Insights from about two decades of ground and space-based aerosol observations over the Central Himalayas

Priyanka Srivastava*(1), Manish Naja(1), Hema Joshi(2), T.R. Seshadri(3), Mukunda M Gogoi(4), S. Suresh Babu(4), P. Bhardwaj(5), Rajesh Kumar(6)

(1) Aryabhatta Research Institute of Observational Sciences (ARIES), Nainital, India; e-mail: srivastava.priyanka.res@gmail.com

(2) Indian Institute of Technology, Kanpur, Uttar Pradesh, India.

(3) Space Physics Laboratory, Vikram Sarabhai Space Centre, Thiruvananthapuram, Kerala, India.

(4) Center for Study of Science, Technology and Policy (CSTEP), Bengaluru, India

(5) National Center for Atmospheric Research (NCAR), Boulder, USA

Abstract

Satellite observations are being increasingly utilised over the Himalayas where in-situ aerosol observations are limited due to complex terrain and logistic challenges. While past studies have done short-term validation of these satellites, long-term comparisons are rare, especially in view of sensor degradation over time and the relatively low levels of aerosols in the Himalayas. To this end, we utilise the foremost ~two decades of aerosol in-situ observations over the Central Himalayas. The findings reveal a statistically significant decreasing trend in mass-absorption corrected surface black carbon (BC) concentrations during 2004-2020. Piecewise trend analysis unveils a reversal in BC trend from positive to negative after 2017 due to a decline in fossil fuel fraction. This contrasts with an overall rising trend observed in columnar aerosols from MODIS and CALIOP. Emissions of BC from bottom-up inventories and aerosols from CAMS and MERRA2 also showed positive trends opposite to those of surface observations. The trend in vertical aerosol profiles from CALIOP and POLDER reveals an elevated aerosol layer at around 2-4 km a.m.s.l. around the region which could be a potential answer to the observed discrepancy in the surface and columnar aerosol measurements. The role of anthropogenic emissions and biomass burning in this declining trend is also analysed. These observations thus provide a unique long-term characterization of aerosols over the Himalayas used to test and validate satellite and model results, and can be further used to ascertain the impact of aerosols in the Himalayas.

1. Introduction

The Himalayas are facing consequences of the ongoing climate change such as rising temperatures, extreme precipitation, glacier retreat, thawing, and reduced snow cover [1]. Anthropogenic greenhouse gases and aerosols are known to be the key drivers behind this climate change. Aerosols remain the single largest source of uncertainty [2] in this analysis. In particular, Black Carbon (BC) aerosols, known for being the most absorbing aerosol species, are associated with significant regional climatic implications over the Himalayas apart from their health-related hazards [3]. Therefore, it is essential to accurately quantify the contribution of different sources to the aerosols over the Himalayas. It is also important because the Himalayas lie in close proximity to the world's most polluted regions of the Indo-Gangetic Plains. Further, while emissions of BC and other pollutants over the USA, Europe, and mid-latitudes have been reported to decrease, those over the developing Asian countries are reportedly increasing [4]. Most of the trend (>10 years) based studies over Asia, particularly in South Asia, are either from model simulations or from satellite measurements. However, these observations are not free from uncertainties and need to be compared against ground-based observations. While past studies have done short-term validation of these satellite products, long-term comparison studies are rare especially because of the sensor degradation over time and given the relatively low levels of aerosols in the Himalayas. Yet only limited datasets from ground-based observations are available over this region and even such observations are either representative of a short period (less than a few years) or lack continuity. To bridge the aforementioned gap, we utilise the foremost ~two decades (2004-2020) of aerosol in-situ observations made in an otherwise sparsely studied complex terrain of the Central Himalayas (ARIES, Nainital, 29.4ºN, 79.5ºE, 1958 a.m.s.l.). We analyse the long-term trends in observed BC and Aerosol Optical

Depth (AOD) and compare the former against aerosol parameters from MODIS (Moderate Resolution Imaging Spectroradiometer), OMI (Ozone Monitoring Instrument), CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarisation), POLDER (Polarization and Directionality of the Earth's Reflectances), CAMS (Copernicus Atmospheric Monitoring Services) and MERRA2 (Modern-Era Retrospective Analysis for Research and Applications", Version 2). We highlight the contrast between the trends observed from surface and columnar measurements and between satellite/models against ground measurements. We also utilise emissions inventories, delineate BC sources and analyse the satellite-derived vertical profile to answer the contrasts observed in the surface and columnar measurements.

2. Observations and Methodology

Region: Measurements have been made at a high-altitude site in the Central Himalayas (ARIES, Nainital, 29.4ºN, 79.5ºE, 1958 a.m.s.l.). The region is away from major industries or metropolitans and is a popular tourist spot due to its cool climatology [5].

In-situ measurements: A seven-wavelength Aethalometer (AE 42) is employed for the measurement of eBC from 2004-2020 at the site. Importantly, the site-specific multi-wavelength monthly mass absorption cross-section (MAC) derived from long-term measurements of elemental carbon at the site is used to convert the absorption coefficient to eBC concentrations. Further details are available in [5]. AOD measurements are made using the handheld, multiband MICROTOPS-II sunphotometer at five wavelengths [6].

Satellite data: We have assessed the trend in the aerosol exponent (AE) and AOD using daily Level 3 MODIS Collection 6.1 combined dark target (for dark surfaces) and deep blue (for bright surfaces) data along with active fire data until 2020 over a 1°×1° region around the site. OMI Single Scattering Albedo (SSA) and UV Aerosol Index (AI) are also utilised for the same period and region. We use aerosol vertical profile (variation of extinction coefficient, depolarization ratio and aerosol type with height) from the level 2 version 4 profile products (5 km horizontal resolution) of CALIOP with appropriate quality flags (not described here for brevity) to get high-confidence data. The AOD of the integrated CALIOP profile is also used for analysing the trend. We also utilised aerosol layer height data v2.1 derived from the POLDER GRASP algorithm over the site.

Models and inventory: MERRA-2 provides data at 0.5°×0.625° resolution at 72 hybrid-eta levels starting from the surface to 0.01 hPa. In this work, we have utilised MERRA-2 reanalysis surface BC data from 2004–2020 at a 1-degree region around the site.

CAMS global reanalysis EAC4 (ECMWF Atmospheric Composition Reanalysis 4) model provides global reanalysis aerosol data at 0.75°x0.75° resolution using the 4D-Var assimilation method. Here we utilise 3 hourly, surface, columnar and multi-level data (100 to 850 hPa) of dust (total: 0.03 to 20 µm), sea salt (total: 0.03 to 20 µm), BC (hydrophilic+hydrophobic), hydrophilic organic matter and sulphate. Data from EDGARv6.1 (Emissions Database for Global Atmospheric Research) BC emissions inventory available at 0.1°x0.1° degree for the countries surrounding Himalayas (China, India, Pakistan, Nepal, Afghanistan and Bangladesh) and different sectors emissions within Uttarakhand is also used.

3. Results and Discussions

3.1 Trends in Surface and Columnar Aerosols

Trends in the observed BC, and BC from CAMS and MERRA-2 from 2004 to 2020 are analysed (Figure 1). We find a statistically significant, negative trend with a slope of –19.911 ng m^{-3} $year^{-1}$ in the observations while both MERRA2 and CAMS show a positive trend (with both nearest neighbour and bilinear interpolation approaches). Analysis of seasonal and diurnal variations in MERRA2 BC data shows that it misses the daytime high and spring high observed at the site suggesting that these models need to be used with care at the Himalayas. Mann-Kendall test results show a similar declining trend in BC for the four seasons, noon and night time and extreme events which are found to decline faster. Therefore, there is an overall negative trend over the central Himalayas, yet the trend is much lower than in other parts of India (-242 ng m^{-3} $year^{-1}$) [6]. The trend in AOD from both ground-based and satellite sensors CALIOP and MODIS is also analysed with the Mann-Kendall test and it is found to be positive and significant (Figure 1).

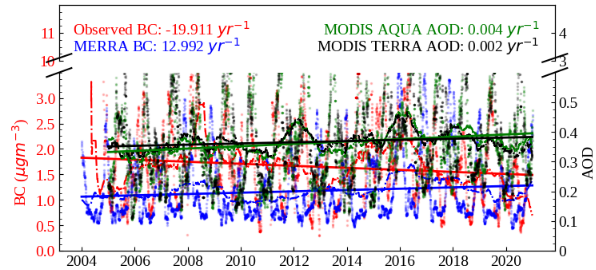

Figure 1. Trend in BC (Observed and MERRA2) and AOD (MODIS AQUA and TERRA). Scattered points are the monthly rolling mean values and curved lines are over 365-days moving averages. The linear fit is estimated using fit to the 365-days moving average.

Satellite observations, however, do not capture the observed seasonal variation in optical depth at the site. Notably, we found a negative trend in surface BC, thus suggesting that the overall columnar aerosol content is rising but the surface BC fraction is declining.

3.2 Why the Contrasting Trends – Elevated Aerosols?

Two possible hypotheses for these contradictory trends in BC and AOD could be, first - a decline in surface BC concentrations with a subsequent rise in the higher altitudes which leads to an escalation of the AOD and, second - a decline in BC with a simultaneous rise in other aerosol species at different heights.

For testing the first hypothesis of a rise in the aerosol concentrations at higher altitudes, we analysed the aerosol layer height from POLDER, extinction coefficient, and frequency of occurrences (Figure 2) of five aerosol types (dust, polluted dust, clean continental, polluted continental and smoke) using CALIOP. All the results consistently reveal the presence of an elevated aerosol layer in ~2-4 km region. Further, the trend in AE from MODIS and AI from OMI is found to be rising while that of SSA from OMI is declining. The rising levels of PDR above 1.5km from CALIOP also support the analysis. All this indicates a possibility that anthropogenic aerosols such as black carbon might be reducing in the lower altitudes while they are building at higher altitudes. Other studies have also suggested the presence of elevated layers over South Asia [7]. However, the observed trends are not significant according to the Mann-Kendall test. Therefore, licensing deeper analysis before any concrete decisions can be drawn.

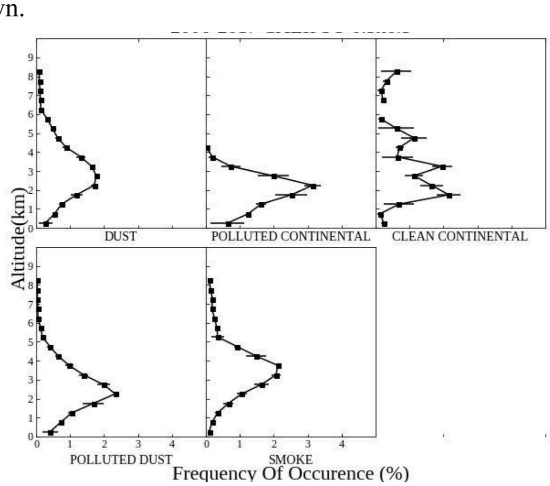

Figure 2. Frequency of occurrence (%) in CALIOP aerosol profiles.

We tested the second hypothesis of a decline in BC with a simultaneous rise in other aerosol species at different heights by analysing the trend in different aerosols from CAMS. We found that dust and sea salt showed a significant decline over all the levels. However, CAMS BC showed a rise in aerosols at the surface and an insignificant trend at upper levels. In contrast, organic matter (Figure 3) and sulphates showed a significant positive trend. Thus, lending credibility to the hypothesis that these aerosols may be growing at higher levels. Yet it is to be noted that the CAMS trend (+21.63 ng m^{-3} $year^{-1}$) did not compare well with the surface BC trend at the site. Therefore, more in-situ observations are needed to draw any conclusions.

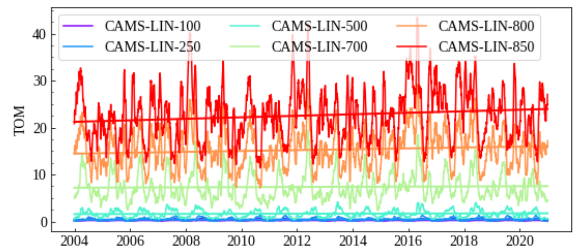

Figure 3. The trend in CAMS total organic matter (TOM) at different altitudes (hPa) in legend.

3.3 Role of fires and anthropogenic emissions in the observed trends in BC and AOD

BC concentrations were segregated into fossil fuel and biomass fractions following [5]. The fossil fuel fraction was found to dominate by about 76% in the ambient BC fraction. The trend analysis reveals a greater decline in the fossil fuel burning rate (-20.29 ng m^{-3} $year^{-1}$) compared to the biomass burning BC fraction (-3.77 ng m^{-3} $year^{-1}$) implying that the decrease in BC is largely attributed to the decline in the fossil fuel combustion fraction. We further investigate the trend in anthropogenic BC emissions from the EDGARv6.1 for countries bounding the Himalayas (Figure 4). It is found that there is a significant positive trend in BC emissions in all six countries. China has the highest BC emissions, followed by India. BC emissions from China have shown a decline since 2013. Further, while the emissions in China are observed to be declining, those in India are still on the rise, though at a slower rate. Furthermore, the trend in observed BC till the year 2016 has a significant and positive slope of 27.77 ng m^{-3} $year^{-1}$. The trend reversed thereafter with a negative slope of -138 ng m^{-3} $year^{-1}$ from 2017 to 2020. Similar features are observed in the fossil fuel and biomass BC fractions. This indicates that the trend in BC has reversed from positive to negative in 2017 possibly as a result of stringent control policies imposed. The results also bring out the importance of mitigation strategies and the continuous ground-based monitoring of these aerosols in the ever-changing emission scenario.

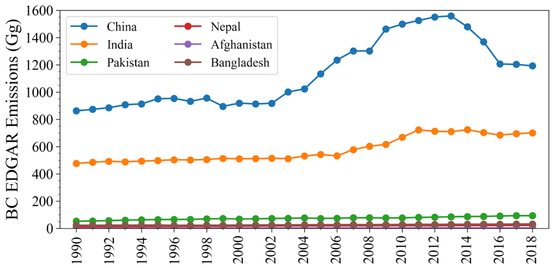

Figure 4. BC Emissions estimates from EDGARv6 for countries surrounding the Himalayas.

4. Summary and Conclusions

This work integrated about two decades of ground, model and space-based observations and attempts to fill the gap in our understanding of aerosols over the complex and important region of the Himalayas. It shows a statistically significant negative trend (–19.911 ng m^{-3} $year^{-1}$) in BC led by a faster reduction in fossil fuel fraction. This decline is still lower than the BC decline reported in other Indian regions. A reversal in BC trend from positive to negative since 2017 and a declining fossil fuel fraction points to a relative success of the control policies imposed. However, more work is needed to increase the rate of decline, especially for biomass emissions. BC from emissions inventories, CAMS and MERRA2 fail to reproduce this decline and depict a rise in BC instead thereby raising questions about the studies using them for drawing conclusions over the region, simultaneously highlighting the importance of continuous ground monitoring. CALIOP and MODIS reproduce the AOD rising trend. Vertical aerosol profiles from satellites and models are used to explain this contrast in decline in surface BC and rising columnar aerosols. Aerosols observed around the 2-4 km layer and rising aerosol trend in upper layers from CAMS points that aerosols might be building in the higher altitudes. More detailed in-situ measurements in future will be crucial to test this hypothesis.

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