

Reviewers Response

Reviewer 2

The authors present an analysis of particle optical property measurements made over India, focusing on measurements made during an approximately two-week period during the overall study. They use their measurements to characterize the air masses encountered during the two-week period into a “background” period and an “event” period, where the “event” period is referred to as a “polluted dust” period. They use their measurements to derive changes in the complex refractive index of the particle ensemble from the background to the event period, concluding that the “results demonstrate that polluted dust differs fundamentally from pure mineral dust and exhibits enhanced optical efficiency per unit mass and altered aerosol radiation interactions due to chemical transformation during transport.”

We thank the reviewer for the assessment of our manuscript. A point-by-point response to the comments is provided below.

Comment: I find that the overall measurements seem of good quality and the general framework for the analysis is fine. However, I have serious concerns about the authors interpretation and presentation of their work. As noted above, they conclude that “that polluted dust differs fundamentally from pure mineral dust.” They have in no way demonstrated this. The fundamental problem with their interpretation is that they have not separated absorption by dust from absorption by black carbon. The authors determine complex refractive index values for the entire particle ensemble and make no effort to distinguish between black carbon and dust (and give no consideration to potential contributions from brown carbon). They refer to a particle classification scheme to argue that they are primarily looking at dust. However, they insufficiently recognize that the classification scheme is inherently imprecise, and mixtures of different particles types can drive the results. Based on the current analysis, the authors can at best conclude that the ensemble average imaginary refractive index increased during the event period over the background period. But they have not done an analysis that allows them to identify the reasons for this change and certainly have not demonstrated that the change is a result of chemical transformation of the dust during transport.

Response: We thank the reviewer for his comment. We have revised and toned down the language in the manuscript to avoid overinterpretations of the results. We now emphasize in the revised manuscript that CCSEM-EDX classification provides a qualitative representation of particle composition and mixing state rather than a quantitative apportionment of aerosol types. Accordingly, we have revised the discussion to focus on the observed increase in the bulk aerosol IRI during the event period relative to the background period.

We now write:

Line 370: The CCSEM–EDX analysis primarily provides information on the particle composition and mixing state. Because individual particles can exhibit overlapping elemental signatures and may contain mixtures of components, particle classification should be interpreted as a qualitative representation of aerosol composition and mixing rather than a quantitative apportionment of distinct particle types.

Therefore, the observed increase in the bulk aerosol IRI during the event period relative to the background period cannot be attributed exclusively to a specific aerosol type based on this classification.

Comment: I think this manuscript could be acceptable for publication if the authors either fundamentally change their interpretation and discussion (including rewriting the abstract almost entirely, and certainly the second half) or fundamentally changing their analysis to try and distinguish between contributions from black carbon, brown carbon, and dust, and then pinpointing the dust properties. I suspect this latter approach will be very difficult and will likely end up with statistically insignificant results. Now, if the authors simply mean by “polluted dust” that it is “dust that is internally mixed with BC” then they should say this clearly. But, they should also make clear that the change is driven by the introduction of BC into the system, and not by “chemical changes” to the dust itself.” They should also reframe their work, as this is not an especially novel result. If they mean instead to argue that it is chemical changes to the dust itself (rather than dust-containing particles) that drive the apparent optical changes, then I do not find that the analysis or data support the conclusions.

Response: Thank you for the suggestion. We have rewritten the abstract and the conclusion section.

We now write:

Abstract

Atmospheric aerosols over the western Himalayas are frequently influenced by long-range transport of mineral dust mixed with anthropogenic pollution from the Indo-Gangetic Plain, yet the optical properties of these mixed aerosol populations remain poorly constrained. During a three-month field campaign at the Dhundi Research Site (2950 m a.s.l.), we combined in situ particle mass concentration, optical measurements, aerosol size distributions, black carbon observations, and single-particle chemical analysis to characterize a seven-day polluted dust event. Relative to the background period, the event was associated with enhanced coarse-mode aerosol loading and exhibited approximately threefold higher scattering, a 2–2.5-fold increase in absorption, and an approximately twofold increase in mass extinction efficiency. Single-particle analysis revealed progressive internal mixing of mineral dust with carbonaceous material and secondary inorganic species, indicating substantial atmospheric processing during transport. Using an optical-closure approach based on Mie theory, we retrieved the effective aerosol

complex refractive index of the bulk aerosol population. At 520 nm, the effective real part increased from 1.64 to 1.74, while the effective imaginary part increased from 0.023 to 0.033. These region-specific optical properties provide observational constraints on the representation of polluted dust aerosols in radiative transfer calculations and regional climate models.

Conclusion

This study reports the effective ACRI of a polluted dust aerosol population transported over the western Himalayas and examines the accompanying changes in its optical, microphysical, and chemical characteristics. During the dry period of 18–24 May 2023, enhanced coarse-mode aerosol mass together with elevated BC concentrations indicated the arrival of a dust-laden air mass that had interacted with anthropogenic emissions during transport across the IGP.

Relative to the background period (11–17 May 2023), the event was characterized by approximately threefold higher aerosol scattering, a 2–2.5-fold increase in aerosol absorption, and an approximately twofold increase in mass extinction efficiency. Single-particle chemical and morphological analysis revealed an increasing abundance of internally mixed polluted dust, carbonaceous material, and secondary inorganic species as the event evolved, consistent with the progressive atmospheric processing of the transported aerosol population.

Using an optical-closure approach based on Mie theory, we retrieved the effective aerosol complex refractive index of the bulk aerosol population. At 520 nm, the effective real refractive index increased from 1.64 to 1.74, while the effective imaginary refractive index increased from 0.023 to 0.033. 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. The concurrent increase in BC concentrations and decrease in AAE are consistent with an enhanced contribution of carbonaceous aerosols to the retrieved effective IRI.

The results demonstrate that the optical properties of aerosols in the western Himalayas during polluted dust events cannot be adequately represented using the refractive index characteristic of pure mineral dust or urban aerosols alone. The effective ACRI values reported here provide observational constraints for radiative transfer calculations and aerosol optical parameterizations in regional climate models, while highlighting the importance of representing the aerosol mixing state in simulations of dust transport over the Himalayas. Future work that combines particle-resolved optical modeling with direct measurements of individual aerosol components, particularly brown carbon, will further constrain the contributions of different species to the effective optical properties of polluted dust.

Comment: I will also note that the authors provide no statistical analysis to justify their conclusion that the properties during the event period differ from the background period. They show smoothed (very smoothed) distributions, but a statistical analysis comparing the background

and event period is necessary if the authors wish to contend that there were real changes.

Response: Thank you for the comment. We have added statistical analysis to Section 3.2 and summarized it in a new Table 1.

We now write:

Line 153: Statistical tests were applied to the daily averaged aerosol properties to evaluate whether the differences between the identified background and event periods were statistically robust. 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. Because multiple aerosol properties were tested simultaneously, the resulting p -values were adjusted using the Benjamini–Hochberg false-discovery-rate (FDR) procedure. Differences were considered statistically significant when the FDR-adjusted p -value was less than 0.05.