

Reviewers Response

Reviewer 3

Chandel et al. present observations of aerosol optical properties over the western Himalaya, with a particular focus on a selected dust-influenced period. The study combines in situ optical measurements, aerosol size distributions, black carbon measurements, and single-particle chemical analysis to characterize changes in aerosol properties during this period. The topic is potentially important because observations of aerosol optical properties in high-altitude Himalayan regions remain limited, and such measurements may be useful for evaluating the regional climatic effects of transported dust and pollution. However, I have substantial concerns about the current manuscript. In my view, the manuscript is difficult to follow, the interpretation is often stronger than what the data can support, and the logic is not sufficiently consistent throughout the paper. The measurements themselves may be valuable, but the present analysis and discussion do not yet provide a robust basis for the major conclusions. I therefore do not think the manuscript is suitable for publication in ACP in its current form.

We thank the reviewer for the thorough assessment of our manuscript. We have undertaken a fundamental revision that addresses each of the reviewer's comments.

Comment: My first major concern is that the authors appear to overstate the distinction between the background and dust-event periods. The event is defined using a relatively modest PM_{coarse} threshold, but the evidence for a strong dust event at the sampling location is not fully convincing. From the temporal evolution of PM_{coarse}, a more pronounced coarse-mode enhancement appears later, around 26 May–3 June, yet this period is not selected or discussed in sufficient detail. The authors should clearly justify why 18–24 May was chosen as the primary dust-event period instead of the later period with apparently higher PM₁₀ or PM_{coarse} levels. The MERRA-2 dust flux and CALIPSO information are used to support the selected period, but the connection between these regional-scale products and the actual surface measurements at the site needs to be made more quantitatively and cautiously. At present, the event selection appears somewhat arbitrary, and several of the subsequent conclusions depend strongly on this period definition.

Response: We thank the reviewer for this point. We have now revised Section 3.1 to provide a quantitative justification for the event selection.

We now write:

Line 204: Figure 2 illustrates the identification of a heavy dust loading episode observed during May 2023 and its subsequent characterization using surface measurements, reanalysis products, and satellite observations. The temporal variability of coarse particulate mass (PM_{coarse}), black carbon (BC), brown carbon (BrC) absorption, secondary brown carbon (Sec BrC) absorption, and their mass-normalized optical ratios is presented in Figure 2a–c. Figure 2a shows the

temporal evolution of the $\text{PM}_{\text{coarse}}$, which increased from 20–30 $\mu\text{g m}^{-3}$ during 11–17 May 2023 to 45–55 $\mu\text{g m}^{-3}$ during 18–24 May 2023. This approximately twofold increase in coarse-mode aerosol mass concentration is a characteristic signature of dust intrusion. Concurrently, BC concentrations increased from about 0.3–0.5 to 1.0–1.5 $\mu\text{g m}^{-3}$ (Figure 2b). The absorption coefficient of BrC at 370 nm ($b_{\text{abs,BrC}}$) increased from approximately 2–3 to 5–6 Mm^{-1} , while the absorption due to secondary BrC ($b_{\text{abs,SecBrC}}$) increased from approximately 1.5–2.5 to 4–5 Mm^{-1} . Similarly, the mass-normalized absorption ratios increased from approximately 0.05–0.10 to 0.15–0.25 for BrC/ $\text{PM}_{2.5}$ and from 0.05–0.10 to 0.10–0.20 for Sec BrC/ $\text{PM}_{2.5}$ (Figure 2c). The concurrent enhancement of the coarse aerosol mass, the absorbing components of carbonaceous aerosol, and their mass-normalized optical ratios indicate that the observed dust intrusion was accompanied by substantial anthropogenic aerosol loading and secondary aerosol formation, consistent with a polluted dust plume rather than a relatively pure mineral dust event.

The spatial distribution of the dust mass flux from the MERRA-2 reanalysis (Appendix Figure A3a) provides evidence for the enhanced aerosol loading observed during the last part of May 2023. Between 18 and 24 May, dust mass fluxes exceeding 0.4 $\text{kg}\cdot\text{m}^{-2}\cdot\text{s}^{-1}$ extended from North West India towards the western Himalayas, while considerably weaker dust fluxes were present during 11–17 May. The spatial continuity of the enhanced dust flux is consistent with long-range transport from the Thar Desert and adjoining arid regions. The vertical and horizontal transport pathways associated with the observed aerosol variability are illustrated by CALIPSO lidar profiles (Appendix Figure A3b–c). On 16 May, relatively weak aerosol backscatter was observed, with aerosol layers extending from the planetary boundary layer (PBL) into the free troposphere. In contrast, 19 May was characterized by substantially greater aerosol extinction, with most of the aerosol loading confined within the PBL before reaching the measurement site. The enhanced near-surface aerosol layer is consistent with the transport of dust-laden air masses to the western Himalayas. Quantitative evidence from the reanalysis further supports this interpretation. In the MERRA-2 grid cell closest to DRS (32.5°N, 77.0°E), the mean dust column mass density (Appendix Figure A2) increased from $0.029 \pm 0.015 \text{ mg m}^{-2}$ during 11–17 May to $0.063 \pm 0.039 \text{ mg m}^{-2}$ during 18–24 May, corresponding to a 2.1-fold increase. Similarly, the attenuated backscatter profile of CALIPSO 532 nm acquired on 19 May revealed a pronounced aerosol layer centered at $3.2 \pm 0.5 \text{ km}$, close to the elevation of the DRS site (2950 m), indicating that the transported aerosol plume directly influenced surface observations.

The concurrent increase in coarse aerosol mass, carbonaceous aerosol absorption, dust column mass density, and satellite-observed aerosol loading identified 18–24 May 2023 as a distinct polluted dust episode. To objectively distinguish this episode from background conditions, we applied a composite criterion based on three independent indicators: (i) daily $\text{PM}_{\text{coarse}}$ concentrations exceeding the 60th percentile of the May–June distribution for at least four consecutive days, (ii) daily precipitation below 1 mm to minimize the influence of wet scavenging, and (iii) evidence of dust influence from MERRA-2 dust column mass density and dust flux,

together with the vertical aerosol distributions observed by CALIPSO. Based on these criteria, 11–17 May 2023 was identified as the background period, whereas 18–24 May 2023 satisfied the criteria for a sustained dry, dust-influenced event.

A subsequent period, 26 May–3 June, also exhibited elevated $\text{PM}_{\text{coarse}}$, with concentrations reaching approximately $90 \mu\text{g m}^{-3}$. However, this period was not included in the event definition because it followed substantial precipitation on 25–27 May, with a daily accumulation of approximately 10 mm. This precipitation can efficiently remove the coarse aerosol and modify the aerosol size distribution, thus introducing an additional removal process that is difficult to separate from the effects of dust transport. Including this precipitation-affected period would therefore complicate the interpretation of the observed changes in aerosol optical properties and the subsequent optical-closure analysis. We consequently selected 18–24 May as the event period because it represents the longest continuous period that meets the quantitative event criteria and remains relatively free of precipitation-related aerosol removal.

Comment: Second, I am concerned that the interpretation of the optical measurements is not sufficiently rigorous. The authors report increases in scattering, absorption, SSA, SAE, MEE, and changes in AAE during the selected event period. These changes indicate that the aerosol ensemble changed during the event, but they do not by themselves demonstrate that the intrinsic optical properties of dust were fundamentally modified. In particular, absorption increased during the event together with BC concentration, and AAE decreased toward values more characteristic of BC-like absorption. This suggests that enhanced BC or other anthropogenic absorbing components may explain much of the absorption increase. The manuscript does not adequately separate the contributions of mineral dust, BC, and possible BrC to the retrieved absorption and imaginary refractive index. Therefore, the authors should avoid implying that the absorption enhancement reflects chemical transformation of dust itself unless they can explicitly demonstrate this separation.

Response: Thank you for pointing it out. We have completely revised the interpretation throughout the manuscript. We now explicitly state that the observed changes demonstrate modifications in the effective optical properties of the bulk aerosol composition, not the intrinsic changes in the dust mineralogy.

We now write:

Line 329: These changes demonstrate that the bulk aerosol composition during 18–24 May was optically distinct from the background aerosol population.

Line 419: These values represent the bulk aerosol optical properties of the internally mixed aerosol population and therefore reflect the combined influence of mineral dust and coexisting anthropogenic aerosols rather than the intensive refractive index of the mineral dust component alone.

Comment: Relatedly, the current wording throughout the manuscript gives the impression that the authors have characterized the refractive index of “polluted dust,” but the retrieval actually appears to represent an ensemble-average aerosol refractive index. This distinction is critical. If “polluted dust” simply means an aerosol population containing dust mixed with BC, sulfate, and other anthropogenic components, then the authors should state this clearly and interpret the results as ensemble-average optical properties of a mixed aerosol population. However, if the authors intend to claim that chemical aging of mineral dust itself caused the enhanced absorption or modified refractive index, the current data and analysis do not support that conclusion. The abstract, discussion, and conclusion should be substantially rewritten to avoid overclaiming.

Response: We agree and have systematically revised the abstract, discussion and conclusion with phrases like “refractive index of bulk aerosol composition during a polluted dust storm event” throughout the manuscript. In Section 3.1, we emphasize that the term ‘polluted dust’ in this study refers to an aerosol population in which mineral dust particles coexist with and are internally mixed with anthropogenic components (BC, organic carbon, sulfate, nitrate). The retrieved CRI is the effective optical parameter of this bulk composition.

We now write:

Line 216: Polluted dust refers to mineral dust aerosol that has undergone mixing and chemical aging through interactions with anthropogenic aerosol constituents, such as black carbon, organic matter, sulfate, nitrate, and related species, resulting in dust-containing particles or plumes with modified physicochemical and optical properties.

Comment: Third, the discussion structure is difficult to follow. The manuscript often presents results in a report-like sequence rather than developing a coherent scientific argument. For example, the aerosol-type classification based on SAE–AAE appears suddenly in the optical-properties section, but it is not well integrated with the later chemical-composition analysis. The classification scheme is also inherently qualitative and should not be used as strong evidence that the observed optical changes are specifically caused by polluted dust. The chemical data should be used more directly and carefully to support, constrain, or qualify the optical interpretation. As written, the optical, chemical, and refractive-index sections are not sufficiently connected.

Response: We thank the reviewer for this comment. We have removed the SAE–AAE aerosol-type classification matrix from the main text and moved it to the Appendix, as it was qualitative and not well integrated with the chemical analysis. Instead, we now use the chemical data (Section 3.3) as the primary evidence for aerosol composition and mixing state.

Comment: Fourth, the diurnal section does not yet provide a clear scientific contribution. It is not obvious why the diurnal analysis is central to the manuscript, and the current interpretation is not fully convincing. The authors argue that the diurnal behavior reflects boundary-layer growth and valley-scale transport, but the figures suggest that the differences between background and

event periods are persistent across many hours rather than being limited to specific transport windows. If local or valley-scale processes enhance absorption around 08:00–09:00, the authors should explain why a similar feature is not more clearly observed during the background period. Likewise, if boundary-layer evolution controls the transport of polluted aerosol to the site, this mechanism should produce a more clearly defined enhancement during the event period. The discussion should also be revised because some interpretations do not appear consistent with the plotted data; for example, absorption appears to be strongly enhanced in the afternoon as well, not only in the morning. Unless the authors can provide a more quantitative and physically consistent interpretation, this section should be shortened or moved to supporting information.

Response: We thank the Reviewer for this comment. The detailed diurnal heatmaps and probability density functions have been moved to the Appendix, while only a concise description of the main features is retained in the revised manuscript. During the background period, the absorption coefficient does not exhibit a clear morning maximum between 08:00 and 09:00 local time, possibly because BC concentrations are very low and cannot be distinguished on the colorbar. In contrast, a distinct increase in absorption in the morning is observed during the event, when BC concentrations are substantially higher. This may be due to high local emissions during that period. Both σ_{sca} and σ_{abs} show a broader enhancement from the afternoon into the evening during the event, consistent with the combined effects of PBL development and upslope transport. We have revised the text accordingly to clarify that the morning enhancement is superimposed on this broader afternoon-to-evening increase, rather than implying that the absorption enhancement is confined to the morning hours.

Comment: Fifth, the chemical-composition analysis is not well aligned with the main background-versus-event framework. The main discussion up to Section 3.3 is based on a comparison between background and dust-event periods. However, Section 3.4 shifts to a comparison between two selected days within the dust-event period. The authors should explain why 20 May and 22 May were selected, whether they are representative of the event, and why comparable chemical measurements from the background period are not available. Without background chemical composition, it is difficult to use the SEM-EDX analysis to support claims about changes from background to polluted dust conditions. The chemical data may show that dust and carbonaceous particles coexisted during the event, but they do not clearly establish the magnitude or mechanism of the transition from background to event conditions.

Response: We agree with the reviewer. We have therefore substantially revised Section 3.3 (formerly Section 3.4) to clarify its purpose and interpretation. The samples collected on 20 and 22 May were selected to represent early-event and mid-event conditions, respectively, and to examine changes in particle composition as the dust plume evolved. The chemical analysis is now used to provide evidence for within-event evolution, rather than to infer a compositional transition between the background and event periods. Since no single-particle samples are

available for the background period, we have removed any interpretation of the SEM-EDX data in terms of a background-to-event compositional change. The revised section also makes the connection between the chemical composition and the observed optical properties.

We now write:

Line 343: Single-particle measurements were not available during the background period because the low aerosol loading yielded insufficient particles for statistically robust SEM-EDX analysis. Accordingly, the SEM-EDX dataset is used to examine the chemical and morphological evolution of aerosols within the polluted dust event rather than to compare the background and event periods.

Comment: Finally, the ACRI interpretation needs major revision. The authors state that both RRI and IRI show marked enhancement during the dust event, but this conclusion is not fully convincing from the current presentation. The reported increase in RRI appears relatively modest, and the IRI behavior is more difficult to interpret because it is strongly affected by absorbing components such as BC. The authors should provide formal statistical tests or uncertainty-aware comparisons between background and event periods before claiming marked enhancement. More importantly, the retrieved RRI and IRI should be described as properties of the aerosol ensemble rather than dust-specific values. The manuscript should not compare these ensemble-average values directly with literature values for relatively pure mineral dust without carefully acknowledging the role of BC, BrC, and other internally or externally mixed absorbing particles.

Response: We fully accept these comments and have made the revisions accordingly. We have added statistical analysis to Section 3.2 and summarized it in a new Table 1. Given the relatively small number of daily observations and the potential for non-normal distributions of atmospheric aerosol properties, the two-sided Mann-Whitney U test was used as the primary non-parametric test. The Mann-Whitney U test evaluates whether observations from the two periods exhibit a systematic difference in their rank distributions. Welch's two-sample *t*-test was additionally applied as a complementary parametric test to evaluate differences in the group means without assuming equal variances between the two periods.

Comment: Overall, I find that the manuscript addresses a potentially interesting and under-observed region, and the measurement dataset may be valuable. However, the current manuscript substantially over-interprets the observations. The authors need to either fundamentally reframe the manuscript as an ensemble characterization of a mixed dust-pollution aerosol episode or conduct additional analysis that separates the optical contributions of dust, BC, BrC, and other aerosol components. Without such revision, the conclusion that polluted dust differs fundamentally from pure mineral dust due to chemical transformation during transport is not adequately supported.

Response: We have revised the manuscript and removed all claims about “chemical transformation of dust,” “fundamental difference from pure mineral dust,” and “intrinsic refractive index of polluted dust.” The conclusions now state exactly what the data supports.