al., 2012; Zhang et al., 2015). The influences of South Asia and East Asia on the TP were noticed by previous studies. Most of them were focused on limited locations (Cao et al., 2011; Engling et al., 2011; Chen et al., 2018) or in one or few seasons (Zhao et al., 2017; Wang et al., 2018). Here, we further quantified the influence of South Asia and East Asia over the entire TP in the four seasons, in terms of both fractional contribution (Figure 6) and affected areas in the TP (Figure 4). Moreover, we identified three key areas within South Asia and East Asia and found that the contribution of BC from there to surface BC in the TP is highest among South Asia and East Asia (Figure 3).

The estimate of the fractional contributions from various source regions to surface BC in the TP from this study is generally comparable with that from literature (Lu et al., 2012; Zhang et al., 2015; Yang et al., 2018). (1) The total contribution of South Asia and East Asia to surface BC in the TP is estimated to be 77%, between 69% by Zhang et al. (2015) and 84% by Lu et al. (2012). (2) The contribution of South Asia is stronger in winter and weaker in summer, which was also suggested by Lu et al. (2012), Zhang et al. (2015), and Yang et al. (2018). Yang et al. (2018) modeled that the contribution of South Asia is 61% in non-monsoon season (October-April) and 19% in monsoon season (May-September). (3) The estimate of local contribution is ~10%, comparable with that by Zhang et al. (2015), indicating the dominant role of BC transport from nonlocal regions. Nevertheless, there are some

disagreements between this and previous studies in various extents. A noticeable disagreement is that this study estimates that the annual mean contribution of East Asia is approximately 35% (Figure 6, also see Figure S5), while the estimates by Lu et al. (2012) and Zhang et al. (2015) are respectively 17% and 19%. The discrepancy may be associated with the differences in region definitions and the estimation models.

Biomass burning is an important source ~~of atmospheric~~ BC ~~in aerosols over~~ the TP (Zhang et al., 2015). It was observed that BC emissions from biomass burning in South Asia could be transported to the TP by the atmospheric circulation (Cong et al., 2015), and thus resulted in high BC episodes in the southern TP (Engling et al., 2011; Putero et al., 2014). ~~Only limited~~Limited numerical studies ~~explored~~ ~~connections between~~assessed the impact of biomass burning ~~and on~~ surface BC in the TP over a long-term period (Mao and Liao, 2016). Here, we demonstrated that biomass burning is an important driver of the interannual variation in surface BC over the TP in spring: (Figure 7). In particular, we found that there were extremely strong fire activities over the Indo-Gangetic Plain, central India, and Southeast Asia from 1998 winter to 1999 spring that largely enhanced surface BC concentrations over the entire TP: (Figures 7 and 8). This extreme anomaly in fire activities and associated influence on BC over the TP may have not been fully documented.

The Asian monsoon can influence the atmospheric circulation over the TP and its surroundings (Xu et al., 2014; Han et al., 2019). We found that the Asian monsoon system can significantly modulate the interannual variation in BC transport from South Asia and East Asia to the TP: (Figures 9-12). We revealed that the EASM can modulate the westward transport of BC from central China to the TP in ~~Asian monsoon can influence the atmospheric circulation over the TP and its surroundings (Xu et al., 2014; Han et al., 2019).~~ ~~For~~ summer, (Figure 9), while previous studies

~~mainly~~mostly focused on the transport pathway build by the SASM, ~~which can meridionally transport BC from South Asia to the TP~~ (Zhao et al., 2017; Kang et al., 2019). ~~In this study, we further revealed that the EASM can modulate the westward transport of BC from central China to the TP.~~ 2019). In winter, ~~the Asian monsoon system also significantly~~this study shows significant influences of the EAWM on the BC transport from northern South Asia and central China to the TP. (Figures 11 and 12). These results can also shed some light on the transport mechanisms of other atmospheric species to the TP, such as water vapour.

~~Numerical~~Regarding methodology, numerical simulations, including adjoint (Kopacz et al., 2011), backward-trajectory (Ming et al., 2009, 2010; Lu et al., 2012), adjoint (Kopacz et al., 2011), and tagging tracer (Zhang et al., 2015), and emission perturbation (Yang et al., 2018) simulations, have been used to identify BC transport from sources to the TP. Each of the methods has its advantages and limitations (Lu et al., 2012; Zhang et al., 2015). Modifying an approach proposed by Lu et al. (2012), we developed an efficient and stable method which shows strong performance in revealing the factional contribution of BC transport from different source regions to the TP by season and year. ~~This method can explicitly show~~We also run a set of emission perturbation simulations using GEOS-Chem to compare with the spatial distribution~~outcomes~~ of the trajectory method being developed in this study. The emission perturbation method can estimate the contribution of ~~different~~a source ~~regions~~region to surface BC over the TP. It is ~~feasible for users of a chemical transport model to estimate BC transport from different source regions to concentrations in a receptor region if adjoint~~(i.e. the TP in this study) by turning off BC emissions in that source region. The detailed description on our simulation and tagged modes of the model~~its outcomes~~ are ~~unavailable to them.~~provided in Supplement (Figure S5).

Comparing the results between the backward-trajectory and the emission

perturbation methods (Figure 6 vs. Figure S5), we can observe the following: (1) Both methods show that South Asia and East Asia are two dominant source regions for surface BC in the TP. The total contributions of South Asia and East Asia estimated by the two methods are comparable, which are respectively 77% (Figure 6a) and 82% (Figure S5a). (2) The two methods suggest similar seasonality for the contributions of South Asia and East Asia. The contribution of South Asia to surface BC in the TP is stronger in winter and spring, while the contribution of East Asia is stronger in summer and autumn. (3) The influences of BC transport on different TP subregions estimated by the two methods are generally consistent. For example, both methods show that over 70% of surface BC in the eastern TP comes from East Asia, and over 70% of surface BC in the southern TP comes from South Asia. Therefore, both methods show comparable results, although there are some discrepancies in various details.

Overall, the backward-trajectory method developed in this study can reasonably quantify the relative contributions of different source regions to surface BC in the TP. This method has following advantages. (1) The transport pathway of BC can be visibly expressed (Figures 2 and 3). (2) The spatial distribution of the contribution of source regions can be showed explicitly (Figures 3 and 4). (3) It is feasible for users of a chemical transport model to investigate the source-receptor relationships if adjoint and tagged modes are unavailable to them. Note that this method assumes a fixed lifetime of atmospheric BC (i.e. 7 days in this study), which might lead to some uncertainties. The emission perturbation method does not require this assumption. It is reliable and straightforward. However, the emission perturbation method cannot provide the spatial information showed in Figures 2-4. It can only provide an overall assessment for the total contribution from each of the source regions defined, which are often limited because of constraints of computation expenses.

This study is subject to some limitations. Numerical simulations have advantages of covering large areas over the entire TP and long periods, such as 20 years in this study. ~~GEOS-Chem has been used in multiple studies on BC aerosols over the TP (Kopacz et al., 2011; Lu et al., 2012; He et al., 2014b; Mao and Liao, 2016).~~ However, according to this and earlier studies, there are discrepancies between observations and simulations from GEOS-Chem. He et al. (2014b) suggested that BC aerosols over the TP may be underestimated by GEOS-Chem. ~~A smaller~~ Simulations in this study have a weaker seasonal variation in BC concentrations ~~from simulations~~ than ~~from~~ observations ~~is also revealed in this study from previous studies (Figure S1).~~ All of these imply uncertainty in simulating the absolute BC concentrations over the TP by GEOS-Chem. ~~Additionally~~ Furthermore, the simulations are with a resolution of 2° latitude by 2° longitude. Such a resolution may not fully capture processes in the sub-grid scale, such as the mountain-valley wind (Cong et al., 2015). Using a regional model, Zhang et al. (2020) demonstrated that compared with simulations with lower resolution, simulations with higher resolution can better resolve the effects of topography and consequently yield stronger transport flux of BC from South Asia to the TP. Therefore, our results may somewhat underestimate the contribution of South Asia because of the model resolution. Using regional models at higher resolutions in the future can better describe the terrain effect in the TP.

In addition, anthropogenic BC emission inventories used in GEOS-Chem simulations may add uncertainties in our estimates. Figure S6 compares the anthropogenic emissions in 2000 used in this study and those in 2010 from the Task Force on Hemispheric Transport of Air Pollution Phase 2 (TF HTAP2). It shows that anthropogenic BC emissions in most areas of South Asia have substantially increased from 2000 to 2010 (Figure S6). On regional mean, anthropogenic BC emissions in East Asia have also increased from 2000 to 2010 (Figure S6). Inside East Asia, anthropogenic BC emissions in most regions of eastern China have increased, while

decreased in Korea and Japan from 2000 to 2010 (Figure S6). Therefore, our results may underestimate the BC transport from South Asia and East Asia to the TP in 2010. After 2010, anthropogenic BC emissions have changed as well (Zheng et al., 2018). Zheng et al. (2018) reported that anthropogenic BC emissions in China have reduced 27% from 2010 to 2017. BC emissions from biomass burning used in this study cover 1997-2011 and fire emissions in 2011 are used for simulation years after 2011. However, fire activities in South Asia have strong interannual variations and have been significantly increased from 2001 to 2016 (Earl et al., 2018). Therefore, fire emissions used here may lead to biases in the BC simulations after 2011. This study is focused on natural drivers (biomass burning and meteorology) that are connective to the interannual variation in BC over the TP. The impact of interannual anthropogenic emissions ~~warrant~~warrants further studies.

6 Conclusions

Using a global chemical transport model, GEOS-Chem, we characterized the variation in surface BC over the TP in 20 years from 1995 to 2014. Applying an approach that combines the BC simulations from GEOS-Chem and backward trajectories from HYSPLIT, we identified the source regions for surface BC in the TP and demonstrated the influences of atmospheric transport and biomass burning on the interannual variation in surface BC over the TP. The major conclusions are drawn as follows.

Based on the 20-year mean, surface BC concentrations are higher in the southern and eastern TP than in the other TP subregions. Surface BC in the TP is mainly influenced by two source regions: East Asia and South Asia. The two regions totally contribute 77% surface BC in the TP. The influence of East Asia is dominant in summer while the influence of South Asia is dominant in winter. By subregion, surface BC in the southern and eastern TP comes mainly from South Asia and East

Asia, respectively. Over the western and central TP, surface BC comes mainly from South Asia. Over the northern TP, the most important source region is central Asia in spring and autumn, East Asia in summer, and South Asia in winter.

Interannually, from 1995 to 2014, biomass burning can explain over 75% of the variation in springtime surface BC concentrations over the TP if biomass burning and meteorology are both considered in GEOS-Chem simulations. Springtime surface BC in the TP is significantly correlated to the total number of fire counts from ATSR satellite data at 1 km resolution over the Indo-Gangetic Plain in South Asia ($r=0.76$, $p<0.05$). In the spring of 1999, the extremely strong biomass burning in South Asia largely elevated surface BC concentrations ($0.08 \mu\text{g m}^{-3}$ or 31% relative to the climatology) over the TP. The strong biomass burning in South Asia in the winter of 1998 also enhanced BC concentrations over the TP.

The interannual variation in surface BC over the TP are greatly influenced by meteorology. Specifically, the Asian monsoon system alters the long-range transport of BC to the TP by modulating the atmospheric circulation. In summer, when the EASM is stronger, the more frequent or stronger westward wind in the lower troposphere can enhance BC transport from central China to the TP. When the SASM is stronger, the increased meridional wind over the northeastern South Asia in the middle troposphere can enhance BC transport from northeastern South Asia to the TP. In winter, when the EAWM is stronger, the reduced zonal wind in the lower troposphere tends to increase BC transport from central China to the TP. In winters with a stronger Siberian High, the enhanced zonal wind in the middle troposphere over the TP tends to carry more BC from northern South Asia to the TP.

Data availability

The GEOE-Chem model is publicly available at <http://geos-chem.org>. The HYSPLIT

model can be acquired from http://www.arl.noaa.gov/HYSPLIT_info.php. The meteorological and fire data were download from <https://www.esrl.noaa.gov/psd/data/gridded/data.ncep.reanalysis.surface.html> and <https://earth.esa.int/web/guest/home>, respectively.

Author contributions

JL, YW, and HH designed the research. HH and YW performed the study under supervision of JL and TZ. HH, YW, YL, HC, ~~and~~ YZ analyzed the data. HH, YW, and JL wrote the paper. ~~TZ, with valuable input from~~ BZ, HW, HL, QW, SL, TW, MX, and ML ~~contributed insight and comments~~.

Competing interests

The authors declare no conflict of interest.

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