

Anonymous Referee #2

1. From what is stated in the paper, the authors may wrongly assume that H₂SO₄ in the model refers to liquid sulfate (SO₄=), which is an H₂SO₄ solution. However, in WACCM, H₂SO₄ is a gas that will nucleate and form sulfate aerosols. To derive the sulfate burden in WACCM, one needs consider use so4_a1, so4_a2 and so4_a3 variables. The lifetime of sulfate can be identified in using the excess sulfate burden to decrease by 1/e from its peak, where excess burden is the amount above the pre- injection burden. However, this can be tricky due to seasonal and natural variability.

Thank you for bringing this to our attention. We have replaced our analysis of H₂SO₄ with SO₄ burden.

2. I don't see any support for the conclusions that "studies appear to have under- estimated the potential coverage and therefore reflectance of geoengineered sulfate- aerosols". The coverage of sulfate aerosols has not been investigated in this paper based on the global interactive climate model.

We have expanded on the analysis of aerosol burden in the global interactive climate model. The results in section 3.2.1 of this manuscript are concerned with the ability of the DBS-informed injection schemes to achieve better global coverage in the first three months of transport, which support our statement that other geoengineering model experiments have not optimized injections for efficiently achieving global coverage and maximizing reflectance.

Also, the authors have not investigated changes in coagulation. They have been looking at the effective radius, which does not give a conclusive result. There are several other statements in the conclusions that have not been addressed in the paper, for example, this study has also not demonstrated that fixed aerosols result in "heating hot spots".

We have now included comments that explicitly state that rates of coagulation are correlated with the effective radius, with more coagulation causing larger radii (L440) (e.g. Mills et al., 2017). We have clarified our statement about hot spotting (L265) and instead refer to homogeneous cover and more uniform radiative forcing, which we did address through normalized entropy analysis and global coverage.

3. The dynamical injection method proposed here has demonstrated that within the first 10 days an idealized aerosol disperses faster if injected in regions in dispersion regions (which is not a new or surprising result).

This mathematically rigorous identification of the time-varying locations of maximal dispersion in the stratosphere is indeed new. As well, nobody has addressed the combination of enhancing dispersion and the role that global attractors play (backward DBS ridges) in constraining the spread of aerosols. While these injection locations may sometimes coincide with a user's intuition, there has been no mathematically rigorous effort to identify where these positions are

at a given point in time, as all previous efforts rely on average long-time behavior or proxies for dispersion. The present research provides a way in which the geoengineering community can be more strategic and precise with their injections.

Perhaps the reviewer could provide us with a citation.

Any discussion about the efficiency of SO₂ injections vs sulfate with regard to geoengineering is irrelevant, because the benefit has only been shown in a simple model and for a short time period. What happens if the new method is applied for 10 years and not just for 1 season? Will there be any difference in the aerosol burden?

Thank you for asking this. We have reframed our results to focus on aerosol burden.

As is stated in the manuscript, these experiments present the role that short-time diffusive transport barriers can play in dispersion of aerosols at much longer time-scales. Because this is an initial investigation, we did not want to complicate the results with multiple injections into the same model simulation and have compounding influences, as would likely be done in a real geoengineering application. As well, our method was not applied for one season. We are looking at the evolution from a single day of injections over the course of a year. After the span of one year, turbulence in the stratosphere has effectively mixed sulfates by all approaches, though there are still improvements possible in global energy fluxes as indicated in the new Table 2 and Figure 9.

We have also mentioned this explicitly in the results of the neutral tracer experiment in the following line (L292): *“After 10 weeks, DBS-informed injections were still more effective at global coverage than the fixed-location protocol, even with the airport restrictions, but at yearly timescales, the average improvement was minimal.”*

With the inclusion of aerosol burden values, it is now mentioned in section 3.2 that there is significant difference in performance of the DBS-enhanced and fixed injection protocols. The current simulations are not available for a 10-year analysis.

4. Finally, it is not really clear how the approach of injecting into diffusive regions can improve controllability and improve a controller that relies on the fact that specific injection locations will result in specific AOD and temperature changes. Injecting into regions of increased mixing, how can this be used for the controller development?

There may be some confusion regarding this comment. We do not reference a controller in the manuscript and are uncertain what is meant by controller development. The present research is focused on increasing dispersion at short timescales in order to overcome previously described limitations in geoengineering associated with rates of injection and subsequent coagulation, as well as other downstream effects. We do mention now (L446) that having a shorter time window from injection to achieving global coverage provides less time for interference, such as the influence of unforeseen attracting structures or coagulation.

Line 90: This part needs more explanation, since people should not be required to have to read the referenced papers.

This section has been expanded as follows: Recently, Haller et al. [2018, 2020] derived an additional objective criterion that specifically identifies the strongest barriers and enhancers of diffusive particle transport. That is, one can identify the time-varying locations of material barriers in a fluid flow that maximize or minimize the diffusive contribution in the advection-diffusion equations over a given timeframe. They have obtained a diffusion barrier strength (DBS) field whose ridges highlight the strongest diffusive transport barriers in forward-time fluid flow analysis and strongest diffusive transport enhancers by running a backward-time fluid flow analysis. Neither of these simulations require modeling the evolution of a diffusive scalar field, but still rigorously define the structures that are most influential to diffusive transport.

Additionally, more detail is included explicitly in the methods section 2.2

Line 105: How were geoenvironmental objectives improved in this study? No temperature targets have been met with this method.

We have rephrased the sentence as follows: *“We find significant improvement in the ability of injected aerosols to both quickly surround the earth, and to be able to achieve similar coverage with fewer injection sites.”*

Line 111: What years has been chosen? The SSP8.5 experiment covers 2015-2100? Did you use a standard future fully coupled model configurations?

The analysis was performed on a simulation using 2015-2016 conditions.

Line 118: The importance of transport barriers varies with altitude so differences with altitudes can be expected, what do you mean with “fundamental differences”?

This part of the manuscript has been rewritten in the new 128-136:

A preliminary dispersion analysis was first conducted by approximating aerosol concentration evolution from the behavior of neutral tracers that perfectly follow the wind fields (Figure 3, left column). At the beginning of each month for the full 18.75 years of CESM2 (WACCM6) model simulation, injection locations were identified using a short temporal neighborhood of the wind-field output from CESM2 (WACCM6) Run #1. The advection of parcels of neutral tracers from neighborhoods surrounding those injection points was then computed for the following 50 months in the Run #1 wind fields. This approximation of aerosol transport by perfectly fluid-following particles inherently assumes that there are negligible inertial effects and the aerosol vertical motion is not influenced by radiative heating or cooling of the particle. While these assumptions limit any study of climate impacts, these calculations provide a longitudinal comparison of dispersion from dynamics-informed injections and traditional injection protocols that spans multiple modes of interannual climate variability.

Line 119: Please clarify how aerosols are modeled in this simulation, how do they become non-reactive fluid-parcels?

Thank you for bringing this confusion to our attention. We have clarified this section in the new L128-136. See previous comment.

Line 121: Please explain what is meant by “short temporal neighborhood of the wind-fields output”? Also, the following sentence is unclear. Please describe, what you mean by fluid following particles (are those aerosols in the model, if so which aerosols were looked at?)

We have rephrased here and elsewhere so that it is clearer that “fluid following particles” are actually neutral tracers that do not influence the wind, and perfectly follow air particles in the stratosphere. See new L128-136 and previous two comments.

Line 126: Sentence is unclear: what “numerous natural climate cycles” is referred to here? Do you mean you used the entire 18.75 years for informing the injection locations?

We have rephrased this sentence as follows:

“While these assumptions limit any study of climate impacts, these calculations provide a longitudinal comparison of dispersion from dynamics-informed injections and traditional injection protocols that spans multiple modes of interannual climate variability.”

Line 128: Are these 1-year simulations performed with prescribed wind fields? If so, are those wind fields derived from the 18.75 years of simulation? Having a table that describes the experiments would be helpful. What do you mean by seasonal injections of sulfate precursors? Did you inject SO₂ only once or every season, how much?

Thank you for the suggestion. We have included a flow chart (new Figure 3) to detail the two experiments we ran, and how they are related to the 18.75 years of CESM2 wind field.

Line 210-212: What do you mean by aerosol concentrations, what aerosols are used? What do you choose for the size of a bin, since this is a modal model?

We have clarified that the definition of Eq 2 is with respect to infinitesimal neutral-tracer particles in the stratosphere. This is not a modal model, rather we are quantifying the rate of mixing in the stratosphere wind fields.

Line 218ff: The text is very confusing. The authors discuss H₂SO₄ (the aerosol precursor gas) which then nucleates to form sulfate aerosols. After injections of SO₂ and oxidation to H₂SO₄, H₂SO₄ will decay and aerosol will be formed. It is not clear how the authors learn about the aerosol lifetime and microphysical processes, while investigating H₂SO₄.

Thank you for the suggestions. We have changed our analysis to focus on SO₄ burden.

Line 224: Please explain above how the model simulation has been performed without microphysics (do you mean aerosol microphysics)? Again, another very confusing statement here, the authors now state they injected aerosols. What aerosols have been used?

This point has been clarified by the inclusion of a flow chart (Figure 3) that defines the two separate experiments more clearly.

226: If this is a 2D model lon/lat, what are zonal concentrations? Zonal averages?

Zonal concentration refers to the fraction of the total number of tracers in each latitude band. This has been clarified in the text by adding the following sentence: Zonal concentrations of tracers were calculated as the fraction of the total number of tracers present in a given discrete latitude band. (L246)

Were injections performed at one point or over one longitude band?

Injections were performed at seven separate locations. This has been clarified in the following sentence (L243): *For the infinitesimal neutral-tracer advection experiment (Figure 3, left column), the global coverage of pseudo-aerosols injected at seven dynamically varying DBI locations was much greater than coverage from the seven fixed (FI) locations.*

This is also clarified in the description of our search algorithm in Table 1.

How fast are aerosols been transported longitudinal?

These “pseudo-aerosols” perfectly follow the wind fields along the 540 K isentrope.

How do those locations correspond to the transport barriers subtropics and polar jet stream?

Thank you for bringing this to our attention. We have now included DBS surface plots and SO₄ burden fields at two weeks and six weeks after injection (Fig 11, 12). In these images, multiple transport barriers are present as is their influence on collecting or blocking aerosols. We also future work that could connect well known meteorological phenomena with DBS ridges (L503).

Why is there a difference between winter and summer?

These are time-varying wind fields carrying the neutral tracers that we are tracking. The difference in winter and summer can be easily attributed to the time-varying nature of this fluid flow.

228: What do you mean by: there is north movement of “the volume of particles”?

While there is North-South meandering of the injected tracers, the fixed-injection scheme resulted in little-to-no dispersion by the end of the first week.

**Figure 5: From the figure caption one cannot understand what the lines represent?
Fraction of sulfates with regard to what?**

This has been clarified both in the text and in the caption:

Figure 6: DBS-informed injection yields significantly enhanced coverage over fixed-location injections over short-term, seven-day periods. Zonal concentrations in subplots a-d are calculated as the fraction of the total number of neutral-tracers in a given latitude band at a given time. The time evolution of zonal concentration over one week of transport from the two injection protocols are displayed in subplots a-d with their respective normalized entropy values in subplots e-f.

Figure 6: Assuming that the same amount of “aerosol” has been injected for the fixed and the variable injection sides, how can the Relative Entropy be different at the start?

Concentrations were calculated by the number of particles in a lat-long grid, and the area of each cell varies with latitude. As the “area” of the parcel of particles is smaller than the grid cells, having the same number of particles in a grid closer to the equator results in a different computation of concentration. The minor difference present between the “7 sites” and “Fixed Loc.” in this graph is the result of this grid.