In Situ Observations of Light-Absorbing Carbonaceous Aerosols at Himalaya: Analysis of the South Asian Sources and Trans-Himalayan Valleys Transport Pathways

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Abstract

The mountain-valleys along the south margin of the Himalaya-Tibetan Plateau (HTP), referred to as trans-Himalayan valleys, are transport pathways of atmospheric pollutants from the South Asia lifting to the HTP based on atmospheric models and satellite detections. However, few field studies can be conducted to confirm the aerosol transport along these valleys due to the harsh environment and limited power supply. Individual particle collection instruments offer the unique advantage of collecting samples and tracing particle sources in remote areas due to their portability and efficiency. In this study, transmission electron microscopy was used to study individual particles collected in two trans-Himalayan valleys. We found that 49–70% of thousands of particles were light-absorbing carbonaceous aerosols including soot-bearing and tarball-bearing particles in December of 2017. The dominant sizes of soot-bearing and tarball-bearing particles were 151–370 and 198–255 nm, respectively. The WRF-Chem model and wind field analyses show that the intense regional air pollution with high mass concentrations of light-absorbing carbonaceous aerosol occurred in the South Asia and southwest wind normally prevailed along the trans-Himalayan valleys in the afternoon of each day during wintertime. Backward trajectory and CALIPSO data further confirm that these light-absorbing carbonaceous aerosols from the South Asia were transported to the southern HTP. These carbonaceous aerosols exhibited significantly internally mixing with secondary sulfates. Here individual particle analysis coupled with the WRF-Chem model, the meteorological observation data, and satellite data confirm the trans-Himalayan valleys as the direct channels for abundant carbonaceous aerosols lifting from the South Asia to the HTP.

1. Introduction

The Himalaya-Tibetan Plateau (HTP) covers millions of square kilometers with high altitude and is usually known as the “third pole” on Earth (Yao et al., 2012). Because of its complex topography and numerous glaciers, the HTP exerts an important influence on the Asian monsoon and the global climatic change (G. X. Wu et al., 2007). The HTP is far away from the intensive anthropogenic activities and is considered as one of the cleanest but the most vulnerable regions on Earth (Yao et al., 2012). The south margin of the HTP is crisscrossed with hundreds of river valleys within high mountain ranges. The anthropogenic atmospheric pollutants like carbonaceous aerosols either from the faraway South Asia (Kopacz et al., 2011; X. H. Zhang et al., 2018) or from the local residential emissions in the valleys (Bonasoni et al., 2010; Regmi et al., 2019; Yuan et al., 2019) can be uplifted into the HTP along the valleys (Cong et al., 2015; Dhungel et al., 2018; Hindman & Upadhyay, 2002; Marinoni et al., 2010). These valleys act as transport channels for atmospheric pollutants and have been called the “leaking wall” (Cao et al., 2010), the “direct channel” (Bonasoni et al., 2010), or the “trans-Himalayan” valleys (Dhungel et al., 2018). The light-absorbing carbonaceous aerosols from the South Asia were observed in the snowpack in the HTP glaciers (Dong et al., 2018; C. L. Li et al., 2016) and contributed to the glacierretreating (Sarangi et al., 2019; B. Q. Xu et al., 2009; Y. L. Zhang et al., 2017).
In the past decade, aerosol measurements in the HTP were mostly conducted at observation stations far from the trans-Himalayan valleys (Kang et al., 2019; B. Liu et al., 2017; Niu et al., 2018; J. F. Wang et al., 2017; G. M. Wu et al., 2018). Various samplers and online instruments could be run steadily at the observation stations with continuous electric power supply. However, there are still only a few of the remote observation stations in the HTP that are suitable for long-term measurements. The valuable data from these remote observation stations could not directly reflect the atmospheric pollutant transport through the trans-Himalayan valleys. The desolate environments with harsh weather conditions and power shortages do not allow people to deploy long-term field experiments in most trans-Himalayan valleys. To date, only a few bulk measurements were conducted in a high valley at the foot of Himalayas, mostly focusing on the aerosol optical properties (Cristofanelli et al., 2014; Decesari et al., 2010; Gobbi et al., 2010; Kirillova et al., 2016; Marq et al., 2010; Nair et al., 2013). More direct aerosol data in more valleys are needed to show how the aerosol particles specifically are lifted and transported along the trans-Himalayan valleys.

Some studies show that individual particle collection has unique advantages in collecting aerosol samples and tracing particle sources, which is superior to online and bulk measurements in the clean atmosphere with low PM$_{2.5}$ concentration ($<10$ μg/m$^3$) at the remote region of the HTP (W. J. Li et al., 2015; Yuan et al., 2019). Individual particles only need to be collected on the small substrates for electron microscopy analysis. Various portable and battery-powered individual particle samplers have been successfully used to collect aerosol particles in diverse harsh environments such as a valley site of the southeastern Tibetan (Yuan et al., 2019), a mountain site (L. Liu et al., 2018), an Arctic site (Yu et al., 2019), an Amazon forest site (China et al., 2018), a marine observation (W. J. Li et al., 2017), and even in-flight observations (Adachi et al., 2019; Pósfai et al., 2004). All of these sampling methods can collect adequate particles for laboratory analysis with a short period from 10 to 60 min. Moreover, individual particle method has successfully observed the anthropogenic aerosols transported to the Arctic (Chi et al., 2015), East China Sea (W. J. Li et al., 2017), and northern Tibet (W. J. Li et al., 2015). The mixing states and morphologies of fresh aerosols from combustion sources can be clearly identified by the transmission electron microscopy (TEM). Therefore, individual particle analysis is a powerful and effective method for capturing the physicochemical properties of fine particles during the mountain-valley transport in the remote trans-Himalayan valleys.
frequent anthropogenic burning activities, such as daily wood and straw burning for cooking and heating and household waste burning in weekend. Because of the sparse population and harsh environment, the observation conditions at the Jilong site are hampered by the discontinuous electricity supply during the sampling period. In this study, we used two individual battery-powered particle samplers to collect aerosol particles on substrates at the two sites.

Figures 1b and 1c clearly show the topography of the typical trans-Himalayan valleys with Jilong site and Nepal site locating in the high altitude. Three open channels in the south edge extend northward to the Tibetan Plateau (Figures 1b and 1c). A large city (Kathmandu, Nepal) and several small villages are present in lower altitudes within a 200 km north-south region and 2 km lower down (Figures 1b–1f). Kathmandu, with its population of ~5 million people and its bowl-shape at the foothills of Himalayas, is a heavily polluted regions in the South Asia (Panday & Prinn, 2009). Predictably, the anthropogenic pollutants emitted from Kathmandu and these villages would be lifted and transported to the higher altitude HTP area under favorable meteorological conditions.

2.2. Individual Particle Collection and Electron Microscopic Analysis

The individual particle samples were collected by two portable samplers (DKL-2, Genstar Electronic Technology, China). One 0.3 mm jet nozzle and one 0.5 mm jet nozzle were connected as the sampler inlet to collect fine and coarse particles, respectively (W. J. Li et al., 2011). The sample filters were the copper grids coated by carbon film (carbon type-B, 300-mesh copper; Tianld Co., China). Seven individual particle samples in total were successfully collected at Jilong site at the following (Beijing) times daily: 10:30, 16:30, 19:00, 21:50 on 13 December and 10:00, 16:00, 18:30 on 14 December 2017. Two individual particle samples in total were successfully collected at Nepal site at 17:00 (Beijing time) on 8 December 2017. The sampling durations were 30 min at Jilong site and 2–5 min at Nepal site. After the sample collection, the TEM samples were immediately stored in the plastic capsules (100 mesh; Tianld Co., China) and then these capsules were placed in a desiccator under low relative humidity (RH = 20–25%).
The individual particle samples were analyzed by a TEM (JEOL JEM-2100, Japan) at 200 kV and the details can be found in Yuan et al. (2019). The chemical elements of individual particles (≥C, exclude from Cu) were detected by an energy-dispersive X-ray spectrometer (EDS, INCA X-MaxN 80T, Oxford Instruments, United Kingdom). A total of 3,569 individual particles were analyzed to obtain their morphologies, mixing states, sizes, and compositions. The equivalent circle diameter of each analyzed particle was measured by the RADIUS software (EMSIM GmbH, Germany) to represent the particle diameter.

2.3. Particle Mass Concentration Analysis
A portable aerosol dust monitor (Grimm Aerosol Technik GmbH & Co. KG, Germany) was used to measure PM$_{2.5}$ concentrations during the particle collection period at Jilong site (supporting information Figure S1). PM$_{2.5}$ concentrations in the major cities (such as Kanpur, Patna, and Arga) of the northern India on 13 and 14 December were obtained from the Central Control Room for Air Quality Management of India (https://app.cpecbcr.com/ccr/#/caaqm-dashboard-all/caaqm-landing/data). PM$_{2.5}$ concentrations of Kathmandu on 13 and 14 December were obtained from the U.S. Embassy (http://aqicn.org/city/nepal/kathmandu/us-embassy/). We further obtained black carbon (BC) and organic carbon (OC) column concentrations from the Giovanni online data system (https://giovanni.gsfc.nasa.gov/giovanni/).

2.4. Meteorological Data Analysis From Observations and Model Simulations
Vertical wind data at the major meteorological observation stations over part of the South Asia and the southern HTP were gained from the China Meteorological Agency and processed by Meteorological Information Combine Analysis and Process System (MICAPS, China). RH and temperature at the sampling site were measured by an automated weather instrument (Kestrel 5500, United States). There was no precipitation during observation period at the two sites.

The WRF-Chem model (version 4.1) has been proved to be an effective transport model with a combination of meteorological and chemical factors (Grell et al., 2005; M. Li et al., 2019) and is widely implied for simulating the carbonaceous aerosols emission and transport in the South Asia and the HTP (Alvarado et al., 2018; Gogoi et al., 2017). In this study, the WRF-Chem simulated the surface mass concentrations of BC, OC, and PM$_{2.5}$ and the regional wind fields in the South Asia and the southern HTP from 08:00 (Beijing time) on 6 December to 08:00 (Beijing time) on 24 December 2017. The modeling domain covers the northern part of South Asia and the southern HTP (80°E to 88°E, 21°N to 31°N) with 41 × 50 grids and the horizontal spatial resolution is 25 × 25 km. Twenty-four vertical layers are setup from the surface to 50 hPa. The meteorological data for model simulation (1 hr, 1° × 1°) can be obtained from the National Centers for Environmental Prediction (NCEP) Research Data Archive (https://rda.ucar.edu/datasets/ds083.2/). The Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) is used to investigate the aerosol chemistry (Zaveri et al., 2008) with nine aerosol species being specifically treated, such as BC, OC, sulfate, nitrate, ammonium, calcium carbonate, sodium chloride, some unspecified inorganic aerosols, and water. The anthropogenic emission inventory for Asia (MIX) in 2010 with 0.25° × 0.25° resolution (M. Li et al., 2017) is used, including anthropogenic emissions from industry, power generation, agriculture, transportation, and residential sectors. The vertical cross-section along the major wind direction is simulated to understand the vertical variations of atmospheric pollutants and wind fields.

The Hybrid Single Particle Lagrange Integrated Trajectory (HYSPLIT) model is used to calculate the backward trajectories for investigating the air mass paths and the possible particle sources. The 48-hr backward trajectory frequency was calculated with the height of ending point being 50 m above ground level and grid resolution was set up at 0.5° (https://ready.arl.noaa.gov/hypub-bin/trajtype.pl?runtype=archive).

2.5. Satellite Data Analysis
The CALIPSO lidar detection provides a direct measurement of the optical properties and vertical structures of aerosols at both regional and global scales (Winker et al., 2009). Two types of images, including 532 nm total attenuated backscatter coefficient and aerosol subtype, from the CALIPSO website (http://www-calipso.larc.nasa.gov/) were used to investigate the vertical distribution of atmospheric pollutants over the South Asia and the HTP. The active fire spots over the South Asia were obtained from the Fire Information for Resource Management System (FIRMS) (https://firms.modaps.eosdis.nasa.gov).
3. Results and Discussion

3.1. Distributions and Concentrations of Aerosol Pollutants

We analyzed thousands of particles and classified these particles into five major types according to their different morphology and composition: soot, tarball, soot-SK, tarball-SK, and SK-rich. Soot and tarballs are the most abundant carbonaceous particles in all the samples. Soot is a fine light-absorbing carbonaceous aerosol and is mainly emitted from fuel burning or biomass burning (Bond et al., 2013; L. Liu et al., 2017; Yu X. Zhang et al., 2018). Many studies have shown that soot particles exhibit a unique chain-like aggregates morphology, and they can be clearly identified through the electron microscopies (Adachi et al., 2018; China et al., 2015; Y. Y. Wang et al., 2017). The TEM image shown in Figure 2a clearly displays one chain-like soot aggregates.

Tarball is a particular and abundant particle type of brown carbon with a spherical shape, a small size, and a certain viscosity (Adachi et al., 2019; Pósfai et al., 2004). Tarball is mainly emitted from fossil fuel burning or biomass burning and consist of major C, O, N with minor S, K (Adachi et al., 2019; Pósfai et al., 2004). Due to their refractory properties and special morphologies, tarballs can be clearly observed via electron microscopy (Adachi et al., 2019; Hand et al., 2005; Hoffer et al., 2016). In this study, the spherical fine tarball particle is clearly displayed from the TEM image (Figure 2b). Besides the abundant soot and tarballs in the fine particles, we also found many potassium sulfate particles (SK-rich) (Figure 2c). SK-rich particles easily appear bubbles under the strong electron beam and the major chemical elements of SK-rich particles are S, C, O, and K. The SK-rich particles are taken as the secondary inorganic aerosols and mainly contain (NH₄)₂SO₄, K₂SO₄, and NH₄NO₃ (W. J. Li et al., 2016), which are frequently internally mixed with secondary organic aerosols (Figure S2a). Abundant soot and tarballs were mixed with SK-rich particles during the transport, which are defined as soot-SK and tarball-SK particles (Figures 2d and 2e). EDS spectra show that the major chemical elements of soot and tarball are C, O, and Si (Figures 2f and 2g), while the major chemical elements of soot-SK and tarball-SK particles are C, O, Si, S, and K (Figures 2h and 2i). To perform a detailed study of carbonaceous aerosols, we further considered soot/SK, tarball/SK to be soot-bearing particles and tarball/tarball-SK to be tarball-bearing particles. Besides the soot-bearing, tarball-bearing, and SK-rich particles, we also found extremely small number of mineral and Fe particles (<1% of total particles, Figures S2b and S2c). In this study, we excluded these trace particles from the statistics.

Soot-bearing and tarball-bearing particles were the major particles in the ambient air, accounting for 71% of the total particle counts at remote Jilong site and 49% at polluted Nepal site, respectively. The number fractions of the total carbonaceous aerosols including soot-bearing and tarball-bearing particles consistently stayed at fairly high levels at each sampling time (52–98%) at Jilong site. The soot-bearing and
tarball-bearing particles mostly resided in the fine mode and 80% of them were smaller than 500 nm (Figure 3a). Size distributions of carbonaceous aerosols (Figure 3b) showed that soot and tarball had a peak at 151 and 198 nm, respectively, which are consistent with other observations (Adachi et al., 2019; Moffet & Prather, 2009; Pósfai et al., 2004) and much smaller than the peaks at 370 nm for soot-SK and 255 nm for tarball-SK in this study. The size differences suggest that SK-rich internally mixed with soot and tarball during the transport shifted the carbonaceous aerosols into the larger sizes.

Soot and tarball mainly originate from fossil fuel burning and biomass burning (Adachi et al., 2019; Bond et al., 2013; Laskin et al., 2015). Satellite and ground-based data showed that fossil fuel burning and biomass burning are the dominant contributors to the high concentrations of BC and OC in the South Asia during wintertime (Rajput et al., 2014; Sarkar et al., 2018). Indeed, MODIS data show that several dense active fire spots occurred in the South Asia during the sampling period (Figures 1a, S3a, and S3b). We can identify a haze layer over the South Asia from the MODIS images on 13 and 14 December 2017 (Figures S3a and S3b). Mass concentrations of ground-level PM$_{2.5}$ show the high values of 100–200 μg/m$^3$ in major cities in the South Asia during the sampling days (Figures S3a and S3b). Figures S3c and S3d further show that BC and OC column mass density were highest in the northern India. Satellite data and ground-based aerosol measurements have confirmed that the increase of BC mass concentrations in the northern India during wintertime is mainly related to the dense crop residue and biomass burning (Sarkar et al., 2018). Because of a lack of electrical power, biomass burning is the major energy sources for household heating and cooking in Kathmandu in winter (Chen et al., 2015), contributing to the high concentrations of BC and OC (Wan et al., 2019). These emission sources coupled with vehicular emissions in the cities significantly contributed to the haze formation in the South Asia.

Interestingly, we found that the total proportion (71%) of light-absorbing carbonaceous aerosols at Jilong site was much higher than those at background and remote sites in the HTP, such as Nam Co (14%), Renlongba (26%), Lulang (64%), Shangri-La (25%), Menyuan (12%), Qomolangma (11%), Nielamu (~8%), and even higher than at the town and urban sites, such as Dhunche (~10%) and Kathmandu (~8%) in Nepal, Lhasa (9.8%) in the central TP, and Arga (38%) in India (Table 1). One possible reason is that large amounts of carbonaceous aerosols were emitted in the upwind rural and urban areas in the valley channels. Indeed, we detected large number of soot-bearing and tarball-bearing particles (49%) in two samples collected at polluted Nepal site in a traditional village in a trans-Himalayan valley. Based on our observations, the local residents often use various biomass for daily cooking and heating and also burn household waste at a garbage disposal station out of the village in weekend. One recent study reported the intense emission sources for the high concentrations of carbonaceous aerosols (monthly average OC in TSP: 62.8 μg/m$^3$, monthly average EC in TSP: 11 μg/m$^3$) in the Kathmandu Valley in winter (Wan et al., 2019). In addition, the narrow channels are subject to being receptor areas for particle number concentrations of carbonaceous aerosols from wide and flat areas like the South Asia. Therefore, the intense anthropogenic emissions from the valley residents and the South Asia were the major contributors to the high number fractions of soot-bearing and tarball-bearing particles detected in this study under the favorable meteorological conditions.

![Figure 3](https://example.com/figure3.png)

**Figure 3.** (a) Relative abundance of soot, tarball, soot-SK, tarball-SK, and SK-rich particles at different size ranges during the sampling period. (b) Size distributions of soot, tarball, soot-SK, and tarball-SK particles.
| Site        | Location          | Setting | Altitude (m a.s.l.) | PM (μg/m³) | Particle count | Externally mixed (%) | Internally mixed (%) | Externally mixed (%) | Internally mixed (%) | Date       | Reference          |
|------------|-------------------|---------|---------------------|------------|----------------|----------------------|----------------------|----------------------|----------------------|------------|--------------------|
| Jilong     | Central Himalaya  | Remote  | 2,830               | 2.6 (PM$_{2.5}$) | 3,447          | 17                   | 34                   | 13                   | 7                    | 2017.12    | This study          |
| Nepal      | Northern Nepal    | Village | 2,720               | -          | 122            | 5                    | 24                   | 14                   | 6                    | 2017.12    |                    |
| Nam Co     | Central TP        | Remote  | 4,730               | 6.7 (TSP)  | 1,800          | 8                    | -                    | 6                    | -                    | 2006 winter | Cong et al. (2009) |
| Renlongba  | Eastern Himalaya  | Remote  | 5,000               | -          | 413            | 25                   | -                    | 1                    | -                    | 2010.04    | Hu et al. (2018)   |
| Lulang     | Southeast margin TP | Remote  | 3,300               | -          | 4,059          | 64                   | -                    | -                    | -                    | 2016.05    | Yuan et al. (2019) |
| Shangri-La | Eastern margin TP | Background | 3,580               | -          | 397            | 8                    | -                    | -                    | -                    | 2016.07–08 | Fan et al. (2016)  |
| Nenjuan    | Northern margin TP | Background | 3,295               | 17 (PM$_{2.5}$) | 4,253          | 25–38                | -                    | -                    | -                    | 2013.09–10 | W. J. Li et al. (2015) |
| Qomolangma | Central Himalaya  | Background | 4,276               | 2–6 (TSP)  | 900            | 8                    | -                    | 3                    | -                    | 2005.05–06 | Cong et al. (2010) |
| Nielamu    | Central Himalaya  | Town    | 4,166               | -          | >1,200         | -                    | -                    | -                    | -                    | 2014 winter | Dong et al. (2017) |
| Dhunche    | Central Himalaya  | Town    | 2,051               | -          | >1,200         | -                    | -                    | -                    | -                    | 2014 winter | Dong et al. (2017) |
| Kathmandu  | Central Nepal     | Urban   | 1,314               | -          | >1,200         | -                    | -                    | -                    | -                    | 2014 winter | Dong et al. (2017) |
| Lhasa      | Central TP        | Urban   | 3,650               | ~25.7      | 408            | 9.8                  | -                    | -                    | -                    | 2013.02    | Duo et al. (2015)  |
| Nyingchi   | Eastern Himalaya  | Urban   | 2,900               | ~200       | 406            | 67                   | 6                    | -                    | -                    | 2010.04    | Hu et al. (2018)   |
| Agra       | India             | Urban   | ~300                | 90 (PM$_{2.5}$) | -                | 38                   | -                    | -                    | -                    | 2010.04–06 | Pipal et al. (2011) |

Note: Date is presented in a “year.month” format.
3.2. Transports of Carbonaceous Aerosols Along the Trans-Himalayan Valley

The WRF-Chem simulates the horizontal distribution of BC, OC, and PM$_{2.5}$ mass concentrations and shows an obvious spatial gradient distribution with increasing from the HTP to the South Asia and one high emission zone in the central Indo-Gangetic Plain (Figure 4). The simulated spatial distributions of carbonaceous aerosols exhibit a clear spatial gradient from the HTP to the South Asia. The horizontal distributions of BC, OC, and PM$_{2.5}$ concentrations are shown in Figure 4(a)–(c). The color scales for BC, OC, and PM$_{2.5}$ are shown in Figure 4(d)–(f). The red circle dot represents the Jilong site. The thick red arrows (a–c) represent the prevailing southwest winds along the valleys from the South Asia to the Himalayas during 12:00–19:00 (Beijing time). The thick white arrows (d–f) represent the opposite prevailing northeast winds along the valleys from the Himalayas to the South Asia during 20:00–11:00 (next day, Beijing time). The typical hourly simulated data of each day are listed in Figures S4–S6 and all the hourly simulated data of each day are available from the Figshare database (https://doi.org/10.6084/m9.figshare.12504947).
aerosols are consistent with the MODIS data (Figures S3c and S3d). Moreover, some previous studies used the WRF-Chem model to reveal large anthropogenic emissions in the central Indo-Gangetic Plain in wintertime of every year (R. Xu et al., 2018; Yang et al., 2020). Figure 4 shows that southwesterly and westerly synoptic-scale air masses prevailed during the sampling period, covering the northern South Asia and the southern HTP. Bonasoni et al. (2010) and R. Xu et al. (2018) reported similar meteorological fields in winter season. As shown in Figures S4–S6, we noticed two opposite wind fields in each day in the south of Himalayas: a continuous southwest wind along the trans-Himalayan valley directions during 12:00–19:00 (Beijing time) and a northeast wind during other time. The continuous southwest wind in the noon and afternoon can lift the carbonaceous aerosols from the high emission zone in the low central Indo-Gangetic Plain to the high Himalayas. Decesari et al. (2010) and Kirillova et al. (2016) reported the higher mass concentrations of carbonaceous aerosols in the afternoon than those in the night in the Himalaya, due to the polluted air transported from the South Asia in the afternoon.

To clearly display the transports of carbonaceous aerosols along the trans-Himalayan valley, we choose a southwest-northeast cross-section passing Jilong site (red line in Figure S7) to show the vertical variations of BC, OC, and PM$_{2.5}$ mass concentrations in the afternoon (Figure 5). Steady southwesterly wind prevailed above 3-km height along the whole cross section and below 3-km height at the foothill of Himalaya. Figure 5 shows that the high aerosol loading containing abundant OC and BC occurred in the foothill of Himalaya and most of aerosols were blocked by the high elevated Himalaya mountain. However, we found that these atmospheric pollutants penetrated into Himalaya mountain along the trans-Himalayan valley directions during 12:00–19:00 (Beijing time) and a northeast wind during other time. The continuous southwest wind in the noon and afternoon can lift the carbonaceous aerosols from the high emission zone in the low central Indo-Gangetic Plain to the high Himalayas. Decesari et al. (2010) and Kirillova et al. (2016) reported the higher mass concentrations of carbonaceous aerosols in the afternoon than those in the night in the Himalaya, due to the polluted air transported from the South Asia in the afternoon.

To confirm the meteorological influence on carbonaceous aerosols transport, we further examined the actual wind observation data at the 500 hPa height from the major meteorological stations in the South Asia and the southern HTP on 13 and 14 December 2017 (Figure 6). Figures 6a and 6b show that the meteorological stations were actually dominated by the southwesterly winds. Vertical wind fields at the two closest low-altitude meteorological stations south of the Jilong site in the South Asia were southwest above the 3 km height and west or northwest below the 3 km height at 8:00 (Figures 6d and 6e), which is consistent with the vertical wind profile from the WRF-Chem model (Figure 5). During the sampling period, this strong and large-scale southwesterly wind system was available to carry atmospheric pollutants emitted from the South Asia toward the HTP.

The 48-hr backward trajectory frequency was calculated to provide further evidence for indicating the long-range transport of anthropogenic carbonaceous aerosols from the upwind polluted regions (Figure 6c). The backward trajectories mostly originated from the northern India, passed through Nepal, and climbed to the sampling site (Figure 6c) and more than 70% of air mass trajectories were along the south mountain-valley pathways, suggesting that atmospheric pollutants were mainly transported along the mountain-valley channels as shown in the Figure 1.
The CALIPSO data (Figures 7a and 7b) show that a thick atmospheric pollutant layer containing large amounts of polluted continental and smoke aerosols stayed near the ground on the South Asia at 15:30 (Beijing time) on 13 December, which is consistent with the single visible haze layer from the MODIS image (Figure S3a). These polluted continental and smoke aerosols climbed along the south ridge of the Himalayas following the elevated air masses to the sampling site (Figures 7a and 7b). At 16:30 (Beijing time) on 13 December, we also collected an individual particle sample at Jilong site. We found large amounts of soot-bearing and tarball-bearing particles in the sample (Figures 7c–7f), which confirmed the CALIPSO observation at the sampling area. In accordance with the meteorological analysis discussed above, we can conclude that the anthropogenic carbonaceous aerosols from the South Asia could be transported to the sampling site.

In this study, the WRF-Chem model simulation, meteorological observation, air mass backward trajectories, CALIPSO data, and MODIS images all proved that these anthropogenic carbonaceous aerosols collected at the sampling sites were mostly sourced from the biomass and fuel burning in the South Asia. Indeed, a few previous observations and model work have revealed that the atmospheric pollutants from the upwind polluted area can be elevated along the south slope of the Himalayas in the wintertime within a few days (Bonasoni et al., 2010; Kopacz et al., 2011; Lüthi et al., 2015). In view of the topographic and positional similarity in the south margin of the HTP, these south-north valleys provided abundant important ventilation channels for air flows to carry large amounts of anthropogenic carbonaceous aerosols onto the HTP from the low altitude and flat South Asia, which are usually covered by the highly polluted haze layer (Sarkar...
Therefore, we specifically confirmed the north-south mountain-valley as one important transport channel for atmospheric pollutants lifting and penetration into the HTP.

### 3.3. Aging of the Carbonaceous Aerosols

Besides abundant fresh carbonaceous particles as soot and tarballs, there were large amounts of aged carbonaceous particles like soot-SK and tarball-SK. In general, the total number fractions of soot-SK and tarball-SK were 35% at 10:00, 36% at 16:00, 39% at 19:00, and 69% at 21:00 at Jilong site (Figure 8a). This result reflects a significant increase of aged particle number fraction from morning to evening (Figures 8b–8e). Moreover, the WRF-Chem simulation (Figure 4) shows that the continuous southwest wind along the trans-Himalayan valleys in the noon and afternoon could carry the anthropogenic carbonaceous aerosols from the South Asia to the sampling sites, leading to a peak of PM$_{2.5}$ at 19:00 and 18:30.

![Figure 7](image1.png)

**Figure 7.** CALIPSO vertical profiles of (a) 523 nm total attenuated backscatter coefficient (km$^{-1}$ sr$^{-1}$) and (b) aerosol subtype from 15:28 to 15:42 (Beijing time) on 13 December 2017. (c–f) TEM images of the individual samples at 16:30 (Beijing time) on 13 December 2017.

![Figure 8](image2.png)

**Figure 8.** (a) Relative abundance of individual particles at Jilong site on 13 and 14 December 2017. (b–e) TEM images at 10:30, 16:00, 18:30, and 21:50 (Beijing time) at Jilong site.
4. Atmospheric Implications

Soot has been proved to be an important contributor to the climate warming due to the light-absorbing property (Bond et al., 2013; Jacobson, 2001; Ramanathan & Carmichael, 2008) and tarball is another nonnegligible light-absorbing carbonaceous aerosol (Alexander et al., 2008; Hoffer et al., 2017). We found large amounts of soot and tarballs directly emitted from the resident activities in a traditional village in Nepal as one upwind polluted site of a trans-Himalayan valley. These fine particles are the major light-absorbing carbonaceous aerosols from anthropogenic combustion sources (L. Liu et al., 2017; Saleh et al., 2014). According to the regional WRF-Chem simulation, ground-based sampling and subsequent electron microscopy analysis at a remote site and a village site in the trans-Himalayan valleys, large amounts of these light-absorbing carbonaceous aerosols can be transported from the South Asia to the south margin of the HTP along the trans-Himalayan valley. Note that the climatic effect of brown carbon in the trans-Himalayan valleys even in the HTP might be reevaluated due to the occurrence of tarballs (Kirillova et al., 2016; Q. Wang et al., 2019).

The sampling sites are situated in two of numerous trans-Himalayan valleys with similar terrain in south margin of the HTP. The atmospheric pollutants from the valley resident or the faraway upwind area can be transported along all of these valleys to the HTP under favorable weather conditions (Q. Y. Wang et al., 2018; Zhao et al., 2013). Our findings based on individual particle analysis provide clear evidence that the daily extending of the haze over the South Asia lifted up to the HTP along the trans-Himalayan valleys. Obviously, the typical mountain-valleys in the HTP likely play an important role for atmospheric pollutant transport from the South Asia to the HTP. Therefore, more atmospheric pollutants could be deposited on the surface of glaciers in the trans-Himalayan mountains connecting with these valleys. These light-absorbing carbonaceous aerosols might further accelerate the retreating glacier problems and consequently affect the environment and climate there (Kang et al., 2019). Our aerosol observations provide an excellent indication of the mountain-valley transport in the HTP. Our study suggests that the atmospheric models should incorporate atmospheric pollutants being transported in the mountain-valley channels, thereby better evaluating the radiative forcing of the abundant light-absorbing carbonaceous aerosols in the atmosphere and ice glaciers in the typical topography from the South Asia to the HTP.

Data Availability Statement

Fire spots data in Figure 1; PM$_{2.5}$ mass concentrations in the major cities of the northern India and Kathmandu in Figure S3; BC and OC column concentrations distribution in Figure S3; 48-hr backward trajectory frequency endpoint file, individual particle data in Figures 2, 3, 8, S2; PM$_{2.5}$ mass concentration in Figure S1; and all the hourly simulated data are available from the Figshare database (https://doi.org/10.1073/pnas.1900129116). CALIPSO data in Figure 7 are available from the NASA CALIPSO aerosol products (https://www-calipso.larc.nasa.gov/products/lidar/browse_images/std_v4_showdate.php?browse_date=2017-12-13).

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