The manuscript "**Analysis of the Composition and Direct Radiative Effects of Polluted Dust Using Satellite Observations and Reanalysis Data**" by Wang et al. uses 17 years of CALIOP level 2-layer data to locate three climatological polluted dust hotspots and characterizes their collocated composition with MERRA-2. The study then performs idealized core-shell Mie and RRTM experiment for May 2010 over eastern China to examine how the dust-sulfate mixing state modifies the direct radiative effect. The authors report a contrasting the SW-LW response, with internal mixing weakening SW cooling substantially while producing a smaller enhancement of LW warming.

The topic is important and the combined use of satellite observations, reanalysis, and radiative-transfer modeling is potentially valuable. However, the observational analysis constrains aerosol co-occurrence rather than particle-level mixing state, while the radiative calculations rely on idealized assumptions that are not yet sufficiently justified or documented.

The main concerns that should be addressed are as follows:

1. The manuscript builds directly upon the theoretical framework of Zhang et al. (2024), which investigated how dust-sulfate surface coating modifies shortwave and longwave radiative effects relative to external mixing. The present study cites Zhang et al. (2024) as the basis for the core-shell framework (Page 10, Line225), attributes the opposite responses of 550 nm and 10 μm AOD to this previous work (Page 13, Line 298-299), and discusses the atmospheric lensing response in the context of the same mechanism (Page 18, Line 429). However, the manuscript subsequently states that the SW-LW asymmetry in the impact of internal mixing on dust DRE is "to our knowledge, a new result of the present study" (Page 21; Line 507).

The distinction between the previous theoretical finding and the present contribution therefore requires clarification. Please explicitly state what aspects of the SW-LW asymmetry were already demonstrated by Zhang et al. (2024) and what new insights are provided here. For example, if the primary advance is the application of a previously demonstrated physical mechanism to observationally constrained polluted-dust environments using CALIOP/MERRA-2-derived regional aerosol conditions, this should be clearly stated as the main contribution. The regional quantification of mixing-state sensitivity is a valuable contribution; however, the current wording risks overstating the novelty of the underlying physical mechanism.

2. The headline DRE response appears strongly depndent on an unconstrained property of the external mixing reference. The manuscript reports that moving from fully external to fully internal mixing reduces net cooling by

48.4% (land) and 42.4% (ocean) at the TOA, and by 38.1% and 40.1% respectively, at the surface (Page 16, Line 365-369). Importantly, the Discussion itself states that the SW contrast driving these differences is governed primarily by whether the sulfate mass contributes additional externally mixed extinction, rather than by coating-induced changes in the optical properties of the dust particles (Page 20, Line 497-499). The prescribed properties of the external sulfate population therefore become a central control on the magnitude of the principal SW and net-DRE result.

That population is represented using a single dry effective radius of 0.35 μm (Page 10, Line 235), for which I could not find a physical justification or sensitivity analysis. No hygroscopic-growth treatment is described. Because sulfate mass is fixed, the assumed particle size strongly controls particle number, geometric cross section per unit mass, and hence external-mixture extinction. I am therefore not questioning the qualitative external-internal contrast

obtained under the adopted assumptions; my concern is whether the reported magnitude of that contrast is adequately constrained.

I suggest that the authors (i) justify the 0.35 µm assumption and report the corresponding dry sulfate Mass Extinction Efficiency at 550 nm, together with sufficient information on the wavelength- or band-dependent sulfate extinction used in RRTM-SW; (ii) test the principal DRE result across a physically plausible range of sulfate particle sizes or size distributions and show how the reported percentage differences change; (iii) clarify whether the sulfate AOD in Fig. 7b is the native MERRA-2 sulfate-AOD diagnostic or is recomputed from the MERRA-2 sulfate mass using the prescribed particle properties, and compare the two where appropriate; and (iv) state the sulfate density and chemical form used when converting MERRA-2 sulfate mass to external particle number and internal shell volume.

3. The manuscript states that "neglecting this coating enhancement biases the net DRE of polluted dust toward stronger cooling" and presents the associated SW-LW asymmetry as a new finding (Page 21, Lines 505–507). However, the reported external-internal differences indicate that the net-DRE response is overwhelmingly dominated by the SW component rather than the LW enhancement. For instance $DRE_{net,external}$ = SW_{ext} + LW_{ext} = -7.83+2.12 = -5.71 W m-2; similarly $DRE_{net,\ internal}$ = -5.3+2.24 = -3.06 Wm-2. Thus, the total external-internal difference is approximately +2.65 W m-2 (net-DRE difference). Now if we decompose this difference, it shows that the shortwave component contributes: ($SW_{DRE,\ TOA,\ internal}$ - $SW_{DRE,\ TOA,\ external}$)/Difference DRE_{netTOA} = (-5.3 – (-7.83))/2.65 ~95% whereas the longwave contribution accounts for only approximately 4.5% of the net-DRE difference. A similar partition is found at the surface, where the LW contribution represents only about 6% of the external-internal net-DRE difference.

Therefore, although the LW enhancement represents an interesting consequence of dust-sulfate mixing state and may be important for understanding spectral radiative responses, it is not the dominant contributor to the reported net-DRE correction. The conclusions should distinguish between the dominant SW-driven reduction in cooling and the smaller LW warming enhancement. In particular, the statement that neglecting the LW coating enhancement biases polluted-dust DRE toward stronger cooling should be moderated to reflect the relative contribution of the LW term to the total DRE change.

The atmospheric DRE requires a somewhat different interpretation. For this component, the external-internal difference is not dominated by the SW term. From the manuscript it can be seen that a SW atmospheric DRE change of approximately +0.06 W m-2 and an LW change of approximately -0.05 W m-2. However, this also highlights a robustness concern, because the LW atmospheric difference is comparable to the uncertainty implied by the spread among internal-mixing schemes. In Fig. 12, the reported uncertainty represents only the range among Schemes I-III and does not include uncertainties associated with the optical or radiative-transfer assumptions.

The internal-external comparison does benefit from cancellation of some common uncertainties; however, several assumptions do not cancel because sulfate enters the two configurations through fundamentally different optical representations; as independent sulfate particles in the external mixture and as a shell component in the internal mixture. In particular, the adopted sulfate LW optical properties, treatment of sulfate water uptake, and the idealized concentric core-shell representation may directly affect the differential LW response. The manuscript should therefore clarify the sensitivity of this small LW enhancement to the assumptions that control the difference between the two mixing states.

4. The atmospheric profiles used in the radiative calculation are described inconsistently, and this is particularly important for the LW DRE. Section 3 states that the 3-hourly MERRA-2 fields enable direct coupling with solar geometry, “instantaneous atmospheric profiles,” and the radiative-transfer model (page 6, Line 141-142), whereas Section 4.2 states that the aerosol optical properties are combined with “standard atmospheric profiles.” (Page 15, line 341-342)

It is therefore unclear whether the RRTM calculations use time- and location-specific meteorological profiles or a prescribed standard atmosphere. If the intended meaning is that MERRA-2 supplies the aerosol fields while the thermodynamic profiles are taken from a standard atmosphere, this should be stated explicitly and the choice justified, particularly because time- and location-specific meteorological fields are available from MERRA-2.

LW DRE is sensitive to the thermal contrast between the surface, aerosol layer and surrounding atmosphere, as well as to the atmospheric temperature and water-vapour structure. A prescribed standard atmosphere may therefore affect both the magnitude and regional representativeness of the May 2010 eastern-China LW DRE. The manuscript specifies the shortwave surface albedo and longwave surface emissivity, but I could not identify the surface temperature used in RRTM_LW.

Please state explicitly which temperature, water-vapour and ozone profiles were used, whether they vary by grid cell and 3-hourly time step, and which surface skin temperature was prescribed. Two additional aspects of the RRTM setup also need clarification: (i) are the reported calculations clear-sky or all-sky? No cloud treatment is described in Section 4.2; and (ii) are the DRE values instantaneous, diurnally averaged, or monthly averages constructed from the 3-hourly calculations? This is particularly important for the SW results because the treatment and temporal averaging of solar zenith angle directly affect the reported radiative-effect magnitudes.

5. The three hotspots are identified from the DJF climatology, the vertical composition profiles in Fig. 3 are also DJF, and the China/India source-transport analysis in Fig. 5 is presented for DJF. The radiative experiment, however, is conducted for May 2010. Figure 2c shows that the aerosol regime changes substantially between these seasons, with Capital Region dust AOD peaking in MAM and being considerably lower in DJF. It is therefore not clear that the sulfate-to-dust mass ratio and vertical overlap underlying the DJF compositional interpretation are representative of the May 2010 radiative case.

This matters because the amount of sulfate available per unit dust mass and its vertical overlap with dust directly affect the shell volume and the external-internal optical contrast. I therefore suggest showing the May 2010 dust and sulfate mass profiles, their relative burden, and their vertical overlap over the radiative domain. If the May 2010 composition differs materially from the DJF regime, then the use of a sulfate-only shell in the radiative experiment requires a separate justification for that case.

A separate concern is the case selection itself. The manuscript states that May 2010 was chosen because the sulfate-rich background coincides with substantial dust loading, “thereby amplifying the sensitivity of AOD and radiative effects to the assumed internal mixing scheme.” (Page 11 Line 253-254). However, no quantitative selection criterion is given. Please clearly state how May 2010 was selected from the 2007-2023 record and indicate where it lies relative to other months in terms of dust burden, sulfate burden and their relative abundance. I do not object to using an intentionally favourable case for an idealized sensitivity experiment.

The issue is how the resulting numbers are interpreted. If May 2010 was selected because it produces an enhanced response, the reported DRE percentages should be described explicitly as case-specific high-sensitivity results, rather than as representative regional values. A seasonal or multi-year calculation would be needed to support a climatologically representative quantitative conclusion.

6. The manuscript cites Li and Shao (2009) as evidence that dust particles in northern China are frequently coated, noting that approximately 90% of dust particles were coated and that the coatings were predominantly nitrate compounds (L39-41). However, the radiative calculations subsequently represent polluted dust using sulfate core-shell particles based on MERRA-2 composition, where sulfate is identified as the dominant pollutant component over the Capital Region (L165-166). However, MERRA-2/GOCART does not include nitrate as an aerosol species. Therefore, the conclusion that sulfate dominates refers only to the aerosol components represented by the reanalysis and does not establish that sulfate is the dominant coating material of atmospheric dust particles.

This limitation is particularly relevant because the central novelty of the manuscript concerns the longwave radiative response of internally mixed dust. Sulfate and nitrate are both weakly absorbing inorganic aerosols in the shortwave, with comparable real refractive indices and negligible imaginary refractive indices. Therefore, representing soluble scattering coatings using sulfate is a reasonable approximation for many shortwave radiative applications. However, this assumption is less straightforward in the thermal infrared, where aerosol optical properties are strongly controlled by wavelength-dependent vibrational absorption features of the coating material. Consequently, even if sulfate and nitrate coating produced comparable SW scattering, they may produce different LW extinction efficiency and spectral AOD response. The SO_4^{2-} ion asymmetric stretching vibration absorption band occurs near the 9-10 μm region, whereas nitrate exhibits absorption features at different wavelengths in IR. Therefore, the use of sulfate optical constants may not necessarily represent the longwave response of nitrate-containing soluble coatings, particularly for the 10 μm extinction diagnostic adopted in this study. The authors should therefore clarify whether the 10 μm response is representative of the broadband RRTM_LW forcing and whether the reported enhancement depends on sulfate-specific infrared optical properties.

At minimum, the manuscript should report the source and phase assumption of the sulfate refractive index used in the Mie calculations and discuss the extent to which the reported longwave enhancement should be interpreted as a property of sulfate-coated dust rather than polluted dust with arbitrary soluble coatings.

7. The dust microphysical representation used in the Mie calculations requires further clarification. The manuscript should specify the five MERRA-2 dust size bins used and explain how each bin is represented in the optical calculations. In particular, are the transported dust bins treated as single representative radii, or is an internal size distribution applied within each bin? This is important because coarse dust particles have large size parameters at visible wavelengths, and a single-radius representation may introduce sensitivity to the selected representative radius. Relatedly, the coarse-mode representation has two limitations that should be discussed: the treatment of size within each resolved bin and the upper size range represented by the aerosol model. MERRA-2/GOCART represents dust only up to a maximum particle radius of 10 μm, which may omit larger dust particles that contribute substantially to

thermal infrared extinction. The authors should discuss how this size truncation may influence the magnitude of the reported LW response.

The dust density used to convert MERRA-2 dust mass into particle volume should also be explicitly stated because this conversion directly determines the dust core radius and sulfate shell thickness, which are central to the internal-external mixing comparison.

8. Line 361 states that internal mixing enhances longwave warming “even though the total column extinction cross section is reduced.” However, Fig. 8 shows that at 10 μm the internally mixed cases have higher AOD than the external mixture, whereas the reduction in extinction occurs for the shortwave (550 nm) response shown in Fig. 7. Please clarify that the reduced extinction refers to the shortwave response and revise the wording to avoid implying that the longwave warming enhancement occurs despite reduced longwave extinction.

9. The manuscript states that the three internal mixing schemes produce very similar LW extinction profiles, indicating that the detailed size dependence of the coating has little impact on LW extinction (Page 13, Line 293-294). However, Fig. 10 shows a systematic scheme dependence at 10 μm: Scheme III produces the largest increase in bulk extinction efficiency, Scheme I the smallest, and Scheme II an intermediate response (Page 14, Line 326-328). While these differences may be smaller than the corresponding shortwave differences, they indicate that coating allocation does influence LW optical properties. Please clarify whether "little impact" refers only to the relative magnitude of the effect compared with other uncertainties, rather than implying that the coating distribution is unimportant for LW extinction.

10. The manuscript attributes the different behavior of Scheme III to the balance between enhanced LW extinction and changes in single-scattering albedo (Page 18, Line 419-421). However, Fig. 10b shows that the 10 μm single-scattering albedo remains low (~0.35-0.375) in all schemes; the distinction is only relative among the three internal mixing configurations. Therefore, describing Scheme III as having “relatively high LW single-scattering albedo” may overstate its physical significance. Please clarify whether the atmospheric DRE difference arises primarily from relative changes in SSA, changes in LW extinction efficiency, or their combined effect.