

Response to reviewer

Review:

The authors have appropriately addressed most of my comments, but there are still questions and concerns about the paper that require substantial revision of the text.

First point: It seems like there is still confusion about injecting aerosols vs aerosol precursors and there needs more clarification and discussion throughout the text. The text often refers to aerosol injections, which is however not applied in most model studies and potentially for applications. Injecting aerosol precursors (not aerosols) will result in a different evolution and growth of aerosols, than aerosols. This is because the time it takes for nucleation of sulfur (SO₂), as for example discussed in Mills et al. (2017). The lifetime of SO₂ is usually about 30 days, but it depends on the availability of OH. After a large volcanic eruption, or large SO₂ injections, OH depletion can lead to a prolonged lifetime of SO₂ (47 days after MT Pinatubo). The application of the dispersion model using a tracer or aerosol is therefore different than injecting SO₂. To outline the complexity, it has been shown that SO₂ injections at a point location results in smaller aerosols than injections at a longitudinal band. This is because, the zonal wind is already dispersing the gas quickly and therefore reduce the amount at the injection location. More nucleation is induced instead of condensation on existing particulates. Furthermore, after enhanced aerosol burden has been established in the stratosphere, sulfur injections will condensate on existing particles independent on the dispersion efforts.

1) We thank the reviewer for pointing out that we need to be careful in our references to aerosols versus aerosol precursors; we have clarified this distinction throughout that none of our simulations are injecting aerosols but either consider passive tracer (referred to as neutral tracers or pseudo-aerosols) or an aerosol precursor. While we acknowledge the reviewer's point about differences in chemistry and aerosol microphysics, it is possible that there is some confusion about our methods. The purpose of the DBS-informed injection is to identify transport barriers and transport-enhancing features. While we identified these features using pseudo-tracers (effectively infinitesimal radiatively inert aerosols), identification of the barriers does not depend on the form of the tracers. The transport barriers will affect aerosols, gases, or anything else that is advected by the wind fields, and our second set of simulations indeed inject the precursor gas SO₂ and not aerosols.

This has now been stated more clearly in lines 144-149:

"We note that although Run #1 involves calculation of neutral tracers (resembling infinitesimal radiatively inert aerosols), Run #2 involves injection of the gaseous aerosol precursor SO₂. SO₂ requires time to convert to sulfate aerosols (e.g., Mills et al., 2017), and the injection strategy of SO₂ (for example along a longitudinal band instead of into a single grid box) has been demonstrated to affect aerosol size and hence radiative effects of the injection (e.g., English et al., 2012). Nevertheless, the purpose of these DBS-informed simulations is to describe the effects of recognizing transport barriers or atmospheric features that enhance transport. The applicability of this method is not dependent on whether a gas or particle is injected."

Second point: From what is shown in the paper, I am not convinced that this method leads to significant improvements. As shown in the paper, after a one-year simulation of the fully coupled model, it seems that there are no significant differences in coverage between the fixed injection method and the dynamically derived injection method in terms of efficiency (Fig 9). Fig 9 is showing a strong reduction in coverage in the first 30 days, which may have to do with the lifetime of SO₂, and how long it takes to build up a larger sulfate coverage. It would be helpful to also show the absolute values of sulfate here, which are likely to be very small initially. Furthermore, Table 2 indicates, that after 30 and 365 days, in 2 out of 4 cases, the radiative forcing is more strongly reduced in the fixed injection case and the third case shows almost no difference between the DI method. I agree that this method may be useful to consider for the onset of sulfur injections and should be explored in more detail. However, I don't think the authors can support that there are "long-term gains in sulfate burden and radiation" as stated in the conclusions. I would therefore at this point advise against recommending this method as "a benchmark improvement in injection protocol".

2) We appreciate the reviewer's point about overstating some of our conclusions. We have removed the two statements identified by the reviewer and have gone through the manuscript to ensure that our conclusions are supported by our results. Other statements have also been modified to most accurately reflect our findings.

As it stands, we have a rigorous, mathematically-supported theory for enhancing gas or aerosol dispersion along atmospheric structures. This theory is supported by our initial neutral-tracer experiments in simplified climate model wind fields. The influence of these injection choices is also present in our Run #2 simulations, though the intended effect is harder to predict. We do not feel that this negates our efforts, rather indicates that dynamic methods of injection location optimization should be further investigated.

Related to point #1, we reiterate that transport barriers apply to anything advected by the wind fields, be it aerosols or gaseous precursors.

Finally, as stated in my earlier review, two co-authors of this study work on a feedback controller to improve climate impacts. It is important to address the question whether this method is suitable for applying a feedback-controller. Injection locations and amount have been chosen to improve the climate outcomes in particular of surface temperature. How could this method be integrated?

3) We have now added a short description of how a feedback algorithm might be used with DBS-informed injection in L485-491:

"To manage uncertainties in atmospheric flow and climate response, several recent geoenvironmental climate modelling studies have employed a feedback algorithm that regularly adjusts the SO₂ injection rate (Jarvis and Leedal, 2012; MacMartin et al., 2014; Kravitz et al., 2017). Future studies involving DBS-informed injection could pursue something similar. Outlining this process, every week of simulation, new injection locations would be determined

based on wind fields from the previous week, using the DBS algorithm described previously. The model would then be run forward for a week with the SO₂ injected at those locations. This process, which essentially constitutes a form of Model Predictive Control (Garcia et al., 1989), could be carried out for the length of the simulation.”