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Atmospheric aerosol brown carbon in the high Himalayas

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Anthropogenic light-absorbing atmospheric aerosol can reach very high concentrations in the planetary boundary layer in South-East Asia ("brown clouds"), affecting atmospheric transparency and generating spatial gradients of temperature over land with a possible impact on atmospheric dynamics and monsoon circulation. Besides black carbon (BC), an important light-absorbing component of anthropogenic aerosols is the organic carbon component known as 'brown carbon' (BrC). In this research, we provided first measurements of atmospheric aerosol BrC in the high Himalayas during different seasons. Aerosol sampling was conducted at the GAW-WMO Global station "Nepal Climate Observatory-Pyramid" (NCO-P) located in the high Khumbu valley at 5079 m a.s.l. in the foothills of Mt. Everest. PM_{10} aerosol samples were collected from July 2013 to November 2014. The sampling strategy was set up in order to discriminate the daytime valley breeze bringing polluted air masses up to the observatory and free tropospheric air during nighttime. Water-soluble BrC (WS-BrC) and methanol-soluble BrC (MeS-BrC) were extracted and analyzed using a UV/VIS spectrophotometer equipped with a 50 cm liquid waveguide capillary cell. In the polluted air masses, the highest levels of the BrC light absorption coefficient at 365 nm (b_{abs365}) were observed during the pre-monsoon season (1.83±1.46 Mm^{-1} for WS-BrC and 2.86±2.49 Mm^{-1} for MeS-BrC) and the lowest during the monsoon season (0.21±0.22 Mm^{-1} for WS-BrC and 0.32±0.29 Mm^{-1} for MeS-BrC). The pre-monsoon season is the most frequently influenced by a strong atmospheric brown cloud (ABC) transport to NCO-P due to increased convection and mixing layer height over South Asia combined with the highest up-valley wind speed and the increase of the emissions from open fires due to the agricultural practice along the Himalayas foothills and the Indo-Gangetic Plain. In contrast, the monsoon season is characterized by a weakened valley wind regime and an efficient removal of aerosols by wet scavenging. The wavelength dependence of the light absorption by BrC expressed as Ångström Exponent (AAE) within 330-500 nm was on average lower for MeS-BrC (3.9±1.1) compared to WS-BrC (4.8±0.8) and exhibited no diurnal or seasonal trend. The light absorption coefficient of BrC at 365 nm was about 13-17% (WS-BrC) and about 21-29% (MeS-BrC) of that of BC (AAE_{BC}=1). Relative light absorption of BrC and BC considering the whole solar spectrum showed that at NCO-P WS-BrC absorbs 5±2% and MeS-BrC absorbs 12±7% compared to equivalent BC, as measured by Absorption Photometer (MAAP). These results are in line with previous in situ measurements at low altitude stations in South-East Asia, and do not support the strong enhancements of brown carbon absorption contribution in the upper part of the boundary layer and in the free troposphere suggested by remote sensing observations.