Response to Referee 1 (B. Kravitz)

Manuscript egusphere-2026-2772: "Solid-particle stratospheric aerosol injection: a 2-D modeling exploration of the design space"

Lederer, Wygoda, Halbertal, and Rose

We thank Referee 1 for the assessment. In what follows the referee's text is reproduced in black italics and our replies are given in blue; underlined blue text marks a commitment to revise the manuscript. Full references with DOIs for all works cited below are listed at the end of this response.

> *"I will admit that I do not think highly of Stardust. I have moral qualms with the way they operate, and I think their conduct is inconsistent with the norms I value in the scientific research community. The currency of academic research is an honest search for how the world works, and I perceive an effort to use community-based legitimacy in support of Stardust's broader goals. I do not expect the authors to respond to this comment, nor am I particularly interested in a response to it."*

The referee opens with personal views regarding Stardust and states that he neither expects nor seeks a response to them. We agree that such a discussion is not suitable for the ACP open discussion. We note the referee's stated commitment to assess the manuscript objectively notwithstanding these views; that is precisely the standard we ask be applied to the responses below, which address every scientific point in turn.

> *"Having gotten that off my chest, as a reviewer, it is my job to assess the quality of the manuscript objectively. And my honest assessment is that, while I think there are some things in here that are interesting, ultimately this paper is just that – a collection of things. A few of those things are novel, but most are not. A few of those things are relevant to studying SAI, and many are not. A few of them cite the now ample literature, and a shocking number of them do not. A few of them are related to what I can ascertain as a potential main point of the article, but most are tangential. As such, I am recommending rejection of the article."*

On the characterization of the manuscript as "a collection of things," we disagree. The paper is a study of two candidate solid materials, amorphous silica and calcite, at the level of radiative forcing, which is the established first-stage methodology for candidate SAI materials (e.g., Pope et al., 2012; Ferraro et al., 2011; Weisenstein et al., 2015; Dykema et al., 2016; Dai et al., 2018). It resolves how the injection latitude, altitude, and season, in interplay with particle-related parameters, shape the stratospheric aerosol layer, and maps the resulting forcing efficacy jointly with the stratospheric heating across that design space. To our knowledge, this joint mapping has not previously been carried out for solid-particle materials, and has been carried out for sulfate only in part (see the comparison table below). This mapping of the trade-off between forcing efficacy and stratospheric heating is useful in its own right, as it can guide subsequent, more detailed study with higher-complexity climate models. We will add to the introduction of the revised manuscript a section that clearly states the scope of the paper and where it differs from previous works in the field, referring to the studies in the comparison table. The structure of the manuscript supports this: Sect. 2 builds and validates the three components needed to compute the two diagnostics, Sect. 3 maps them across the design space, and Sect. 4 draws the design conclusions. What may read as "a collection of things" is the dimensionality of the design space, which is the subject of the paper, not a lack of a main point. More broadly, forcing-level radiative mapping is the appropriate first step for any new candidate material: it identifies which materials and designs merit the fuller experimental and design effort, and requiring that characterization to be complete before the radiative properties may be studied would invert that sequence.

Study	Material	Model	RF	ΔT_{str}	Detailed injection maps?	Coagulation	Heat→circ. fb.
Dykema et al. (2016)	calcite, diamond, alumina, SiC	1-D RRTM	yes	yes	no	no	no
Weisenstein et al. (2015)	alumina, diamond	AER 2-D	yes	no	no	yes	no
Dai et al. (2018)	sulfate	AER 2-D	yes	no	yes	no (small-rate limit)	no
Vattioni et al. (2024)	calcite, alumina, diamond, SiC, TiO_2	SOCOL-AER, 3-D	yes	yes	no	yes	yes
Duffey et al. (2025)	sulfate	UKESM1, 3-D	yes	no	yes	yes	yes
Bednarz et al. (2023)	sulfate	CESM, 3-D	no	yes	4 cases	yes	yes
This work	**amorphous silica (new), calcite**	**2-D**	**yes**	**yes**	**yes**	**yes**	**no**

Representative solid-particle (and sulfate) SAI studies and the diagnostics they report.

On the citation of the literature, we agree in part: while the manuscript cites a substantial body of sulfate and solid-particle SAI work, the design-oriented sections are under-anchored in the design-specific literature of the past decade, and we accept this criticism. Consistent with the scope of the paper, the literature most relevant to this assessment is that addressing candidate materials and injection design at the forcing level; climate-response studies are cited where they bear on the forcing-level diagnostics. Our replies to Comments 5 and 6 below identify the references being added and where; the remaining design and injection-strategy references are given in our response to Referee 2, who raises the same point. Any of the works cited in this response that are not yet in the manuscript's reference list will likewise be added to the revised manuscript at the points where they support the text. We note that this is a matter for revision, not a ground for rejection. The novelty of the paper's contribution against the prior literature is stated above and placed in context by the comparison table.

Comment 1.

"The authors have come up with a new modeling framework, but I'm not entirely sure why."

The scope of the paper is stated in our general reply above: a study of amorphous silica and calcite at the level of their radiative forcing, across the injection and aerosol-design space. That analysis requires resolving the transport of the injected particles, their coagulation into aggregates, their sedimentation, and their radiative effect, and the framework was built to couple exactly these processes, and no more: it is the minimal model required to survey this design space at the forcing level. We consider the 2-D model, which also includes stratospheric transport driven by ERA5 reanalysis and validated against observations, a useful addition to the tools available to the community; the model is made available as stated in the manuscript, and Referee 2 describes its scripts and code as "extremely well documented and accessible."

"The model has an assumed climate sensitivity and no feedbacks of aerosol heating onto circulation or stratospheric water vapor, so it's not useful for understanding climate in any capacity. (Relatedly, I don't know why Table 1 is in the paper, considering it omits the single largest potential effect on your performance metric.)"

The paper does not claim to compute the climate response. It computes forcing-level diagnostics, a distinct and standard object of study (IPCC AR6 Ch. 7; Forster et al., 2021); as noted in our general reply above, forcing-level evaluation is the established first-stage methodology for candidate SAI materials. The absence of feedbacks is not a deficiency of the calculation but the definition of the diagnostic: the stratosphere-adjusted radiative forcing (SARF) is computed with the troposphere and circulation held fixed and the stratosphere relaxed radiatively (Forster, Freckleton and Shine, 1997). Aerosol heating does act back on the circulation at the response level, on intermediate and long time scales, but its effect on the forcing is expected to be a second-order effect: for example, in fully coupled sulfate simulations it reduces the forcing efficiency by about 10 to 14% even at 4 to 5 K of tropical heating (Niemeier and Schmidt, 2017). For reference, according to Table 1 of the manuscript, mid-latitude injection of amorphous silica would induce a low-stratosphere tropical heating of about 1.1 K $(\mathrm{W\ m^{-2}})^{-1}$. The penetration of water vapour into the stratosphere is likewise part of the climate response, acting on intermediate and long time scales, and therefore lies outside the forcing diagnostic by its definition; the excluded feedbacks are stated explicitly in the manuscript (Lines 73-81). Mapping the forcing for a new material is an informative first step toward the response: to leading order the forcing and its latitudinal structure set the surface-temperature response and influence even the hydrological response (Modak et al., 2016; Bala et al., 2008; Roose et al., 2023). The climate response, moreover, depends only weakly on the particle's identity beyond the forcing it produces: this separation, forcing as the agent-dependent quantity and response as largely agent-independent, is an approximation rather than an exact property of the climate system, but it is a well-established and useful one, and it is what makes the forcing level an appropriate level at which to compare materials and morphologies. We will add to the introduction of the revised manuscript an explanation of the relationship between the radiative forcing and the climate response, and of why forcing-level mapping is a useful first step toward that response.

Table 1 in the paper follows the logic of the forcing-response framework described above: the forcing is the agent-dependent quantity this paper computes, and translating it into a temperature response requires a climate-sensitivity assumption that is external to the forcing level. Eq. 3, which introduces $\eta_{T_{\mathrm{surf}}}$, makes that bridge in a deliberately limited and explicit way, so that a reader can substitute the sensitivity of any preferred model. No comparative conclusion of the paper, across materials, morphologies, or injection strategies, depends on the climate sensitivity value. The manuscript treats $\eta_{T_{\mathrm{surf}}}$ as an approximate metric and uses it only in Table 1. As an initial check, Referee 2 notes that our tropical calcite $\eta_{T_{\mathrm{surf}}}$ is close to the value from a fully-coupled Earth-System-model simulation of calcite injection (Stefanetti et al., 2024). We regard this agreement as preliminary, warranting careful like-for-like verification, and not as a substitute for the coupled three-dimensional climate-response calculation that remains necessary.

> *"There is no mention of chemistry, although given the authors' assertions about chemical effects on the aerosols (more on that later) I'm not sure I would trust the model to do chemistry well anyway."*

This is a declared simplifying assumption in the tradition of preliminary solid-particle studies, which have examined candidate materials as idealized pure substances (e.g., Dykema et al., 2016) or under simplifying assumptions regarding the particle-sulfate interaction (Weisenstein et al., 2015). To what extent the assumption holds for a deployed aerosol depends on the explicit design of the particle, including possible coatings and surface engineering, and on experimental characterization under stratospheric conditions; neither is within the scope of this paper, and we will state explicitly in Sect. 1, as a working assumption subject to verification, that the particles' optical properties are not significantly altered by stratospheric chemistry over their residence time. The radiative significance of the sulfate interaction specifically, the referee's "more on that later," is addressed in our reply to Comment 2.

> *"The active parts of the model appear to be aerosol-radiation interactions, coagulation, and sedimentation. And there's no wet deposition, plus there's no reliable measure of deposition locations anyway*

(it's a 2-D model), so I can't say that sedimentation is an important aspect here either."

Wet deposition is indeed not modeled, and this again follows from the scope of the paper: wet deposition acts in the troposphere, downstream of the stratospheric layer whose forcing this study computes. We agree that in reality it can affect the particle removal pattern and, near the tropopause, the burden profile of the layer's lower edge. How the particles are scavenged will depend on the particle design and its aging in the stratosphere, through hygroscopicity, coatings, and surface state; as with the surface chemistry discussed above, these are outside the scope of this work. The tropopause sink, with mass removed on crossing the tropopause, is therefore a simplifying assumption suitable for a first study at the forcing level, and its cost is bounded. For example, the AER-2D study (Weisenstein et al., 2015) applies a 30-day tropospheric residence time rather than instantaneous removal, and replacing our instant sink with a 30-day residence time would only modestly change the mean stratospheric lifetime, with negligible effect on the stratospheric distribution and the radiative response. The sink treatment is already stated in the model description, which specifies that "the lower-boundary loss is a prescribed-tropopause sink rather than a wet-deposition scheme."

On sedimentation, the reasoning runs in the opposite direction: sedimentation to the tropopause is the dominant control on stratospheric residence time, and coagulation drives it through aggregate growth; the residence-time response across the injection grid (Figs. 6-8) is shaped explicitly by this coagulation-sedimentation balance, which makes sedimentation central to the reported results. On deposition locations we agree, and claim none: the paper reports no deposition maps, and quantifying deposition is outside the scope of the framework (see our reply to Comment 3).

> *"So we are left with a parameter sweep of aerosol scattering, where the most important parameter is the aerosol size distribution. (Actually a chain/agglomerate, where the agglomeration rate can change depending on atmospheric conditions or transport, but since they translate that back into equivalent Mie scattering, it's essentially size.) Plus some window dressing of some other parameters that aren't actually being tested because the model isn't well suited to do so. I am calling transport window dressing, because if you look at what the model is actually capable of testing, the effect of transport is just to modify the coagulation rate, so again, size distribution."*

The stratospheric size distribution is not prescribed: it emerges from coagulation in interplay with transport and the injection latitude, altitude, season, and rate, and mapping that dependence is part of the contribution of the paper. Nor is the problem reducible to size: transport dictates the burden profile of the layer, where and how much aerosol accumulates, and hence the latitudinal and vertical shape of the meridional SARF; this is set by transport rather than by the size distribution, and it underpins the latitudinal-flexibility result in Sects. 3.5 and 4.1.4. We will sharpen, in Sects. 1 and 3.2.1, the framing that the meridional structure of the forcing is set by transport as well as by the resulting size distribution.

> *"But you're not testing this, because on lines 267-268 you're assuming the optimal size distribution. So even then I'm confused about what you're actually testing."*

Since coagulation is a modeled process and the stratospheric size distribution is an evolved output (Sect. 2.4, and above), we assume the comment refers to the injected particles. These are indeed idealized as monodisperse monomers at the per-material optimal diameter, and deliberately so: Lines 267-268 fix the reference injection at the bare-monomer shortwave Mie peak of each material so that materials and injection strategies are compared at a like-for-like optical baseline, and the sensitivity to the injected size is then examined explicitly by varying the monomer diameter d_0 (Figs. 8 and 10). What is being tested is therefore stated: the dependence of the forcing efficacy and heating on material, injected monomer size, fractal dimension, and injection latitude, altitude, and season, with the evolved size distribution as an intermediate output. Injection of a polydisperse initial distribution, as would arise from a real manufacturing and dispersal process, is outside the scope of this study; we will state explicitly in Sects. 2.4 and 4 that the reported

results are for monodisperse monomer injection at the reference diameters, and that polydisperse injection is outside the scope of this study.

> *"I'll note that the fact that the coagulation model is just a means of getting to transport actually works in the authors' favor, because the results in Figure 2 are, contrary to the authors' description, not great until you get to very agglomerated particles (which are heavy and fall out quickly, so probably not nearly as relevant as the particles with fewer monomers)."*

Figure 2 is a verification of the microphysics implementation, not a test of independent physics: the coagulation and sedimentation microphysics follows the standard sectional formulations of Seinfeld and Pandis (2016), the same framework used by the AER-2D model (Weisenstein et al., 2015) against which we benchmark, so close agreement is the expected outcome. The residual differences are likely transport-driven: the two models are driven by different advection-diffusion fields, as AER-2D does not use ERA5 reanalysis. We will state, in the revised Fig. 2 caption, the verification role of the comparison against AER-2D and the transport origin of the residuals.

> *"The radiation model for that parameter sweep is RRTMG, which is pretty good for GCM-scale modeling, but there are better ones if you're testing scattering of a size distribution."*

On the solver: the modified RRTMG scheme used here is benchmarked against a MODTRAN reference with 64 scattering moments (Fig. 3), that is, against the class of higher-fidelity radiative transfer tools the referee refers to. The substantive question for the particles considered here, however, is not the solver but the input optics: whether volume-equivalent Mie spheres represent the scattering of fractal aggregates. This is examined quantitatively with discrete dipole approximation (DDA) simulations of the aggregates in Appendix B6, in which the solar-spectrum-weighted backscattering efficiency of the aggregates, computed across nine aggregate morphologies (up to $N = 5$ monomers), falls close to the Mie volume-equivalent-sphere values, with deviations that are small compared with the uncertainties in stratospheric aerosol optical properties and size distribution. The DDA validation will be extended to $d_0 = 0.3$ μm silica and to calcite as part of our reply to Referee 2's comments, where the same question is examined in detail.

> *"Put simply, this is not an appropriate tool for what the authors are actually doing. I suppose doing this parameter sweep for silica-coated calcite aerosols hasn't been done before, but that leads me to my next point."*

Whether a tool is appropriate is determined by the question it is built to answer; we have answered each of the referee's points above in the context of the question this paper asks, and the comparison table in our general reply above places the framework and its results in the context of prior work. We note the referee's closing acknowledgment that this parameter study has not been done before; the relation between the pure-material optical properties computed here and the coated-particle design is stated in our reply to Comment 2.

Comment 2.

> *"The authors state, without evidence, that the particles are non-reactive in the stratosphere. Given what is known about stratospheric chemistry, it seems highly likely that the silica coating would simply chemically erode away, leaving hygroscopic calcite aerosols, which (according to several reputable studies by Dai et al. and Vattioni et al.) would likely get coated very quickly with background sulfate. The authors' claim that the particles would remain non-reactive is, in my opinion, extraordinary and thus requires extraordinary evidence. And without this evidence, the authors' parameter sweep is invalid because they're exploring the wrong aerosol."*

The design context of this study is a coated particle, engineered to be inert in the stratosphere, with the coating a small perturbation to the optical properties. The working assumption of the paper follows: the radiative calculations use the pure-material optical properties of amorphous silica and of calcite (Sect. 2.3, Appendix B), taken as unaltered over the particle's stratospheric

residence time. The specific process the referee describes, sulfate uptake and the associated compaction of the coated particle, has been examined at the forcing level and found to be second-order: Weisenstein et al. (2015) modelled solid particles that become more compact once coated by background sulfate (their "compact coated" case) and found only a minor difference in the radiative forcing per unit burden relative to bare particles (their Fig. 7), the sulfate coatings being thin (order 10 nm). The forcing-level diagnostics computed here are therefore not expected to be materially altered by this interaction, even where it occurs. Whether this holds for a deployed particle depends on the explicit particle design and on experimental characterization under stratospheric conditions, both outside the scope of this paper; we will state the optical-stability assumption (that the pure-material optical properties are unaltered over the particle's stratospheric residence time) explicitly in Sect. 1, together with the other simplifying assumptions of the framework.

Comment 3.

> *"Let's assume that the particle are pure silica. That leads me to another big problem, which is the authors' claim about the aerosols' 'bio-compatibility'. Breathing silica is deadly – I invite the authors to read about silicosis. The authors don't mention this anywhere in their paper, which is in my opinion a terrible oversight, considering it could make this idea a complete non-starter. More realistically, silica deposition should be quantified, but the authors have a 2-D model that cannot handle deposition adequately. Although there may not be reason that the deposition of these studies would be any different from what is reported in other studies."*

One technical clarification first: silicosis is a disease of crystalline silica (quartz and its polymorphs), whereas Sect. 1 specifies engineered amorphous silica, a distinct non-crystalline material of lower density (2200 against about 2650 kg m^{-3} for quartz) whose regulatory and toxicological classification differs from that of crystalline silica. The premise that this consideration makes the idea "a complete non-starter" therefore rests on a property of a different material.

That said, we accept the underlying point: "bio-compatibility" is an overstated term in a paper that does not analyse deposition, exposure, or health impacts, and the health and ecosystem effects of any injected material are a prerequisite question for deployment that this forcing-level study does not address. We will drop the "bio-compatibility" phrasing from Sect. 1 and retain the neutral amorphous-silica specification. On deposition, we agree with the referee's own observation that there may be no reason to expect the deposition of these particles to differ from that reported in other studies; the deposition profile itself is a transport-and-removal quantity outside the scope of a forcing-level study, as noted in our replies to Comments 1 and 2.

Comment 4.

> *"The emphasis on radiative forcing in the paper (the bulk of Section 2.4) seems odd. The efficiency of injection is not well quantified by this model and it's not clear the authors are using the right aerosol anyway (see point 2 above). Also, line 27 about 'larger achievable radiative forcings' has no basis. One can easily get higher radiative forcings with more sulfate injection, and there is both modeling and observational evidence. I feel like most of this discussion is tangential and misguided."*

The comment raises three points. On the emphasis on radiative forcing, see our general reply above and our reply to Comment 1. The concern about the "right aerosol" is addressed in our reply to Comment 2.

On the sensitivity of the forcing efficacy ϵ (SARF per unit injected mass, Eq. 1) to the injection rate, we have carried out a reduced-rate analysis for the cases described in Table 1 of the manuscript: holding all else fixed, we varied the injection rate (see Fig. 1). While the absolute efficacy values change with injection rate, the qualitative behaviour, and the main design conclusions we draw from it, are preserved. We will add the reduced-rate case, the $\epsilon(R)$ curves of Fig. 1 giving the dependence of the forcing efficacy on injection rate, to the revised manuscript.

On line 27, "larger achievable radiative forcings": the referee is correct that the sentence, as written, is ambiguous, and we will reword the Line-27 statement on "larger achievable radiative forcings" to "larger achievable radiative forcing efficacy". One of the design goals is to reach a given forcing at the lowest possible injection rate. On the referee's point that one can obtain higher radiative forcings with more sulfate injection, we do not dispute this; our metric is the forcing achieved per unit injected mass, not the absolute forcing. We are not sulfate experts, and we note that the reported forcing efficacy of sulfate itself shows a large spread across models, driven in part by the treatment of coagulation and the resulting size distribution (Yu et al., 2026). We will add to the introduction of the revised manuscript a note on this inter-model spread in the sulfate forcing efficacy and on the role of coagulation in setting SAI efficacy.

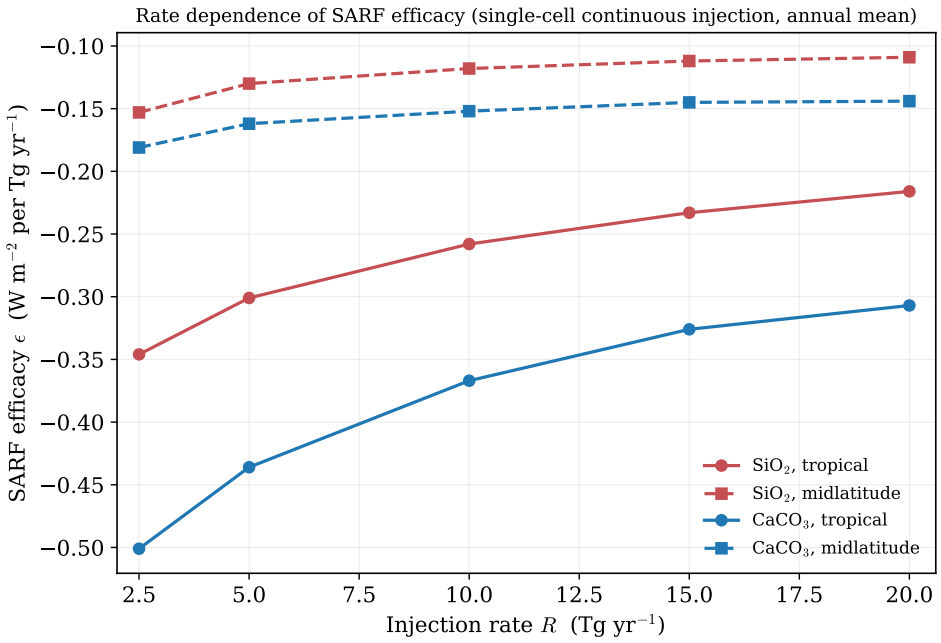

Figure 1: Rate dependence of the SARF efficacy ϵ (more negative = more cooling per unit injected mass) for amorphous silica ($d_0 = 0.5$ μm) and calcite ($d_0 = 0.3$ μm) at the tropical (solid) and mid-latitude (dashed) injection points; single-cell continuous injection, annual mean. The efficacy weakens by roughly 30–40% between 2.5 and 20 Tg yr^{-1} at the tropical point, and less at mid-latitude. To be added to the revised manuscript as part of the reduced-rate case.

Comment 5.

> *"Section 3.2 doesn't seem particularly useful, considering other models that have more advanced capabilities have already done this work and have been doing so for about 10 years. I'm surprised to not see any citations about the relationship between injection location and residence time (e.g., Tilmes et al., Richter et al.)."*

On the missing citations we agree; we will add Tilmes et al. (2017) and Richter et al. (2017) on the injection-location/residence-time relationship and the qualitative dependence of the burden profiles on the injection location to the references, along with the further forcing-level design references given in our response to Referee 2. On the usefulness of the section, Sect. 3.2 serves two purposes. First, it demonstrates that the ERA5-driven transport reproduces this established location/residence-time relationship, the validation step that licenses the design-space maps that follow. Second, as part of the separate-effect approach of this study, it examines the sensitivity of the stratospheric residence time to the solid-aerosol degrees of freedom, the monomer core size and the fractal dimension (Figs. 6-8), degrees of freedom that have little relevance to sulfate and are therefore not addressed by the prior sulfate-SAI work the referee refers to. We will state explicitly

at the start of Sect. 3.2 its two roles: validating that the ERA5-driven transport reproduces the established injection-location/residence-time relationship, and examining the sensitivity of the residence time to the solid-particle core size and fractal dimension.

Comment 6.

> *"I don't know why the authors aren't doing any comparisons with sulfate SAI studies. Surely it would be useful to point out that your aerosol distributions as a function of latitude of injection are similar to those, and if not, you have a mechanism as to why? In fact, the authors have not cited over ten years of literature about design in SAI, even though they specifically mention design parameters."*

We take the point on the literature; we will add the relevant SAI design and injection-strategy references, as committed in the general reply above. As noted there, the literature most relevant to this work is that reporting the forcing-level diagnostics we compute throughout the paper. On the comparison with sulfate: sulfate enters this paper as a reference point rather than as the object of study, and the comparison table in our general reply, which we will add to the revised manuscript, places our results against both solid-particle and sulfate studies (the sulfate cases including Dai et al., 2018; Duffey et al., 2025; Bednarz et al., 2023), providing a direct source for comparison with the sulfate literature.

Comment 7.

> *"I do think the idea of ERA-derived transport in a zonal model is interesting. That strikes me as a sort of data assimilation mode of ISCA, which could be an interesting capability to add to that model. The validation of those aspects (Sections 2.1 and 3.1) are promising."*

We thank the referee for the positive assessment of the ERA5-derived transport approach and of the Sect. 2.1 and 3.1 validation.

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