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# Atmospheric Pollution in the Hindu Kush–Himalaya Region

## Evidence and Implications for the Regional Climate

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*This paper presents a detailed review of atmospheric pollution observed in the Hindu Kush–Himalaya (HKH) region and its implications for regional climate. Data from in situ measurements made at high-altitude stations in the HKH region, observations from satellite-based instruments, and global climate modeling study results are discussed. Experimental observations discussed include both atmospheric measurements and data from snow and ice core sampling from different glaciers in the HKH region. The paper focuses on the atmospheric brown cloud loadings over the Himalayas, particularly black carbon (BC) and ozone, which have links to regional climate and air-pollution-related impacts. Studies show elevated levels of anthropogenic ozone and BC over the Himalayas during the pre-monsoon season with concentrations sometimes similar to those observed over an average urban environment. The elevated concentration observed over the Himalayas is thought to come from the lowlands, especially the highly populated areas of the Indo-Gangetic Plains. The implications of high BC loading in the Himalayan atmosphere as well as elevated BC deposition on snow and ice surfaces for regional climate, hydrological cycle, and glacial melt are discussed.*

**Keywords:** Black carbon; ozone; brown cloud; air pollution; climate change; Hindu Kush–Himalaya.

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### Introduction

In the recent past, field experiments, in situ observations, and satellite monitoring have pointed to the existence of so-called atmospheric brown clouds (ABCs), wide polluted tropospheric layers characterized by anthropogenic aerosol optical depth (AOD) greater than 0.3 and by absorbing AOD greater than 0.03 (Ramanathan, Ramana, et al 2007; Ramanathan et al 2008). ABCs typically consist of particles (referred to as primary aerosols) and pollutant gases. The brownish color of these “clouds” is due to the absorption and scattering of solar radiation by anthropogenic black carbon (BC), fly ash, soil dust particles, and nitrogen dioxide gas. ABCs’ implications for regional and global climate were rapidly recognized; they represent a global climate issue (Ramanathan and Carmichael 2008).

ABC hotspots have been identified in East Asia, the Indo-Gangetic Plains, Southeast Asia, Southern Africa, and the Amazon Basin, and substantial ABCs have also been observed over the eastern United States and Europe (Ramanathan, Li, et al 2007). However, in these extra-tropical regions, the atmospheric concentrations of ABCs are large mainly during the summer, since precipitation removes the aerosols efficiently during the other seasons.

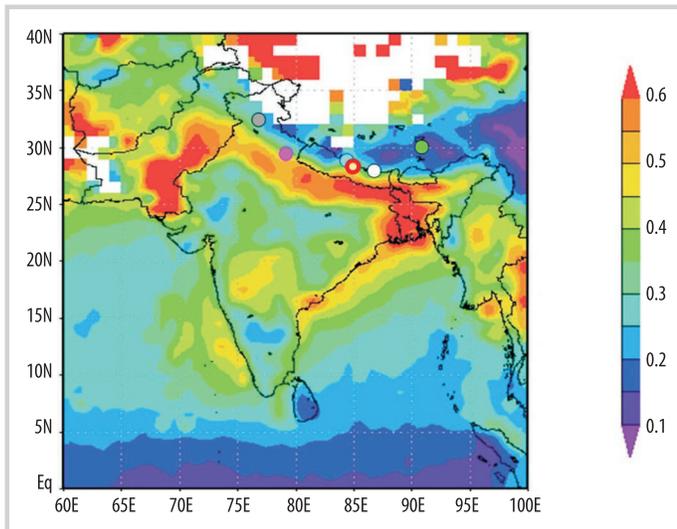
Satellite observations (Figure 1) show that the ABC over the Indo-Gangetic Plains, while extending thousands of miles southward and

over the north Indian Ocean during the dry season, is characterized by a sharp boundary to the north, due to the barrier of the Hindu Kush–Himalaya (HKH) ridge. Thus, these high mountain regions can be affected by dust and pollutant-rich air masses, as indicated by the relatively high AOD values observed by Ramana et al (2004) over the Nepal Himalayas, and by Cloud Aerosol Lidar and Infrared Pathfinder Satellite (CALIPSO) measurements, which indicate that the brown cloud over northern India can extend from 3 to 5 km above sea level (a.s.l.) (Ramanathan, Ramana, et al 2007).

The transport of polluted air masses rich in optically active aerosol and ozone to the HKH region is a matter of great concern. In fact, in addition to influencing the regional radiative forcing of the atmosphere, surface O<sub>3</sub> can strongly affect ecosystems, with possible severe impacts on biodiversity (Royal Society 2008) and agriculture (Agrawal et al 2008). Lau et al (2006) proposed that once aerosols like BC are transported to elevated altitudes (above 3 km), their absorption may affect the South Asia monsoon cycle, which suggests that the regional atmospheric heating by absorbing aerosol over the Himalayas can lead to a northward shift of the monsoon rain belt, resulting in increased rainfall in northern India and the foothills of the Himalayas in the late spring and early summer (the “elevated heat pump effect”).

Increased absorption of aerosols can affect the mountain cryosphere in several ways. As proposed by

**FIGURE 1** Satellite (MODIS) integrated atmospheric optical depth over South Asia from February to May 2007 (courtesy of Giovanni online data system from the NASA GESDISC, after Bonasoni et al 2010, reproduced with permission). The colored scale represents the geographical distribution of the atmospheric optical depth values. The colored circles represent locations for which atmospheric measurements are reported in this work: Manali and Kothi (gray), Manora Peak/Nainital (purple), Langtang (cyan), Godavari and Nagarkot (red and yellow), Nepal Climate Observatory-Pyramid (white), and Nam Co (green).



Ramanathan, Ramana, et al (2007), the atmospheric warming related to the ABC haze on the south slope of the Himalayas might be sufficient to account for the observed retreat of the Himalayan glaciers. Additionally, even small BC amounts can significantly modify snow reflectance, thus altering snowmelt rate and snow spatial coverage, which influences the climate through the snow-albedo feedback. Flanner et al (2009) showed that the absorption by BC impurities in snow exceeds the “dimming” effect and becomes significant for BC concentrations greater than 10 ng/g. These processes may lead to a significant global snow/ice/climate feedback but also, on a regional scale, to critical changes in the local hydrology and water availability related to glacier and snow discharge. However, despite these issues, systematic activities devoted to the characterization of atmospheric composition variability in the HKH region are still sparse, and there is a need for more

comprehensive documentation, assessment, and quantification of the influence of anthropogenic polluted air-mass transport to these mountain regions.

This article summarizes the most recent evidence of the transport of air mass rich in anthropogenic pollution and biomass burning emissions (O<sub>3</sub> and black soot particles) to the HKH region and discusses possible implications for the climate of this very sensitive region. It presents results from experimental activities in the HKH region that involved both direct atmospheric observations and chemical analyses of snow/ice depositions. In particular, it provides typical values and seasonal variability of surface O<sub>3</sub> and aerosol composition at several high-altitude measurement sites in the Himalayas and on the southern Tibetan Plateau. Then it reviews existing studies of air pollution in this high mountain region, also describing the principal mechanisms that favored the

transport of polluted air masses to the HKH region. Finally, it discusses some of the most striking evidence of the possible impacts of these pollution transports on the HKH regional climate.

### Typical levels and seasonal variability of aerosol properties and surface O<sub>3</sub> in the HKH

On a seasonal basis, the air composition in the Himalayan region is strongly influenced by large-scale atmospheric circulation, which alternates between the wet summer monsoon and dry season and determines meteorological conditions that are more or less conducive to the accumulation of pollutants in the lower troposphere and subsequent transport to high mountain regions. Table 1 provides the tentative average time periods of the seasons observed in the South Himalayas using meteorological observations at the Nepal Climate Observatory-Pyramid (NCO-P) in the high Khumbu Valley in Nepal.

During non-monsoon periods the occurrence of dry meteorological conditions over South Asia and a prevalent westerly circulation over the Himalayas lead this region to be particularly exposed to the pollution and biomass-burning products that accumulate over the Indo-Gangetic Plains (Ramanathan, Ramana, et al 2007), as well as to the long-range transport of pollution from far downwind regions like the Middle East, North Africa, and Europe (Carrico et al 2003). During the winter and pre-monsoon seasons, the persistent subsidence and trade wind inversion strongly inhibit the ventilation and dispersion of pollutants, favoring their buildup in

**TABLE 1** Average onset and decay of seasons over the southern Himalayas, based on meteorological observations by the Nepal Climate Observatory-Pyramid.

	Pre-monsoon	Monsoon	Post-monsoon	Winter
Onset	1 March	17 May	5 October	15 November
Decay	16 May	4 October	14 November	28 February

the lower troposphere in the South Asian region (Ramanathan, Li, et al 2007).

Due to the relatively high solar irradiance even during winter and spring in the tropical regions, thermal wind systems can develop along mountain valleys and mountain slopes (eg Henne et al 2005). Together with entrainment processes related to diurnal atmospheric boundary layer (ABL) growth and topographic venting, during daytime such wind systems can efficiently contribute to the transport of pollutants from the foreland plains or from the bottom of the valley to higher altitudes, even favoring the injection of large amounts of pollutants directly from the northern rim of the ABC. Thus, as reported by Bonasoni, Laj, et al (2010), the seasonal variation in surface  $O_3$ , BC, and aerosol particles in the high Himalayas is dominated by higher concentrations during winter and spring (pre-monsoon), minimal concentrations during the summer wet monsoon season, and increasing levels during the autumn (post-monsoon).

Carrico et al (2003) analyzed particle mass concentrations (PM10 and PM2.5), chemical composition, and wavelength-dependent AOD from December 1998 to October 2000 at Nagarkot (on the edge of the Kathmandu Valley, 27.72°N, 85.52°E; 2195 m a.s.l.) and Langtang Mountain (28.13°N; 85.60°E; 3920 m a.s.l.). These observations showed aerosol concentrations peaking in February to May (average PM2.5:  $5.59 \pm 61 \mu\text{g}/\text{m}^3$  at Nagarkot), while a decrease in PM2.5 (average value:  $8 \pm 7 \mu\text{g}/\text{m}^3$ ) was observed from June through September, indicating that the broad-scale South Asian monsoon system plays a fundamental role in modulating the efficiency of aerosol transport to the central Himalayas. This seasonal profile is also observed in the elemental carbon (EC) at Langtang that was characterized by higher average values from October to May (from  $0.52 \pm 0.48 \mu\text{g}/\text{m}^3$  to

$0.48 \pm 0.38 \mu\text{g}/\text{m}^3$ ) and lower concentrations during the summer monsoon ( $0.15 \pm 0.16 \mu\text{g}/\text{m}^3$ ).

Further aerosol measurements (BC and total particle mass concentrations) were carried out at Manora Peak/Nainital (29.37°N, 79.45°E, 1950 m a.s.l.) in the Indian Himalayas, during a comprehensive aerosol field campaign in December 2004. For these earlier measurements, Pant et al (2006) reported that total suspended particulate concentration was in the range 15–40  $\mu\text{g}/\text{m}^3$  (mean value  $27.1 \pm 8.3 \mu\text{g}/\text{m}^3$ ), while BC had a mean concentration of  $1.36 \pm 0.99 \mu\text{g}/\text{m}^3$ , contributing  $5.0 \pm 1.3\%$  to the composite aerosol mass. These relatively high concentrations indicate not-negligible pollution influence at this mountain site.

For this measurement site, from February 2005 to July 2008, Ram et al (2010) reported higher concentrations of carbonaceous aerosol during winter and spring, with monthly average concentrations ranging from 1.7 to 1.9  $\mu\text{gC}/\text{m}^3$  for EC and from 8.8 to 12.3  $\mu\text{gC}/\text{m}^3$  for OC (organic carbon). Conversely, lower concentrations were observed during the monsoon, which was attributed to the more efficient wash-out of aerosols and lower biomass burning emissions. These results are in good agreement with BC measurements (Dumka et al 2010) that described higher BC mass concentrations during spring (mean value:  $1.34 \pm 0.05 \mu\text{g}/\text{m}^3$ ) than during summer ( $0.53 \pm 0.02 \mu\text{g}/\text{m}^3$ ) or autumn ( $1.03 \pm 0.04 \mu\text{g}/\text{m}^3$ ) from November 2004 to December 2007. According to Ram et al (2010), on average, carbonaceous aerosols contribute nearly 25% of the total suspended particulate mass, while the mass fraction of absorbing EC ranged from less than 1% (during the summer) to as high as 7.6% (during the winter), and the absorption coefficient at 678 nm varied between 0.9 and 33.9  $\text{Mm}^{-1}$ .

Ram et al (2010) reported that the average OCEC ratio in 86 aerosol samples was  $7.7 \pm 3.4$  with single

value up to 27.2. This relatively high OC/EC ratio at Manora Peak/Nainital suggests that these species are derived from primary sources (such as biomass burning on the Indo-Gangetic Plains) and contribution of secondary organic aerosols. In particular, the K+/OC ratios (range: 0.01–0.09; average:  $0.05 \pm 0.02$ ), similar to the ratios obtained in the Indo-Gangetic Plains, suggested burning of agricultural waste and fuelwood as a major source of carbonaceous aerosols, especially during winter, when a relative increase in K+ concentrations was observed at Manora Peak/Nainital ( $0.6 \mu\text{g}/\text{m}^3$ ).

Stone et al (2010) carried out a source apportionment study of OC and EC in Godavari (27.59°N, 85.31°E, Nepal), a site located in the central Himalaya foothills at 1600 m a.s.l. During 2006, particulate mass (PM2.5), OC, and EC concentrations in daily aerosol samples followed the occurrence of the ABC in South Asia, with maximum concentrations during the dry winter season and minimum concentrations during the wet summer monsoon. In particular, at this measurement site, the yearly average PM2.5 was  $26 \pm 19 \mu\text{g}/\text{m}^3$  ( $\pm 1$  standard deviation), with a maximum in mid-January ( $120 \pm 10 \mu\text{g}/\text{m}^3$ ) and a minimum in August ( $< 0.8 \mu\text{g}/\text{m}^3$ ). The yearly average OC and EC concentrations ( $\pm 1$  standard deviation) were  $4.8 \pm 4.4 \mu\text{gC}/\text{m}^3$  and  $1.0 \pm 0.8 \mu\text{g}/\text{m}^3$ , respectively.

In the central Himalayas, continuous and long-term atmospheric composition observations and pollution transport monitoring have also been carried out since 2006 at the Nepal Climate Observatory-Pyramid (NCO-P) at 5079 m a.s.l. in the high Khumbu valley not far from the Everest base camp (27.95°N, 86.82°E). The first two years of continuous observation at NCO-P, which are the focus of a special issue on ABCs in *Atmospheric Chemistry and Physics* (McFiggans and Schauer 2010), made it possible to identify and characterize the

atmospheric background conditions, providing information on the variability of the principal aerosol components and trace gases, identifying the periods influenced by the ABC and characterizing the frequency and intensity of acute pollution events. Decesari et al (2010) reported that at the NCO-P, despite its high altitude, the average PM10 mass apportioned by the chemical analyses is of the order of  $6 \mu\text{g m}^{-3}$  ( $10 \mu\text{g/scm}$ ), with almost half of this mass accounted for by organic matter, EC, and inorganic ions, the rest being mineral dust. Organic matter accounted for  $2.0 \mu\text{g/m}^3$  ( $3.6 \mu\text{g/scm}$ ) on a yearly basis, and it is by far the major PM10 component beside mineral oxides. Non-negligible concentrations of EC were also observed ( $0.36 \mu\text{g/scm}$ ), confirming that light-absorbing aerosol produced from combustion sources can be efficiently transported up the altitudes of Himalayan mountains; the carbonaceous species are very abundant ( $\text{TC} = 1\text{--}4 \mu\text{g/scm}$  as seasonal averages), and their concentrations dominate those of total inorganic ions, on average.

As reported by Bonasoni, Laj, et al (2010), atmospheric composition at NCO-P is influenced by the South Asian summer monsoon cycle, as highlighted by the common seasonal behaviors of most of the atmospheric compounds observed here (Figure 2): the lowest seasonal values of  $\text{O}_3$  ( $38.9 \text{ ppbv}$ ) and BC ( $49.6 \text{ ng/m}^3$ ) were observed during the summer monsoon when most (96%) of the air masses reaching the measurement site are related to synoptic-scale uplift and were affected by strongly precipitating low-pressure systems over the Bay of Bengal. Conversely the highest seasonal values of  $\text{O}_3$  ( $60.9 \text{ ppbv}$ ) and BC ( $316.9 \text{ ng/m}^3$ ) were observed during the pre-monsoon season, when far westerly and regional-scale air-mass transport strongly affected the measurement site (Decesari et al 2010; Cristofanelli et al 2010; Marinoni et al 2010; Sellegri et al 2010).

A similar  $\text{O}_3$  seasonality was detected at Manora Peak/Nainital by Kumar et al (2010), who found a maximum in late spring (May:  $67.2 \pm 14.2 \text{ ppbv}$ ) and a minimum in the summer/monsoon season (August:  $24.9 \pm 8.4 \text{ ppbv}$ ) from October 2006 to December 2008. Kumar et al (2010) compared Manora Peak/Nainital  $\text{O}_3$  behavior with observations carried out at a high mountain station in western India, pointing out significant differences that magnify the peculiarity of  $\text{O}_3$  variability in the high Himalayas compared to other locations in central South Asia.

Pollutant observations in high mountain environments also show a differing seasonality than that observed by seasonal distribution of ABC over South Asia (Adhikary et al 2007; Ramanathan, Ramana et al 2007). While these studies reported that ABC concentrations peaked during the post-monsoon season and dissipated with the onset of the summer monsoon over much of South Asia, observations in high mountain environments show that pollutants build up starting in the post-monsoon season while concentrations peak during the pre-monsoon season. The time difference in the peak period could be due to meteorological conditions (such as the atmospheric boundary layer's height and valley winds) and/or seasonal emissions such as those from biomass burning.

While more and more studies are illustrating the atmospheric composition and seasonality of aerosols over the Himalayas, there is still a substantial knowledge gap about sources and sinks of pollutants over this region. While in situ measurements provide information on pollutant concentrations, they are limited by the problem of representativeness and cannot by themselves provide a link with emission sources, a necessity for mitigation strategies. Three-dimensional modeling studies provide a means to link observed concentration with emission sources, but they are also limited by uncertainties about emissions,

meteorology, and aerosol/chemistry interactions. In addition, to simulate the complex flows over steep terrains, high-resolution model studies are required. This is currently lacking in the HKH region. While global model studies (discussed below) certainly provide scientific insights into atmospheric phenomena, they have limited ability, on a country scale, to provide decision support to policy-makers for effective control strategies because of the coarseness of the modeling grid resolution.

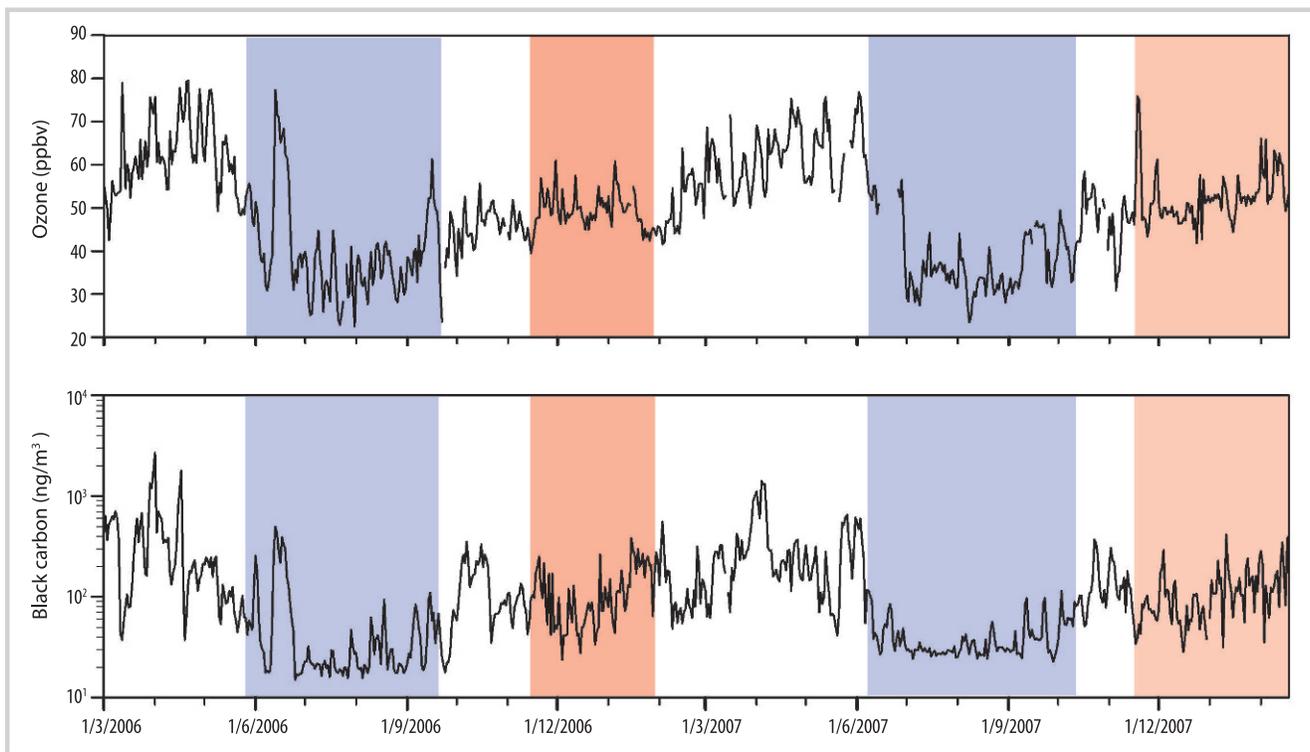
## Evidence of polluted air-mass transport to the Himalayas

### Snow and ice core sampling

Even if precipitation and snow melt can strongly influence the concentration of particles in the snow or ice cores, investigations aimed at determining the impurity contents on glacier surfaces can uncover indirect information about the effect of polluted air masses on mountain environments. For instance, Xu et al (2006), Ming et al (2009), and Xu, Cao, et al (2009) presented BC and EC concentrations in the snow collected from glaciers spanning from Karakoram to the east Himalayas with values ranging from 4 to  $67 \mu\text{g kg}^{-1}$  (see Figure 5). Xu, Cao, et al (2009) discussed long-term (1950s–2000s) measurements of BC content on three glaciers on the south rim of the Tibetan Plateau, pointing out that the amount of black soot has rapidly increased since the 1990s, with snow concentrations almost double those of the 1980s. As discussed by Xu, Cao, et al (2009), these glaciers should receive BC from the south (by the summer monsoon circulation) and from the west during spring and winter.

In the 1950s and 1960s, European emissions probably drove black soot concentration variability, but nowadays the transport of polluted air masses from the ABC over South Asia can strongly affect BC content over these glaciers. In fact, for glaciers on the South Tibetan Plateau, Xu,

**FIGURE 2** Surface ozone and black carbon average daily concentrations at the Nepal Climate Observatory-Pyramid from March 2006 to February 2008. Blue bars indicate the summer monsoon, and red bars indicate winter. The highest ozone and black carbon values are clearly discernible during the pre-monsoon seasons. (Adapted with permission from Bonasoni et al 2010)



Cao, et al (2009) and Xu, Wang, et al (2009) showed a distinct seasonal variability for carbonaceous aerosol concentrations in snow/firn cores, with doubled EC concentrations during the dry season, when the brown cloud is spreading toward the south side of the Himalayas (see previous section). In particular at the Palong-Zanbu No. 4 Glacier (29.96°E, 29.12°E), Xu, Wang, et al (2009) pointed out that EC, water-insoluble organic carbon, and  $\text{SO}_4^{2-}$  concentrations in the ice core were 4.7, 56.0, and 4.2  $\text{ng g}^{-1}$  in 1998 and increased to 16.8, 144.4, and 162.1  $\text{ng g}^{-1}$  in 2005. Analysis of  $\text{EC}/\text{SO}_4^{2-}$  and  $\text{EC}/\text{K}^+$  ratios suggested that fossil fuel burning made a greater contribution to EC deposition than biomass burning, with significant contributions to secondary organic aerosol production as deduced by the low ratios of EC to water-insoluble organic carbon.

Signals of anthropogenic pollution transport can even be found in the foothills of Mt Everest,

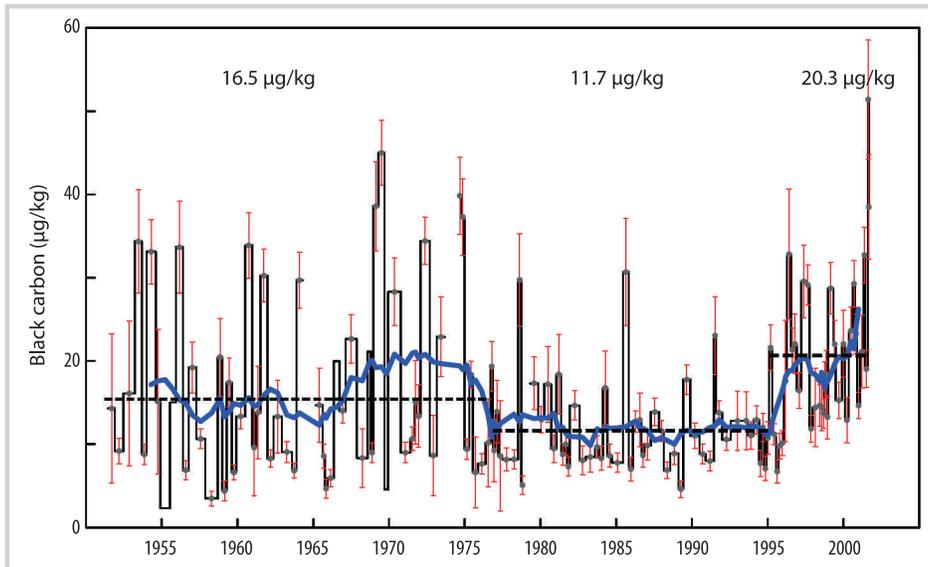
as indicated by Ming et al (2007), who found  $\text{SO}_4^{2-}$  increased (up to 22.20  $\mu\text{eq L}^{-1}$ ) in fresh snow sampling at Repula Col on the East Rongbuk Glacier (28.02°N, 86.96°E, 6500 m a.s.l.) in connection with air-mass transport from the Indo-Gangetic Plains. This evidence indicates that anthropogenic inputs are potentially important even in the remote high-altitude atmosphere in the central Himalayas. It was further corroborated by Lee et al (2008), who analyzed trace element concentrations in a 2.1-m snow pit in the same region from autumn 2004 to summer 2005; these authors found fallout fluxes of anthropogenic trace elements (Cr, Ni, Cu, and Zn) higher than those observed in Greenland and Antarctic snow samples.

The continuous measurement of BC in a 40 m shallow ice core retrieved from the East Rongbuk Glacier in the northeast saddle of Mt Everest (Ming et al 2008) revealed an apparent increasing trend (for the smooth average, see Figure 3) of BC

concentrations in the ice since the mid-1990s (average value:  $20.3 \pm 9.2 \mu\text{g/kg}$  for 1995–2002, with values exceeding 50  $\mu\text{g/kg}$  in summer 2001). Ming et al (2008) hypothesized a significant impact of air mass transported from South Asia, suggesting that BC emitted from this region could penetrate the Tibetan Plateau by climbing over the elevated Himalayas. These results are in agreement with Kaspari et al (2011), who, by analyzing ice core spanning from 1860 to 2000, pointed out that BC concentrations from 1975 to 2000 have increased approximately threefold relative to 1860–1975, from  $0.2 \pm 0.3 \mu\text{g/l}$  to  $0.7 \pm 1.0 \mu\text{g/l}$ .

Finally, measurements of organochlorine pesticides and polychlorinated biphenyls in the 2.1-m snowpack samples collected from the East Rongbuk Glacier in September 2005 have been reported by Kang et al (2009), who found that, compared to other high mountain areas of the world, persistent organic pollutant levels were relatively low over Mt Everest, even if their

**FIGURE 3** Time series of black carbon concentrations for samples sectioned from the East Rongbuk Glacier. Error bars are presented as solid red lines with grey dots, and the thick solid blue curve represents the smooth average. Average black carbon concentration levels during 1951–1975, 1975–1994, and 1995–2002 are shown as dashed lines. (Adapted with permission from Ming et al 2008)



presence clearly testifies to the arrival of anthropogenically polluted air masses mainly from India and China.

#### Atmospheric observations

Possible direct evidence for influence of polluted air masses at high-altitude sites in the northwestern Himalayan region has been provided by Kuniyal et al (2007), who presented  $O_3$  measurements at Manali and Kothi mountain sites ( $32.40^\circ N$ ,  $77.16^\circ E$ , and 2050 and 2530 m a.s.l., respectively). During May and June from 1998 to 2004, average  $O_3$  values of 28–29 ppb were observed at these measurement sites with evident average diurnal variations characterized by nighttime minima (below 25 ppb) and day-time maxima (above 40 ppb at Manali and 30 ppb at Kothi), possibly indicating local photochemical production or upward transport of polluted air masses from the surrounding region and already enriched in photochemical  $O_3$  connected with tourist vehicles or biomass burning related to forest fires, agricultural activities, cooking, and heating during the frequent power failures. At Manora Peak/Nainital, Kumar et al (2010) investigated the influence of

regional pollution from the northern Indo-Gangetic Plains on seasonal  $O_3$  variability. Besides suggesting the possible influence of biomass burning emissions on the occurrence of the observed  $O_3$  spring peak, these authors estimated a 3.2 ppbv/d net  $O_3$  photochemical production for air masses traveling over the northern Indian subcontinent during spring. In particular, the average yearly contribution of these regional polluted air masses to the observed  $O_3$  levels at Manora Peak/Nainital was estimated to be 7 ppbv, with a maximum of 16.5 ppbv in May and June.

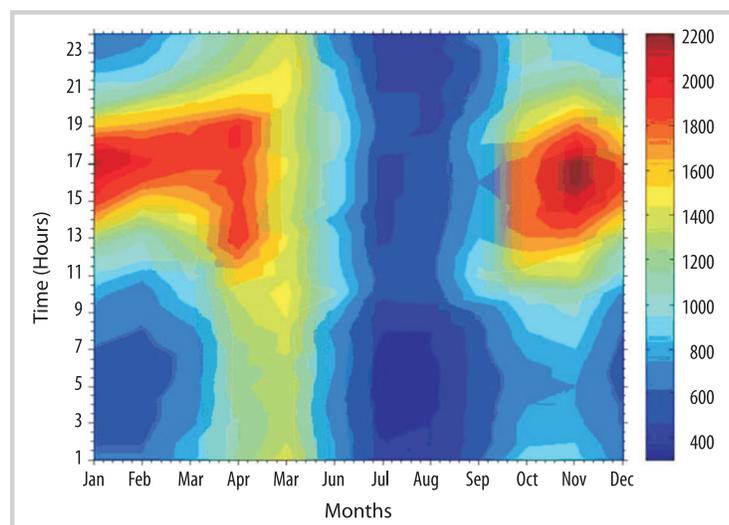
Some of the earlier evidence about the systematic influence of anthropogenic pollution on atmospheric composition over the high Himalayas and aerosol properties was pointed out by Carrico et al (2003) at Nagarkot and Langtang. This study indicated, for both measurement sites, an important influence of combustion aerosols (mainly from fossil fuel use and biomass burning from the Kathmandu Valley and the Indian subcontinent) on atmospheric light extinction and air quality.

The importance of pollution transport to the western Himalayas

has been further confirmed by Dumka et al (2008, 2010) and Ram et al (2010), who examined multiyear BC mass concentrations, aerosol composition, and AOD values at this measurement site. These investigations discovered that boundary layer dynamics and along-valley circulations play a key role in transporting aerosols from the polluted valley region to higher Himalayan altitudes. In fact, during the dry season (October–March), BC values exhibited well-defined diurnal variations with a prominent peak in the late afternoon and mass concentrations always higher by a factor of  $\sim 2$  during daytime than during nighttime, when the measurement site is less influenced by upward air-mass transport (Figure 4).

Panday and Prinn (2009) and Panday et al (2009) executed field measurements and model simulation of air pollution transport and meteorology in the Kathmandu Valley during the dry season of 2004–2005. They found that even if the air mass on nearby mountaintops was disconnected from pollution within the valley during the night, it nevertheless received significant pollution during the morning, when

**FIGURE 4** Monthly mean diurnal variation of black carbon from November 2004 to December 2007 at Manora Peak/Nainital. The color code on the right represents concentrations expressed as  $\text{ng}/\text{m}^3$ . (Modified with permission from Dumka et al 2010)



up-slope flows from the valley bottom began. At Nagarkot,  $\text{O}_3$  up to 75 ppbv was observed in April 2005, clearly indicating that long-valley and slope circulation represent an important mechanism favoring the diffusion of anthropic pollutants to higher altitudes. As reported by Stone et al (2010), at Godavari, over the course of a year of measurements, 24-hour average OC and EC concentrations peaked at  $33.0 \pm 1.7 \mu\text{gC}/\text{m}^3$  and  $4.1 \pm 0.7 \mu\text{gC}/\text{m}^3$ , respectively. Based on the analysis of specific molecular markers, these authors indicated that atmospheric pollution related to motor vehicles, coal combustion, biomass burning including cow dung, vegetative detritus, and secondary organic aerosol was likely to be the source of these carbonaceous aerosols. A significant emission source was not identified; we still need to know more regarding sources and sinks of organic carbon even in a city like Kathmandu.

Overall, these results indicate that the southern side of the Himalayas is impacted by the upward transport of pollutants from a large reservoir in the foothills and/or upwind regions, which represent the northern rim of the South Asian ABC. This has been confirmed by Bonasoni, Laj, et al (2010), who found that 20% of

pre-monsoon days were characterized by strong ABC transport episodes in the period March 2006–February 2008. These pollution transport events were characterized by significant  $\text{O}_3$  and BC increases (13.6% and 522.9%, respectively) over the average seasonal value ( $\text{O}_3$ : 60.9 ppbv, BC:  $316.9 \text{ ng C}/\text{m}^3$ ). This clearly indicates that the Himalayan valleys represent efficient and persistent channels for the transport of large amounts of brown cloud pollutants up to the high Himalayas.

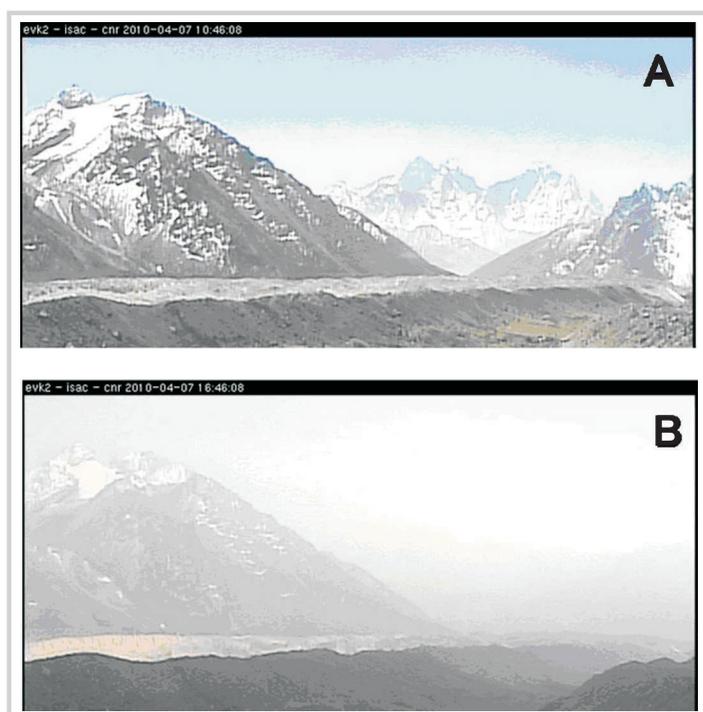
Marinoni et al (2010) showed that due to the efficient daytime up-valley winds, the typical BC and PM1 diurnal cycles are characterized by an afternoon maximum with hourly values peaking, respectively, at  $\sim 550 \text{ ng}/\text{m}^3$  and  $\sim 6 \mu\text{g}/\text{m}^3$  during the pre-monsoon season. Based on work at NCO-P, Bonasoni, Cristofanelli, et al (2010) and Marinoni et al (2010) reported acute pollution events with BC exceeding  $5 \mu\text{g}/\text{m}^3$  (30-minute mean value) and  $\text{O}_3$  peaking at 90–95 ppbv during the pre-monsoon season—values typical for urban areas. The thick haze accompanying these polluted air-mass transports can even be discerned in pictures taken at the NCO-P during pre-monsoon days (Figure 5), testifying to the transport of pollution plumes and their influence in reducing

visibility. Once transported to the high Himalayas, these pollutants can be injected into the free troposphere, where they attain a longer lifetime and/or accumulate in high residual layers. This mechanism has been hypothesized by Marinoni et al (2010), who observed nighttime BC levels at NCO-P that follow the same seasonality as the daytime values. This was particularly evident during the pre-monsoon season when, although lower than the daytime values (average value:  $1042 \text{ ng}/\text{m}^3$ ), the nighttime and early morning particle concentrations still remain high (BC nighttime average value:  $250 \text{ ng}/\text{m}^3$ ).

It should be clear that besides the valley breeze regime, synoptic air-mass circulation can also influence the atmospheric composition at NCO-P. Analysis of BC, PM1, and PM10 observations as a function of large-scale air mass circulation confirms that long-range transport significantly impacts aerosol concentration over the central southern Himalayas. In particular, significant differences are observed within each season between aerosol concentrations in air masses of different origins and paths: polluted aerosols were mainly related to air masses associated with transport from the Middle East, Europe, and South Asia (Marinoni et al 2010).

Possible influence of polluted air-mass transport from South Asia toward the southern Tibetan Plateau has been proposed by Ming et al (2010), who analyzed continuous air and precipitation sampling for carbonaceous particles at the Nam Co Observation Station ( $30.77^\circ\text{N}$ ,  $90.99^\circ\text{E}$ ; 4730 m a.s.l.). Even if relatively low BC concentrations were found at this measurement site for both atmospheric and precipitation samples ( $82 \text{ ng}/\text{m}^3$  and  $7.8 \text{ ng}/\text{g}$ , respectively), a few episodes of atmospheric BC increases (up to  $200 \text{ ng}/\text{m}^3$ ) have been identified during the pre-monsoon season and associated with polluted air-mass transport events from the

**FIGURE 5** Images of the Himalayan Khumbu valley, with the Khumbu glacier in the foreground, from the Nepal Climate Observatory-Pyramid on 7 April 2010. The afternoon conditions (B) clearly testify to the transport of atmospheric brown cloud compounds and their influence in reducing visibility in the high Himalayas, while the morning image (A) reveals the pre-monsoon background conditions. (Reproduced with permission from Bonasoni, Cristofanelli, et al 2010)



Indo-Gangetic Basin. These results are in qualitative agreement with ice core records, which have indicated a significant increase in deposition of both BC and OC over the northern slope of the Tibetan Plateau, especially since 1990 (Ming et al 2008; Xu et al 2009a).

### Recent estimates of the climate impacts of pollution transport to the HKH region

As shown by the atmospheric composition observations and the ice/snow chemical analyses, the pristine Himalayan atmosphere can be impacted by the transport of polluted air masses rich in  $O_3$  and absorbing aerosol. Recent studies have pointed out that BC transport and deposition can significantly affect the cryosphere by modifying snow reflectance and thus altering snowmelt rate and snow spatial coverage. As reported by Xu, Wang, et al (2009), it has been estimated that absorption of visible solar radiation

by fresh snow can increase from 10% to 100% due to a BC amount of 10 ng/g, depending on the size and shape of snow crystals and on whether the BC is internally or externally mixed with the crystals. In particular, Ming et al (2008) estimated that BC present in the Mt Everest ice cores produced a summer darkening effect of 4.5  $W/m^2$  in summer 2001. The research activities carried out at the NCO-P permitted preliminary estimates of key impacts of this anthropogenic pollution transport on the high Himalayas climate. In particular, Yasunari et al (2010) estimated the possible impact of dry deposition of BC on high Himalayan glaciers and snow melting.

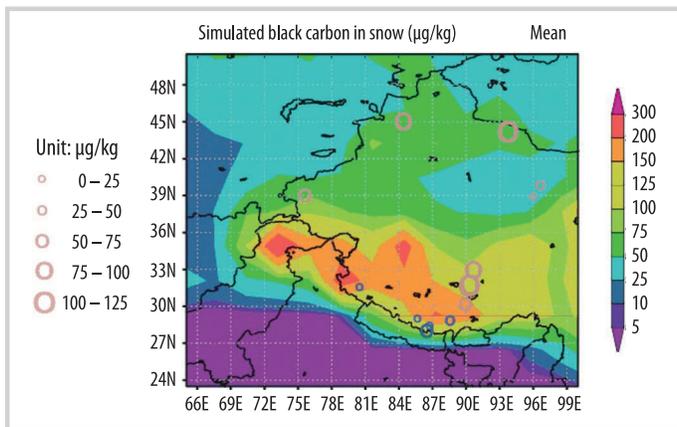
Based on atmospheric composition and meteorological observations at the NCO-P, a total BC deposition of 266  $\mu g/m^2$  has been calculated for the Khumbu glacier during the pre-monsoon season of 2006, which corresponds to a BC snow concentration of 26.0–68.2  $\mu g/kg$ . Such a concentration of BC in snow could result in albedo

reductions ranging from 2.0% to 5.2%. By assuming that these albedo reductions continue throughout the year, a runoff increase of 70–204 mm of water was calculated by Yasunari et al (2010); this value represents 11.6–33.9% of the annual discharge of a typical Tibetan glacier.

Even considering the uncertainties related to these estimates, this investigation clearly indicates that BC deposition on Himalayan glaciers can have not-negligible consequences on surface water runoffs from snow and ice melting. Qian et al (2011) performed a modeling study with a global climate model to simulate the upper limits of radiative effects related to BC deposition in snow over the Tibetan Plateau and in the HKH region (Figure 6). Simulation results show a large BC content (exceeding 150  $\mu g/kg$ ) in snow over the HKH, due to the export of pollution from the Indo-Gangetic Plains. Because of the high aerosol content in snow and large incident solar radiation in low latitudes and high altitudes, large surface radiative flux changes were induced by BC in snow over HKH, with a maximum in the pre-monsoon season (up to 10  $W/m^2$ ) with a surface albedo that can decrease from 2% to 14% over the HKH region. In particular, over the Hindu Kush–Karakoram, BC deposition in snow can increase the surface air temperature up to  $\sim 1.0$  K, reducing the pre-monsoon snowpack by as much as 40%, a more efficient reduction than that caused by  $CO_2$  and carbonaceous aerosol increases in the atmosphere since the preindustrial age.

Kopacz et al (2011) conducted a modeling analysis of the source regions for BC arriving at several remote locations in the Himalayas and Tibetan Plateau. They found that emissions from northern India and central China contributed the majority of BC to the Himalayas, although the precise location varied with the season. For instance, on a yearly basis these authors estimated

**FIGURE 6** Simulated annual mean (shaded) and measured (in circle) black carbon content in snow (unit:  $\mu\text{g}/\text{kg}$ ). As indicated by the legend, the circle size represents the observed black carbon content. The observation data are from Xu et al (2006), denoted by pink circles, and Ming et al. (2009), denoted by blue circles. (Reproduced with permission from Qian et al 2011)



that emissions from India, China, and Nepal delivered about 91% of BC in the atmospheric column over Mt Everest. Based on this study, African biomass burning (in winter and spring) and Middle Eastern fossil fuel combustion (mostly in spring) can significantly contribute to the BC reaching the Himalayas, thus influencing the radiative forcing of these snow-covered regions. These authors also found that the monthly mean direct forcing (with clear-sky and internal mixing adjustment) due to the BC-induced snow-albedo effect was important, varying within  $8\text{--}15\text{ W}/\text{m}^2$  over the central Himalayas and significantly exceeding the direct BC atmospheric forcing (up to  $3\text{ W}/\text{m}^2$ ).

Pant et al (2006) calculated radiative forcing related to  $\text{CO}_2$  aerosol observed over Manora Peak/Nainital in December 2004, finding an average value of  $-4.2\text{ W}/\text{m}^2$  at the surface and  $+0.7\text{ W}/\text{m}^2$  at the top of the atmosphere, implying an atmospheric forcing of  $+4.9\text{ W}/\text{m}^2$ . These values implied a large forcing efficiency of atmospheric aerosol over the western Himalayas ( $88\text{ W}/\text{m}^2$ ) due to the high BC mass fraction. By using the NCO-P observations and a radiative transfer model, Marcq et al (2010) recently calculated the direct local radiative atmospheric forcing due to aerosols for specific classes of air-mass transport. They found that the

presence of absorbing particulate material can locally induce an additional top of the atmosphere (TOA) forcing of  $10\text{ to }20\text{ W}/\text{m}^2$  for the first atmospheric layer (500 m above surface). The TOA positive forcing depends on the presence of snow at the surface, and takes place preferentially during ABC transport episodes along the Khumbu Valley. Warming of the first atmospheric layer is paralleled by a substantial dimming of the amount of radiation reaching the surface (ranging from  $-4\text{ to }-20\text{ W}/\text{m}^2$ ). As shown by this study, the calculated surface forcing is also very dependent on surface albedo, with maximum values occurring over a snow-covered surface. Overall, these estimates suggested a TOA forcing significantly greater than the Intergovernmental Panel for Climate Change (IPCC) reported values for greenhouse gases.

Recently Lau et al (2010) showed that the elevated heat pump (EHP) effect can also lead to widespread enhanced land-atmosphere warming, accelerating the melting of seasonal snow cover in the Himalayas and Tibetan Plateau. These authors suggest that, especially from late April to mid-May, the warming and moistening of atmosphere overlying the Tibetan Plateau and the HKH region causes a reduction in surface sensible and latent heat fluxes from land to atmosphere. The related net heat gain induces excess snowmelt.

This mechanism is proposed to add to the darkening of snow by deposition of soot and dust, leading to increasingly accelerated snowmelt. These results are reinforced by the work of Qian et al (2011), who showed that during the South Asian pre-monsoon season, BC and organic particles can warm the troposphere over the HKH and Tibetan Plateau by absorbing sunlight, while the surface snow polluted by BC absorbs more solar radiation and warms the overlying air masses. Both these effects can enhance the upward motion of air mass and favor deep convection along HKH during the pre-monsoon season, resulting in earlier onset of the summer monsoon and increased convective precipitation (up to  $2\text{ mm}/\text{d}^1$ ) over northern India and HKH.

Efforts to understand the composition and seasonal characteristics of pollutants over the Himalayas to inform possible mitigation strategies are limited by the scarcity of robust knowledge about emission sources. Emissions sources are uncertain not only over mountainous regions but also over urban and rural areas, especially in the HKH region, because of the lack of coordinated and readily available energy use and consumption data. For example, many experimental studies (Carrico et al 2003; Stone et al 2010) cannot fully account for observed concentrations with known emission source categories. Zhang et al (2009) provided a detailed gridded emissions inventory, which was developed for the NASA INTEX-B mission (2006) for all of Asia. The inventory is widely used for studies related to Asia and is one of the best available. In this study, the authors point out the uncertainties related to the emissions of pollutants. For example, for China, uncertainties in emissions are  $\pm 70\%$  for  $\text{CO}$ ,  $\pm 132\%$  for  $\text{PM}_{10}$ ,  $\pm 130\%$  for  $\text{PM}_{2.5}$ ,  $\pm 208\%$  for BC, and  $\pm 258\%$  for OC. The same authors had previously estimated emission inventory in support of another NASA (TRACE-P) mission for

the year 2000 (Streets, Bond, et al 2003; Streets, Yarber, et al 2003). The emissions uncertainty for all of Asia reported then was  $\pm 185\%$  (CO),  $\pm 360\%$  (BC), and  $\pm 450\%$  (OC). In a span of 6 years, the emissions estimate changed substantially for different regions within Asia—for example, BC estimates decreased by 18% over South Asia and increased by 127% over East Asia. The authors argued that the changes in emissions not only are a result of actual growth or reduction but are also due to improvements in the emissions inventory process (such as improved technical knowledge and replacement with local emissions inventories).

Apart from India, emissions inventories for the countries of the HKH region are either nonexistent or not readily available. It is crucial that these local bottom-up inventories be prepared so that effective control strategies can be implemented. While the problem of CO<sub>2</sub>-related warming is global in nature and requires multicountry, multistakeholder negotiations, issues related to regional air quality and regional climate and other co-benefits such as human health—for example, the issues induced by BC—can be effectively tackled on a regional scale.

### Summary and recommendations

On average, air pollution levels in the HKH region are significantly lower than those observed in urban and industrialized areas of South Asia. However, especially during the South Asian dry season, this very sensitive region can be systematically affected by transport of anthropogenic climate-altering pollutants like BC and O<sub>3</sub>. This is clearly reflected by the presence of pollutants in the HKH cryosphere as well as by the atmospheric mass concentration of aerosols, which are more elevated than they are pristine locations in Antarctica, the Arctic, or the remote marine boundary layer (see Hyvarinen et al 2009; Decesari et al

2010; Marinoni et al 2010; Stone et al 2010). In particular, the few existing continuous atmospheric observations reveal that surface O<sub>3</sub>, BC and aerosol particles in the high Himalayas are characterized by higher concentrations during winter and spring (the pre-monsoon season), minimal concentrations during the summer wet monsoon season, and increasing levels during the autumn (post-monsoon season).

Even if the surface O<sub>3</sub> observed during non-monsoon seasons can be also related to natural processes like stratosphere-to-troposphere air-mass exchanges (eg Cristofanelli et al 2010; Kumar et al 2010), long-range and regional-scale pollution strongly contribute to the occurrence of high levels of O<sub>3</sub> and aerosol particles in the HKH region. Favorable meteorological conditions coupled with large emissions from fossil fuel combustion, biomass burning, and biofuel cooking and heating favor the appearance of an ABC extending from the Indian Ocean to the Himalayas during the dry season. Due to conducive atmospheric circulation, this pollution can be transported toward the Himalayas and vented along the mountain valleys, thus directly impacting mountain ecosystems. Polluted air masses originating from far westerly regions like Northern Africa, the Middle East, and Europe can further contribute to determining the variability of anthropogenic pollutants over HKH.

This leads to a number of issues concerning the possible impacts of anthropogenic emissions on air quality and climate in this critical region. In particular, high amounts of BC and O<sub>3</sub> can be effectively transported up to the high Himalayas. Both are short-lived climate-altering compounds and pollutants. O<sub>3</sub> is the third greenhouse gas in terms of direct anthropogenic radiative forcing, and BC is estimated to have a global radiative effect that exceeds that of methane, and thus they both represent important

contributions to the observed global warming. By interfering with solar radiation, atmospheric BC can also modify the surface energy and the atmospheric temperature lapse rate on regional scales, thus impacting hydrological cycles and precipitation patterns.

Moreover, both BC and O<sub>3</sub> have significant toxic effects on human health and ecosystems. In particular, they can significantly impact terrestrial vegetation by affecting leaf structure and physiology as well as plant growth and crop productivity in South Asia (Agrawal et al 2008). A preliminary investigation has pointed out that plant productivity may decrease up to 20% in some HKH regions due to future tropospheric O<sub>3</sub> increases (Royal Society 2008). As shown by recent studies, the presence of BC and soot particles in the HKH atmosphere can modify the atmospheric and surface energy balance, altering atmospheric circulation, precipitation patterns, cloud coverage, snowmelt rates, and snow spatial coverage. This can drive significant changes in glacier shrinking and snow melting, possibly also influencing river regimes related to glacier and snow discharge.

Due to their relatively short atmospheric lifetimes, BC and O<sub>3</sub> emission reductions should be very effective in reducing the anthropogenic impact on climate. In fact, because BC and O<sub>3</sub> remain in the atmosphere for weeks to months, instead of centuries like other greenhouse gases such as CO<sub>2</sub>, their reduction provides a highly climatically effective mitigation option. Therefore, it is very important that governments together with local and international agencies develop appropriate policies to promote significant reduction of emissions of pollutants and greenhouse gases. Focused efforts to tackle reductions in BC emissions and ozone precursor gases could be an effective immediate action plan.

Concerning the HKH, the introduction of more effective

technologies for domestic heating and cooking can be very effective in reducing the absorption of aerosol directly emitted into the atmosphere or O<sub>3</sub> precursors. This goal can be reached by spreading the use of renewable energy (such as thermal solar energy) or by adopting more efficient cooking and heating systems that limit emissions. In the industrial sector, primitive brick kilns are still operating in huge numbers in South Asia and could be improved with existing technologies to reduce the BC/OC emissions arising from them. In the transportation sector, existing technology such as the diesel particulate filter could be made mandatory in the region to reduce emissions from diesel vehicles. Open biomass burning for agriculture purposes is another major source of pollutants and climate-altering compounds in this region (Gustafsson et al 2009). Limiting or regulating this kind of practice can lead to very effective reduction of anthropogenic impacts on the regional climate and air quality.

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