

Response to Referee 3

Manuscript egusphere-2026-2772: “Solid-particle stratospheric aerosol injection: a 2-D modeling exploration of the design space for amorphous silica and calcite”

Lederer, Wygoda, Halbertal, Kushnir, and Rose

We thank the referee for the constructive review and for the specific suggestions. Alongside this response we submit a revised manuscript, in tracked-changes and clean form, together with a revised Supplement. The referee’s report is reproduced below in black italics; our reply to each part follows in blue. Line numbers refer to the clean (non-tracked-changes) revised manuscript.

The main changes of this revision relevant to this report: the diagonal-diffusion discussion moved into Sect. 2.1; the DDA validation extended up to $N = 32$, both materials and both monomer sizes (Appendix B); a line-by-line longwave validation added for both materials, as a main-text figure in the model description (Sect. 2.3, Fig. 4), with the band-weight validation in Appendix B; the MODTRAN6 configuration specified in Sect. 2.3; a QBO sensitivity test with time-dependent winds added (Supplement Fig. S6) together with a sulfate transport comparison against published multi-model results, in which the 2-D model and LAGRANTO are both run on multi-year winds without averaging (Supplement Fig. S7); a comparison under the GSFC 2-D transport fields, with all other settings unchanged, added to Fig. 2; latitudinal and seasonal SW/LW forcing profiles added (Fig. S1); a new subsection (Sect. 3.6) comparing the forcing-level results with previous work, supported by the net-SARF row of Fig. S4 and by the forcing response functions of Fig. S8 of the Supplement; and all five suggested references added.

Summary

This manuscript presents a 2D modeling framework with explicit transport, microphysics, and radiation process representations. It is primarily designed to allow computationally feasible simulations of ensembles of solid particle SAI scenarios that are systematically varied over a parameter space encompassing injection scheme, particle size, particle material (refractive index), and fractal geometry of coagulation agglomerates.

Overall Assessment

The paper’s scientific merit is high, and it makes a novel contribution to the literature.

The paper is suitable for publication in principle.

The major improvements required before publication are improvement of citations, clarification of some methodological points, addition of some caveats regarding some findings, and making the Zenodo depository described in the Data and Code Availability fully populated and live by the time of publication.

The paper is in the category of minor revisions before publication.

All the improvements listed are implemented in the revised manuscript as detailed below. The code and data repository are fully populated and live (github.com/stardust-initiative/solid-sai-2d-paper and dependencies within), with citable persistent DOIs (Zenodo record DOI 10.5281/zenodo.20271742, concept DOI 10.5281/zenodo.20271741; Data and Code Availability, lines 690–694).

Major comments:

Transport

The transport in the model is driven by a climatology for the residual circulation obtained from ERA5 reanalysis (averaged over 2008 – 2017) plus mixing represented by a diagonal diffusion tensor. This averaging period averages out the QBO influence on transport, which could have implications for the fidelity of some injection schemes (see below). Regarding the diagonal diffusion tensor approximation, it is worth noting that the Chemical Lagrangian Model of the Stratosphere (CLaMS) has demonstrated that mixing and residual circulation have a comparable contribution to age of air (Ploeger et al. GRL 2015). This finding seems to raise a question regarding how well a diagonal approximation for the diffusion tensor might represent age of air. For this reason, it seems prudent to move the material regarding the diagonal approximation to the main narrative from the appendix and cite the CLaMS finding.

The diagonal-diffusion discussion has been moved to the main narrative (Sect. 2.1, lines 132–136), citing Ploeger et al. (2015). We add that since mixing contributes to age of air comparably to the residual circulation, the age-of-air and LAGRANTO validations test the combined transport and implicitly the diagonal diffusion approximation.

On the QBO: rerunning the 2-D model on the actual sequential 2008–2017 winds, which contain the real QBO phases, changes the global burden by only 3–4 % (Supplement Fig. S6, lines 434–438). For the sulfate reference case the 2-D model and LAGRANTO trajectories are both run over multiple years of time-dependent winds, with no averaging at all, and their residence times agree to within –12 to +4 %, both inside the published multi-model spread (Supplement Fig. S7). Implications for the seasonal-injection results: see the injection-scheme comment below.

Aerosol Microphysics

The model uses a discrete monomer number bin scheme with power of two bins 1, 2, 4, 8, 16, 32, 64, and 128 monomers. The DDA computations to validate the volume equivalent sphere Mie approximation are rigorous, but are limited in the sense that they cover only $N \leq 5$ monomers. Since this only applies to 3 of the model’s 8 bins, an appropriate qualification regarding the validation of the aerosol optics should be added, or a validation for the larger aggregates should be included (likely as Supplementary Information). This is relevant because of the potential for the five larger bins ($N = 8$ to 128) to carry a more substantial fraction of the aerosol mass burden. For example, Figs. 6 and 8 suggest the formation of larger aggregates in high-injection-rate tropical scenarios.

The DDA validation was extended. It now covers configurations up to $N = 32$ (five of the eight sectional bins), both materials, and both monomer sizes (60 configurations; Appendix B, lines 914–947, and the new two-material Fig. B3). It is found that the equivalent-sphere Mie calculations are accurate to $\lesssim 3\%$ at the reference size, within ≈ 13 – 22% for compact aggregates out to $N = 32$, and deviate most (≈ 31 – 44%) for loose fractal $N = 16$ configurations. Weighted by the actual steady-state bin mass populations, the bin-population-weighted deviation is –3 to +1 % for silica and +8 to +13 % for calcite (Supplement Fig. S2). The main takeaways are stated in the main text (Sect. 2.3, lines 230–239).

Radiation Scheme and Validation

The radiation scheme uses an unmodified version of the RRTMG_LW two-stream approximation and introduces a SZA-dependent value for an optically thin-layer backscatter coefficient into the off-the-shelf RRTMG_SW solver. The assumption of optically thin layers holds well for the scenarios considered, but the dependence of this approximation on layer optical thickness should be moved to the main narrative from appendix B.

Sect. 2.3 (lines 197–199) now states the thin-layer approximation in the main text, with its dependence on layer optical thickness derived in Appendix B1.

The validation of the radiative transfer scheme relies on the utilization of MODTRAN6, which is capable of high spectral resolution radiative transfer calculations as well as multi-stream representations of the polar angle radiation field. The details regarding spectral resolution and radiative transfer solver should be added to the manuscript. In particular, the meaning of using 64 moments is unclear if a multi-stream solver is not used. Boucher JAS (1998) and King (1983) should be cited regarding the phase function and number moments.

Sect. 2.3 (lines 205–209) now specifies the MODTRAN6 configuration: “a detailed radiative-transfer calculation by MODTRAN6, run in its DISORT multi-stream configuration [...], with the aerosol scattering phase function supplied as a 64-moment Legendre expansion; the number of expansion terms required for forward-peaked phase functions follows King (1983), and the sensitivity of aerosol shortwave forcing to the phase-function representation is discussed by Boucher (1998).”

An additional point about the Modtran validation: the authors do not validate the LW heating rate profile using Modtran. This is a lost opportunity because, as Figure B1 shows, there is a large amount of spectral structure in the aerosol longwave absorption efficiencies that is averaged out in the RRTMG LW bands. One of the paper’s main diagnostics, the stratospheric heating metric η_{heating} , has a potentially sensitive dependence on band-averaging, as in Sokolik et al. 1998 (JGR) for aerosols and Toon et al. (1989) for clouds. The authors note a Modtran LW heating rate profile comparison will be provided in future work. For now a qualification due to the unquantified uncertainty in η_{heating} would be welcome.

The longwave heating-rate comparison has now been added. A new figure in the model description (Sect. 2.3, Fig. 4 of the revised manuscript) compares the model’s band-averaged aerosol LW heating-rate profile and per-band LW forcing against LBLRTM v12.17, for both materials, for a 10 Tg layer (18–22 km; the LW analogue of the shortwave MODTRAN case). For silica the peak heating rate agrees to 7 % (0.163 vs. 0.176 K day^{−1}), the in-layer RMS residual is 0.024 K day^{−1}, and the column LW forcing agrees to 13 %. The same comparison for calcite is also included: the column LW forcing agrees to 13 % (0.117 vs. 0.103 W m^{−2}) and the in-layer perturbation, a cooling of −0.014 against −0.029 K day^{−1} at the peak, is reproduced to within a factor of two. The qualification the referee requested is included at the definition of η_{heating} (Sect. 2.4, lines 273–278): with the LBLRTM comparison the spectral-averaging uncertainty is now stated as a quantitative bound, citing Sokolik et al. (1998) and Toon et al. (1989).

Results about Injection Scheme Design and Interpretation

As noted above, the transport scheme uses a climatology which averages over a decade of QBO cycles. Since the climatological averaging doesn’t capture the systematic differences in transport between hemispheres, the impact of the displacement of the NH and SH injection windows in the alternating-summer-hemisphere seasonal strategy relative to the QBO cycle is lost. Because of this, the statements in Sections 4.1.4 and 4.2 regarding the relative performance of seasonal versus symmetric injection should be qualified accordingly.

The seasonal-versus-symmetric statements now carry the requested caveat wherever they appear (Sects. 3.4, 4.1.4, and 4.2). The concern was also tested directly: both schedules were rerun with the actual sequential 2008–2017 winds, which contain the real QBO phases (Supplement Fig. S6; lines 434–438). The burden changes by 3–4 % and the seasonal advantage ($\approx 1.23\times$) is preserved: the schedule comparison is not an artifact of the climatological averaging.

Relationship to Prior and Concurrent Literature

Recent observational work by Lyu et al. 2026 (Science) identifies the ubiquity of a small stratospheric aerosol mode with significant organic content. This new work identifies a number of significant interactions between this new mode and hypothetical SAI deployments, and is largely sulfur-centric.

However, for the purposes of the authors' work, the small mode provides an additional coagulation target that would alter the effective aggregate size distribution of injected particles. For this reason Lyu et al. should be cited and its implications for the present work discussed.

Lyu et al. (2026) is cited and discussed in the Introduction: “The background aerosol includes a recently identified mode of small, organic-rich particles in the lower stratosphere (Lyu et al., 2026), which offers a further coagulation target for injected particles; that interaction is not modelled here (Sect. 2).”

Figures

Figure 1: what is the meaning of the gray shading in this figure? Please add to the caption.

The Fig. 1 caption now defines every element; the gray shading is the uncertainty envelope of the observational age-of-air estimates, reproduced from the digitized Fig. 4 of Chabrillat et al. (2018).

Figure 2: the dimer mass fraction in Weisenstein appears to be ~ 0.2 to 0.25 , compared to the author's ~ 0.05 to 0.1 . This is a large relative difference and should be qualified as something less than "close agreement."

The quoted contrast pairs curves from different scenarios: at 1 Tg yr^{-1} the dimer mass fraction is ~ 0.05 in Weisenstein et al. (2015) against ~ 0.10 here (~ 0.07 under GSFC transport), and at 8 Tg yr^{-1} it is ~ 0.20 against ~ 0.26 . As the coagulation scheme is identical, the differences are attributed to the different transport datasets. This is demonstrated by adding results produced with the GSFC 2-D transport fields that drive AER-2D itself (Weisenstein et al., 2004): under that identical transport the resemblance is much better (monomer mass fractions 0.93 vs. 0.93 at 1 Tg yr^{-1} and 0.65 vs. 0.69 at 8 Tg yr^{-1}). Fig. 2 now shows both transports alongside the AER-2D reference, and the text and caption state the comparison accordingly (lines 170–176 and the Fig. 2 caption).

Figure 6: the third panel would more logically be $\tau_{\text{coagulation}}/\tau_{\text{no coagulation}}$. The effective diameter panel is suggestive as a process description but doesn't completely capture the coagulation effect's magnitude and belongs in SI.

Figure 6 (Fig. 7 of the revised manuscript) panel (c) now shows the residence-time ratio $\tau_{\text{coag}}/\tau_{\text{no coag}}$ (lines 330–332 and the Fig. 7 caption), and the aggregate-diameter map has moved to the Supplement (Fig. S3).

Figure 9: While the meaning of this figure is clear, the collapse of spatial and seasonal structure into a globally- and annually-averaged TOA value hinders comparison with other work in the field. This spatial and temporal averaging makes sense for figures 11 and 12, which focus on how different injection times and locations cause well-defined scalar diagnostics to vary. However, for the purposes of understanding differences in spatio-temporal variability with analysis of other injection schemes, these figures should be modified to visualize the space and time variability in SARF for some injection scheme end-members that bookend the variation with the parameter ensemble.

As suggested, the latitudinal structure was added to the Supplement: Fig. S1 decomposes the forcing of Fig. 9 (Fig. 10 of the revised manuscript) into zonal-mean SW and LW profiles, annual and seasonal (DJF/JJA), for silica, calcite, and sulfate (pointer at lines 375–376 of the revised manuscript).

Figure 13: a qualification regarding the symmetric and seasonal injection protocols producing similar profiles should be added. This is because the similar profiles are potentially a product of the model’s averaged QBO transport climatology, and profiles produced under real-world QBO conditions may not be as similar. To be clear, this is to support clear conceptual interpretation of the result, not its suitability for operational SAI.

The statement accompanying Fig. 13 (Fig. 15 of the revised manuscript; lines 488–491) now reads: “the normalized meridional SARF profiles for the symmetric and seasonal cases are nearly indistinguishable [...]” We add that the direct test with the sequential 2008–2017 winds (Supplement Fig. S6) indicates the similarity largely survives under real QBO phasing: the meridional profiles change by less than the $\sim 10\%$ interannual spread.

References

The following citations are referenced in this review but are absent from the manuscript’s reference list. The authors should consider adding them where indicated in the major comments above.

Boucher, O.: On aerosol direct shortwave forcing and the Henyey–Greenstein phase function, *Journal of the Atmospheric Sciences*, 55, 128–134, [https://doi.org/10.1175/1520-0469\(1998\)055<0128:OADSFA>2.0.CO;2](https://doi.org/10.1175/1520-0469(1998)055<0128:OADSFA>2.0.CO;2), 1998.

King, M. D.: Number of terms required in the Fourier expansion of the reflection function for optically thick atmospheres, *Journal of Quantitative Spectroscopy and Radiative Transfer*, 30, 143–161, [https://doi.org/10.1016/0022-4073\(83\)90090-2](https://doi.org/10.1016/0022-4073(83)90090-2), 1983.

Lyu, M., Ahern, A. T., Schill, G. P., Lawler, M. J., Murphy, D. M., Taylor, S. J., Fodel, A., Abou-Ghanem, M., Gurganus, C., Zhu, Y., Tilmes, S., Ray, E., Thornberry, T. D., Gao, R.-S., Hints, E. J., Moore, F., Dutton, G., Nance, D., Hall, B., Rollins, A. W., Waxman, E. M., Zuraski, K., Diskin, G. S., Choi, Y., Pierce, R. B., Weinzierl, B., Kuder, F., Dollner, M., Jensen, E., and Brock, C. A.: An unrecognized mode of small particles in the lower stratosphere, *Science*, 392, eadw8939, <https://doi.org/10.1126/science.adw8939>, 2026.

Ploeger, F., Abalos, M., Birner, T., Konopka, P., Legras, B., Müller, R., and Riese, M.: Quantifying the effects of mixing and residual circulation on trends of stratospheric mean age of air, *Geophysical Research Letters*, 42, 2047–2054, <https://doi.org/10.1002/2014GL062927>, 2015.

Sokolik, I. N., Toon, O. B., and Bergstrom, R. W.: Modeling the radiative characteristics of airborne mineral aerosols at infrared wavelengths, *Journal of Geophysical Research: Atmospheres*, 103, 8813–8826, <https://doi.org/10.1029/98JD00049>, 1998.

All five are added at the indicated locations: Boucher (1998) and King (1983), Sect. 2.3 (lines 205–209); Lyu et al. (2026), the Introduction; Ploeger et al. (2015), Sect. 2.1 (lines 134–136); Sokolik et al. (1998) with Toon et al. (1989), Sect. 2.4 (lines 273–275) and Appendix B.

References

Boucher, O.: On aerosol direct shortwave forcing and the Henyey–Greenstein phase function, *J. Atmos. Sci.*, 55, 128–134, 1998. doi:10.1175/1520-0469(1998)055<0128:OADSFA>2.0.CO;2.

Chabrillat, S., Vigouroux, C., Christophe, Y., Engel, A., Errera, Q., Minganti, D., Monge-Sanz, B. M., Segers, A., and Mahieu, E.: Comparison of mean age of air in five reanalyses using the BASCOE transport model, *Atmos. Chem. Phys.*, 18, 14715–14735, 2018. doi:10.5194/acp-18-14715-2018.

King, M. D.: Number of terms required in the Fourier expansion of the reflection function for optically thick atmospheres, *J. Quant. Spectrosc. Radiat. Transfer*, 30, 143–161, 1983. doi:10.1016/0022-4073(83)90090-2.

Lyu, M., Ahern, A. T., Schill, G. P., et al.: An unrecognized mode of small particles in the lower stratosphere, *Science*, 392, eadw8939, 2026. doi:10.1126/science.adw8939.

- Ploeger, F., Abalos, M., Birner, T., Konopka, P., Legras, B., Müller, R., and Riese, M.: Quantifying the effects of mixing and residual circulation on trends of stratospheric mean age of air, *Geophys. Res. Lett.*, 42, 2047–2054, 2015. doi:10.1002/2014GL062927.
- Sokolik, I. N., Toon, O. B., and Bergstrom, R. W.: Modeling the radiative characteristics of airborne mineral aerosols at infrared wavelengths, *J. Geophys. Res.: Atmos.*, 103, 8813–8826, 1998. doi:10.1029/98JD00049.
- Toon, O. B., McKay, C. P., Ackerman, T. P., and Santhanam, K.: Rapid calculation of radiative heating rates and photodissociation rates in inhomogeneous multiple scattering atmospheres, *J. Geophys. Res.*, 94, 16287–16301, 1989. doi:10.1029/JD094iD13p16287.
- Weisenstein, D. K., Eluszkiewicz, J., Ko, M. K. W., Scott, C. J., Jackman, C. H., Fleming, E. L., Considine, D. B., Kinnison, D. E., Connell, P. S., and Rotman, D. A.: Separating chemistry and transport effects in two-dimensional models, *J. Geophys. Res. Atmos.*, 109, D18310, 2004. doi:10.1029/2004JD004744.
- Weisenstein, D. K., Keith, D. W., and Dykema, J. A.: Solar geoengineering using solid aerosol in the stratosphere, *Atmos. Chem. Phys.*, 15, 11835–11859, 2015. doi:10.5194/acp-15-11835-2015.