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Altitude-Dependent Decline in Sulphate Aerosol Optical Depth and Its Role in Warming Across the Northwestern Himalayas

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Abstract*— Significant warming has been observed in the Northwestern Himalaya (NWH) in recent decades, but little is known about sulphate aerosols, a key cooling agent that affects regional climate. This study uses sulphate aerosol optical depth (SO_4 AOD) MERRA-2 reanalysis data from 1980 to 2023 to investigate altitude-dependent patterns and their relationships to regional warming. Trend analysis reveals a clear SO_4 AOD decline at high elevations and a modest increase in the foothills. Spatial mapping shows the strongest negative trends along the mountain front, with weaker or positive trends in lower valleys. The high-altitude decline is attributed to the absence of local sulphate sources and inefficient vertical transport of hygroscopic sulphate particles. Under humid boundary-layer conditions, these aerosols absorb water, increasing mass and settling velocity, thereby hindering their ascent across steep Himalayan terrain. Additionally, lower relative humidity at higher altitudes further decreases aerosol growth and scattering. Trends in black carbon and dust AOD are also examined. The decrease in sulphate cooling, combined with stable or increasing warming agents, likely contributes to accelerated warming in the region. These data-driven insights clarify how aerosol distribution and mountain meteorology together influence climate change across the NWH.

Keywords: Aerosol Optical Depth, Northwestern Himalaya, Sulphate Aerosols, Black Carbon, Dust, MERRA-2, Radiative Forcing, Climate Change.

I. INTRODUCTION

Aerosols - fine particles suspended in the atmosphere — significantly influence the climate system through direct radiative effects (scattering and absorption of solar radiation) and indirect effects (modification of cloud properties via cloud condensation nuclei). The Himalayan region, particularly the Northwestern Himalayas (NWH), is especially vulnerable to aerosol-driven climate impacts owing to its proximity to the major anthropogenic pollution sources of South Asia and the long-range transport of pollutants from the Indo-Gangetic Plain (IGP) via enhanced convection and steep pressure gradients (Gautam et al., 2009; Decesari et al., 2010; Zhang et al., 2017). These aerosol loadings, compounded by rising surface temperatures, have accelerated glacier retreat and altered precipitation patterns across the region (Gautam et al., 2013; Krishnan et al., 2019).

This study focuses on two contrasting aerosol types. Black carbon (BC) and mineral dust (DU), both light-absorbing species, enhance atmospheric radiative forcing, reduce snow albedo, and accelerate snowmelt (Ming et al., 2012; Sharma et al., 2022), with BC recognised as the second-largest contributor to global warming after CO_2 (Jacobson et al., 2001). In contrast, sulphate (SO_4) aerosols scatter incoming solar radiation and enhance cloud albedo, producing a net cooling effect (Charlson et al., 1992; Kulmala et al., 2000). However, recent reductions in SO_2 emissions have begun to unmask underlying greenhouse gas warming, with studies quantifying an associated temperature increase of 0.1 -- 0.2°C (Stohl et al., 2023). Notably, aerosols account for more than 50% of total lower atmospheric warming over the Himalayas, with radiative forcing efficiency over the IGP and Himalayan foothills estimated at 80–135 W m^{-2} per unit AOD (Ramachandran et al., 2023).

Elevation-dependent warming (EDW) — the tendency for warming to intensify with altitude — has been documented across the Himalayas and the Tibetan Plateau (Pepin et al., 2015; You et al., 2020). Multiple mechanisms drive EDW, including changes in snow and ice albedo, cloud feedback, water vapour concentration, and aerosol loading. Despite growing evidence for EDW, the role of aerosol species in modulating elevation-specific warming remains underexplored. In particular, the altitude-dependent behaviour of sulphate aerosols and their contribution to differential radiative forcing across the NWH has not been adequately characterised.

Despite growing research, the sources, transport pathways, and climatic implications of these aerosol species over the NWH remain insufficiently characterized. This study aims to address this gap by examining the spatiotemporal variability and radiative impacts of BC, DU, and SO_4 aerosols over the region. Specifically, we (i) quantify long-term (1980–2023) trends in sulphate, black carbon, and dust AOD at contrasting elevations; (ii) identify the physical and meteorological mechanisms responsible for altitude-dependent aerosol distributions; and (iii) assess the climatic implications of

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observed aerosol trends for regional temperature change.

The remainder of the paper is organised as follows: Section 2 describes the study area; Section 3 outlines the data and methodology; Section 4 presents and discusses the key findings; Section 5 addresses the broader climatic implications; and Section 6 provides concluding remarks.

II. STUDY AREA

This study focuses on the Northwestern Himalaya (NWH), encompassing the states of Uttarakhand (U.K.), Himachal Pradesh (H.P.), and Jammu & Kashmir (J&K). Geographically, the study domain spans latitudes 28.43° N to 37.05° N and longitudes 72.40° E to 81.02° E (Figure 1). The region is characterised by highly diverse terrain, ranging from subtropical foothills and river valleys to cold deserts and high-altitude glaciated zones, making it particularly sensitive to aerosol-climate interactions across elevation gradients.

The NWH represents one of the most topographically complex regions on Earth, with elevations ranging from approximately 200 m in the Himalayan foothills to above 8,000 m in the higher ranges. This dramatic relief creates strong orographic forcing, distinct vertical atmospheric layers, and sharp microclimatic contrasts over short horizontal distances. The cold-desert environments of Ladakh and Spiti, situated at elevations exceeding 3,500 m, receive minimal precipitation and are dominated by arid, dust-prone landscapes. In contrast, the sub-Himalayan foothills receive significant monsoonal precipitation and are heavily influenced by anthropogenic aerosol emissions from the adjacent IGP.

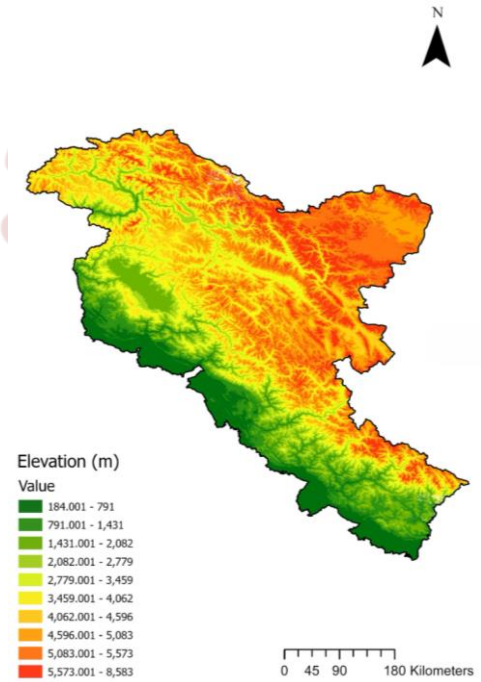


Figure 1. Map of the Northwestern Himalaya study region with elevation gradient

The region is further characterised by distinct seasonal circulation regimes. During winter (December–February), the NWH is influenced by western disturbances that bring precipitation to higher elevations. Summer (June–August) is dominated by the South Asian Monsoon, which delivers moisture from the Bay of Bengal and the Arabian Sea. These seasonal shifts in circulation exert strong controls on aerosol transport pathways, residence times, and optical properties. The proximity of the NWH to the heavily industrialised and densely populated IGP makes it an ideal laboratory for studying the climatic impacts of long-range aerosol transport.

III. DATA AND METHODOLOGY

Aerosol Optical Depth (AOD)

This study uses aerosol optical depth (AOD) as the primary metric, which quantifies the total extinction (scattering + absorption) of solar radiation by aerosols integrated over a vertical atmospheric column. AOD provides an effective proxy for aerosol loading and has been widely used to examine long-term annual and seasonal trends at regional and global scales. Higher AOD values indicate denser aerosol loading and greater attenuation of incoming solar radiation. For cooling aerosols such as sulphates, elevated AOD implies stronger negative radiative forcing, while for absorbing aerosols such as black carbon, higher AOD corresponds to greater positive radiative forcing.

MERRA-2 Reanalysis Data

This study employs MERRA-2 reanalysis data — NASA's second-generation aerosol reanalysis product — which uses the Goddard Earth Observing System, version 5 (GEOS-5), to assimilate multiple aerosol species (Buchard et al., 2017). MERRA-2 AOD data have demonstrated strong agreement with ground-based AERONET observations, with a global correlation coefficient of 0.85 and a root mean square error of 0.12 (Che et al., 2019). The MERRA-2 model incorporates SO_2 emissions from biomass burning, volcanic eruptions, and dimethyl sulphide (DMS) as sulphate aerosol precursors. Black carbon and organic carbon aerosols are primarily driven by fossil fuel combustion and biomass burning, while dust aerosols are generated by wind erosion over arid and semi-arid surfaces (Buchard et al., 2017). The datasets used in this study are summarised in Table 1.

Table 1. MERRA-2 datasets used in this study

Sensor	Variable	Resolution (lat × lon)	Period
MERRA-2 (550 nm)	Black Carbon AOD Sulfate AOD Dust AOD (PM 2.5) 2-metre Air Temperature	0.5° × 0.625°	1980–2023

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Trend Analysis

Long-term trends in Aerosol Optical Depth (AOD) and near-surface air temperature were quantified using **Ordinary Least Squares (OLS) linear regression**, applied independently for each **season–variable–location** combination.

Spatial Analysis and Wind Vector Overlay

Spatial distributions of monthly and seasonal mean AOD were mapped across the NWH domain using gridded MERRA-2 output at 0.5° × 0.625° resolution. Wind vectors derived from MERRA-2 u- and v-component surface wind fields were overlaid on AOD maps to identify the dominant aerosol transport pathways. Altitude-stratified analysis was conducted by classifying MERRA-2 grid cells into elevation bands using a digital elevation model (DEM), enabling the quantification of aerosol optical depth as a function of terrain height. Elevation bands were defined as: foothills (< 1,000 m), mid-elevation (1,000–3,000 m), high-elevation (3,000–5,000 m), and very high-elevation (> 5,000 m).

Relative Humidity and Aerosol Hygroscopic Growth

To investigate the role of hygroscopic growth in altitude-dependent sulphate transport, monthly MERRA-2 relative humidity (RH) profiles were extracted for the study domain and analysed as a function of elevation. Sulphate aerosols are highly hygroscopic and undergo significant growth at elevated RH, which increases particle mass, settling velocity, and gravitational deposition rates. The relationship between RH gradients, aerosol physical properties, and vertical AOD profiles is examined qualitatively in Section 4.3. This analysis provides a process-level mechanistic explanation for the observed altitude-dependent SO_4 AOD decline.

IV. RESULTS AND DISCUSSION

Seasonal Variation of AOD and Near-Surface Temperature

The seasonal time series analysis reveals distinct altitude-dependent aerosol behaviour across the NWH. At cold-desert and high-valley sites in the upper NWH, winter and spring warming is most pronounced, while summer and autumn warming, though positive, is comparatively weaker. Correspondingly, SO_4 AOD exhibits its largest declines during winter and spring — consistent with the region-wide sulphate reduction trend — while summer and autumn declines remain marginal (Figure 2).

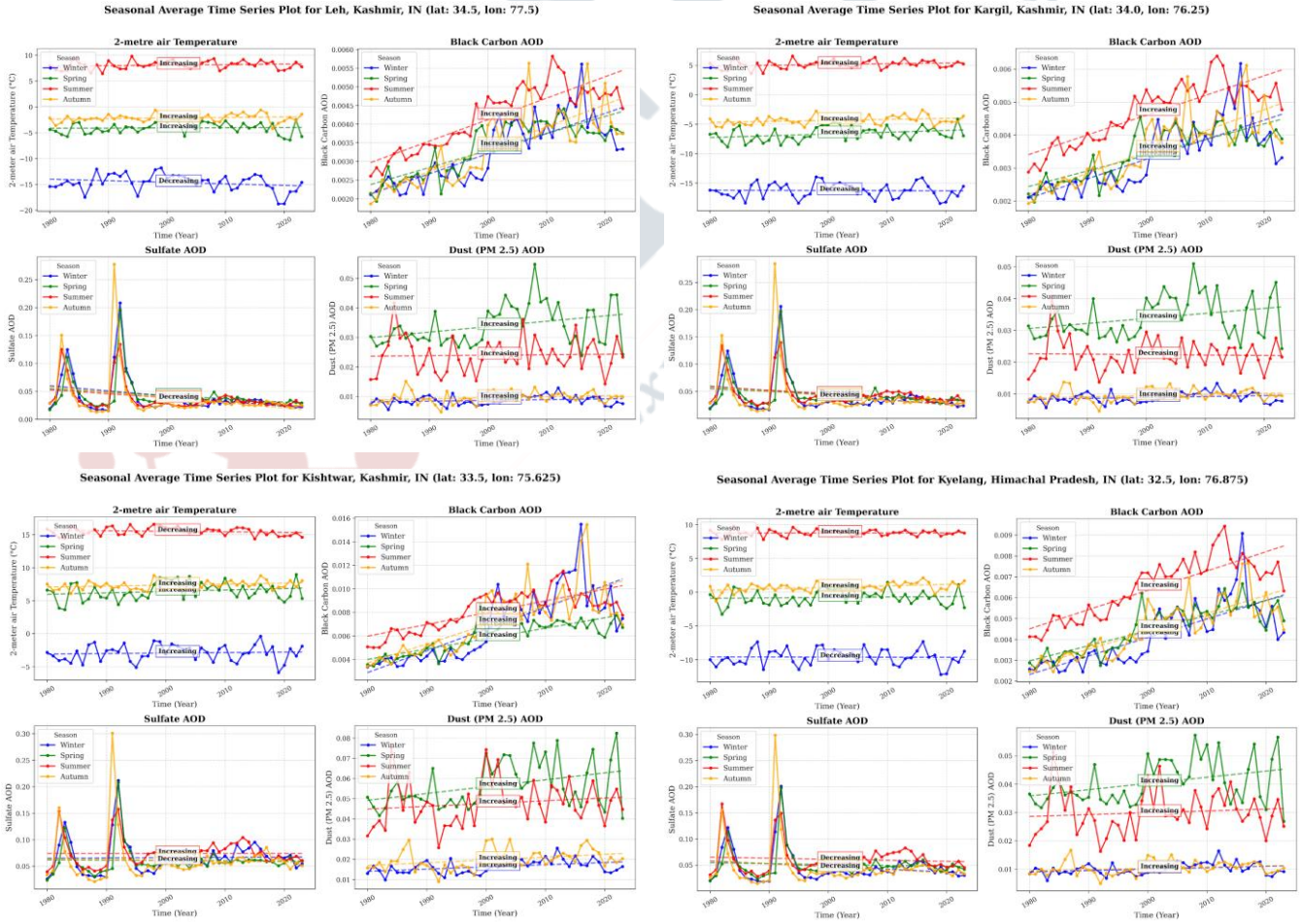


Figure 2. Seasonal average time series plots with trend lines for selected upper NWH locations

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BC AOD increases notably in summer and autumn at these high-altitude sites, suggesting enhanced long-range transport or a rise in local emission activity. DU AOD shows a characteristic springtime peak associated with seasonal dust storms over arid source regions (El-Askary et al., 2006; Badarinath et al., 2007; Gautam et al., 2009, 2013).

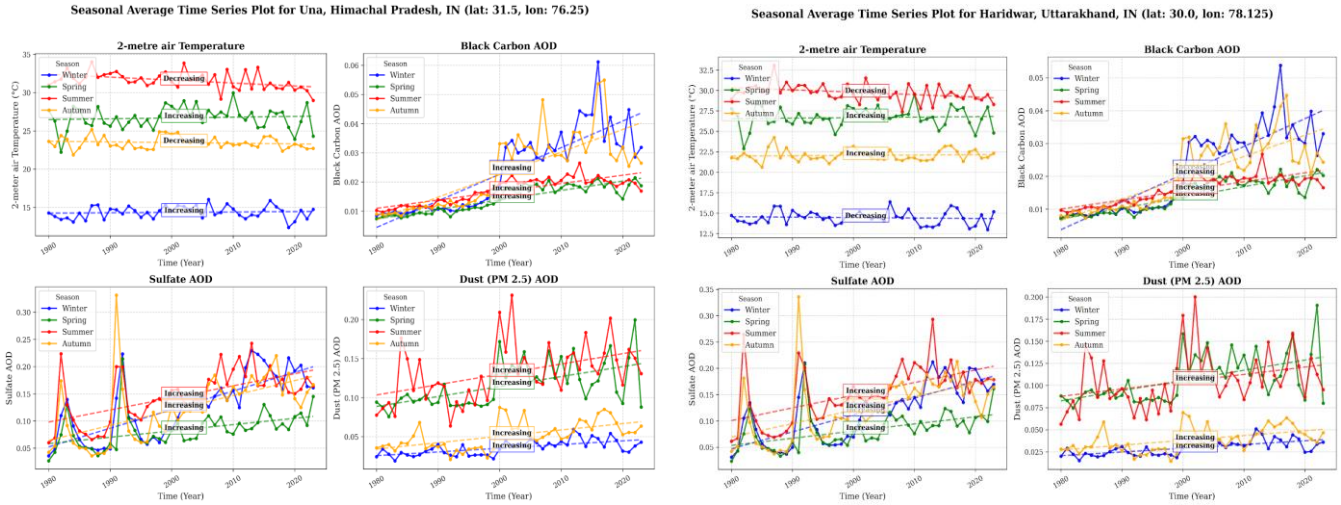


Figure 3. Seasonal average time series plots with trend lines for selected lower NWH (foothill) locations

In contrast, at foothill sites in the lower NWH, SO_4 AOD is increasing, with the steepest rise observed in summer and autumn. Summers here exhibit slight cooling, likely attributable to sulphate-driven scattering, while winter, spring, and autumn show moderate warming that correlates with elevated BC and DU loadings in those respective seasons (Figure 3).

Spatial Patterns and Wind-Driven Aerosol Transport

To examine spatial patterns of aerosol transport, monthly mean BC, SO_4, and DU AOD fields were overlaid with wind vectors (Figure 5a–c), revealing distinct seasonal signatures in aerosol loading across the NWH.

During winter (DJF), BC AOD peaks along the southern foothills (31°–32° N), with values exceeding 0.020, driven by northerly to northwesterly winds that transport pollutants uphill from the IGP under conditions of weak synoptic flow and wintertime atmospheric stagnation (Gautam et al., 2009). SO_4 AOD is also concentrated in foothill areas (AOD ≈ 0.100) but extends further north than BC, reflecting sulphate's longer atmospheric lifetime and the effective upward mixing of secondary aerosol precursors (Decesari et al., 2010). Dust AOD is generally low (< 0.025) in winter, with localised injection near 32° N from westerly winds carrying mineral dust from the Thar Desert.

During spring (MAM), both BC and DU AOD intensify, reaching approximately 0.023 and 0.045 respectively along the 30°–32° N latitudinal belt, driven by strong southwesterly winds advecting aerosols from Punjab and Rajasthan. SO_4 AOD also increases significantly across the NWH, reaching up to 0.135, attributed to rising regional SO_2 emissions and enhanced photochemical processing.

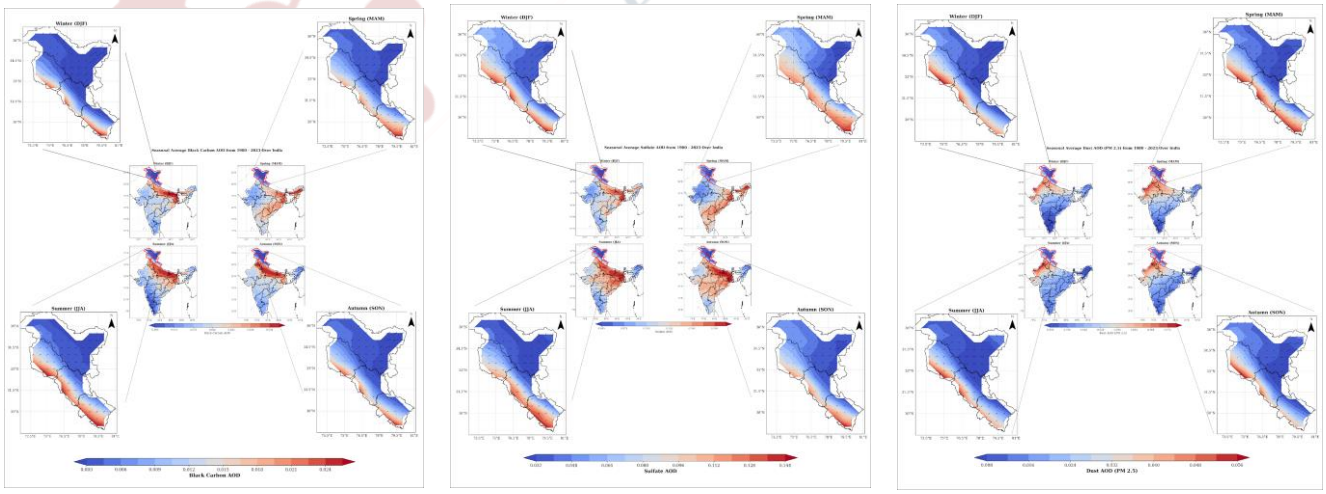


Figure 4(a–c). Spatial distribution of monthly mean (a) BC AOD, (b) SO_4 AOD, and (c) DU AOD (PM2.5) from 1980 to 2023, overlaid with wind vectors

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With the onset of the summer monsoon (JJA), BC AOD decreases to below 0.012 at mid-elevations due to a shift in prevailing wind trajectories, though a modest BC signal persists near the foothills from local emissions. SO_4 AOD peaks slightly (~ 0.115), sustained by enhanced in-cloud aqueous oxidation under moist monsoon conditions. DU AOD drops sharply to below 0.020 owing to efficient wet scavenging by monsoon precipitation.

During autumn (SON), post-monsoon northeasterly winds revive upslope transport, elevating both BC and DU AOD once more along the 31°–33° N belt.

Overall, BC AOD is increasing across the NWH due to a combination of local emissions and long-range IGP transport. Dust accumulation is further enhanced by the convergence of northwesterly and monsoon flows at the Inter-Tropical Discontinuity (ITD), a boundary where distinct air masses meet and create conditions for sustained aerosol loading — typically at altitudes of 1–3 km above ground level (Dumka et al., 2019).

Altitude-Dependent Decline in Sulphate AOD: Mechanistic Explanation

The pronounced decrease in SO_4 AOD at higher elevations can be explained by several interacting physical and chemical mechanisms. First, the NWH at high altitudes lacks significant local anthropogenic sulphur sources. Industrial activity, vehicular emissions, and fossil fuel combustion — the primary SO_2 precursors for secondary sulphate formation — are concentrated in the IGP and Himalayan foothills. In the absence of in-situ production, high-altitude sulphate loadings depend entirely on vertical and horizontal transport from lower levels.

Second, sulphate aerosols are highly hygroscopic. Under the humid boundary-layer conditions typical of the lower atmosphere, SO_4 particles absorb water vapour and undergo rapid growth, substantially increasing their mass and aerodynamic diameter. This hygroscopic growth elevates gravitational settling velocity and increases wet deposition rates, effectively limiting the vertical penetration of sulphate-laden air masses into the free troposphere above the steep Himalayan terrain. The orographic barrier presented by the NWH further impedes upslope transport by promoting precipitation scavenging along windward slopes.

Third, relative humidity decreases markedly with elevation across the NWH. At high altitudes, lower RH suppresses hygroscopic growth, reducing particle size and optical cross-section. While this counteracts the settling mechanism, it also diminishes the total scattering efficiency of residual sulphate particles that do reach higher elevations, further reducing observed SO_4 AOD.

Taken together, the combination of limited local SO_4 sources, hygroscopic growth and gravitational settling in the moist boundary layer, orographic scavenging, and reduced RH-driven optical growth at altitude produces a robust altitude-dependent decline in sulphate AOD across the NWH — a pattern that distinguishes sulphate from BC and DU, which are transported more efficiently to high elevations via convective uplift and long-range advection.

Long-Term Trends in Aerosol Optical Depth (1980–2023)

Analysis of the 44-year MERRA-2 record reveals contrasting long-term trends in aerosol loading across elevation zones. At high elevations (> 3,000 m), SO_4 AOD exhibits a statistically significant downward trend of –0.002 yr^{-1}, equivalent to a reduction of approximately 0.088 AOD units over the study period. This decline is most pronounced along the main Himalayan arc and in the cold-desert environments of Ladakh and Spiti, where background sulphate concentrations are already very low.

In contrast, SO_4 AOD in the foothill zone (< 1,000 m) shows a modest positive trend of +0.013 yr^{-1}, reflecting increasing SO_2 emissions from the expanding industrial and transport sectors of the northern IGP over the study period. This elevation-contrasting pattern — sulphate declining at altitude and rising at the foothills — implies a progressive divergence in the aerosol radiative forcing environment across the NWH elevation gradient.

BC AOD trends are positive across virtually all elevation bands, consistent with increasing fossil fuel combustion and biomass burning across South Asia. The strongest BC increases are observed at mid-elevations (1,000–3,000 m), likely reflecting the combined effects of upslope transport from the foothills and reduced scavenging relative to the precipitation-rich higher elevations. DU AOD trends are more spatially heterogeneous, with significant interannual variability driven by dust storm activity in the Thar Desert and the arid regions of western Asia.

V. CONCLUSION

This study has characterised the altitude-dependent distribution and long-term trends of sulphate, black carbon, and dust aerosol optical depth across the Northwestern Himalaya using 44 years (1980–2023) of MERRA-2 reanalysis data. The key findings are as follows:

A statistically significant decline in SO_4 AOD of –0.002 yr^{-1} is observed at high elevations (> 3,000 m), contrasting with a modest increase (+0.013 yr^{-1}) at the foothills. This elevation-contrasting sulphate pattern is a robust and novel finding of the present analysis.

The high-altitude SO_4 AOD decline is mechanistically attributed to the absence of local sulphate sources, hygroscopic growth and gravitational settling in the moist boundary layer, orographic precipitation scavenging, and reduced optical efficiency of residual sulphate particles at low relative humidity.

BC AOD is increasing across all elevation bands, with the strongest increases at mid-elevations, while DU AOD exhibits seasonally variable trends driven by dust storm activity over source regions.

The combination of declining sulphate cooling and increasing black carbon warming at high elevations contributes to accelerated warming in the NWH, consistent with and potentially reinforcing the observed elevation-dependent warming signal.

The implications of these aerosol trends for glacial melt, freshwater resources, and regional climate feedback are significant and underscore the importance of monitoring aerosol composition and loading at high-altitude sites.

These findings highlight the critical role of aerosol dynamics in mediating high-altitude climate change in the NWH. Future work should integrate ground-based AERONET measurements from high-altitude stations with satellite retrievals and reanalysis products to validate and extend the findings presented here. Model-based attribution studies are needed to quantify the relative contributions of aerosol forcing, greenhouse gas forcing, and natural variability to observed temperature trends across the NWH elevation gradient. Additionally, process-level studies examining aerosol hygroscopic growth under Himalayan orographic conditions would strengthen the mechanistic framework proposed in this study.

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Author Contributions:

Gaurav performed the analysis, produced all the results, and composed the preliminary manuscript. S.A. evaluated the paper, implemented requisite amendments, and confirmed its completion.

Data and Code Availability:

The authors can provide data and code upon request.

Conflict of Interest:

The authors declare no competing interests.

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