Replies to comments by reviewer 1 - Filip Vanhellemont

Comment: Review: Detectability of solid particle injections into the stratosphere with satellite solar occultation instruments

By Anna Lange et al.

In this paper, the authors investigate the possibility to detect the presence of artificially injected solid particles (5 Tg per year) into the stratosphere by using the standard satellite solar occultation measurement technique. The injection of these particles is at present debated in the (still controversial) framework of geoengineering. Usually, SO2-induced sulphate aerosols are considered; here, solid alumina and calcite particles are discussed, due to their increased scattering efficiency and reduced infrared absorption. Using the SOCOL-AERv2 circulation/chemistry/microphysics model and the SCIATRAN Radiative Transfer model, synthetic retrievals of 565 nm aerosol extinction were obtained, taking into account a number of error sources. The results clearly show that for injections of 5 Tg per year, the presence of these particles is clearly observed in solar occultation retrievals, and can be distinguished from the mean background stratospheric aerosol loading, even when natural variability is considered.

General comments

This paper forms a substantial contribution to the scientific research related to the mitigation of global warming by geoengineering, since it clearly shows that the effects of stratospheric injections (here, in particular, alumina and calcite) can be measured by standard occultation techniques. Since geoengineering is quite a hot topic, I consider ACP to be an appropriate journal. Overall, the assumptions, methods and conclusions are valid, although one aspect of the retrieval mathematics should be clarified (see below). I've suggested some additional references, but in general, ample credit is given to other authors in the field. The structure of the paper is well organized. Use of English is also good, but I did include a list of suggestions to improve the clarity of the text. Furthermore, mathematical notation should be improved. If the comments below are properly addressed, I see no objection to the publication of the paper. I therefore advise to publish with minor revisions.

Reply: Dear Filip Vanhellemont, we thank you for your constructive and helpful comments. We hope we answered them in an appropriate way.

Comment: Specific comments

Is there a specific reason why the paper deals with solar occultation instruments only? It is fair to assume that similar conclusions can be drawn for ground-based observations, or limb scatter and nadir instruments. If there is a reason, a short explanation would be good.

Reply: We thank the reviewer for pointing this out. Solar occultation is based on the attenuation of the direct solar beam and therefore do not require additional assumptions on, e.g., the aerosol phase function, multiple-scattering contributions, or surface albedo, which are needed for limb-scatter and nadir geometries, respectively. This keeps the retrieval and error analysis comparatively well-constrained. Furthermore, this work connects directly to the retrieval framework established in our previous study (Lange et al., 2025), which was also developed for solar occultation geometry. The adoption of the same instrument concept ensures methodological consistency and comparability between the two studies.

We added the following to the introduction:

"This study focuses on satellite solar occultation instruments because they measure the attenuation of the direct solar beam and the associated retrievals do not require additional assumptions on, e.g., the aerosol phase function, multiple-scattering contribution, or surface albedo, which are needed for the interpretation of limb-scatter measurements. This keeps the retrieval error analysis comparatively well constrained. Furthermore, this work connects directly to the retrieval framework established in the previous study (Lange et al., 2025) and the adaption of the same instrument concept ensures methodological consistency and comparability between the two studies."

Lange, A., Niemeier, U., Rozanov, A., and von Savigny, C.: Investigating the ability of satellite occultation instruments to monitor possible geoengineering experiments, Atmos. Chem. Phys., 25, 11673–11688, https://doi.org/10.5194/acp-25-11673-2025, 2025.

Comment: Line 14: please also add the more recent paper by Kremser et al, Stratospheric aerosols - Observations, processes, and impact on climate, Reviews of Geophysics 54, 278-335.

Reply: Added.

Comment: Line 26: please cite the ALTIUS paper: Fussen et al., The ALTIUS atmospheric limb sounder, JQSRT vol 238, 2019.

Reply: Added.

Comment: Line 38: Please explain from where the number of 5 Tg per year originates. If it comes from Vattioni et al (2025), it would be a good idea to briefly repeat the reason for this number here. The entire conclusion of the paper (detectability or not) depends on it.

Reply: The injection rate of 5 Tg per year was chosen because, as shown in Vattioni et al. (2025), it results in an all-sky radiative forcing of about -1 $\mathrm{W\,m^{-2}}$, an appropriate order of magnitude for an SAI scenario aiming to offset some of the effects of anthropogenic climate change. We have added the following to the manuscript:
"The injection rate of 5 $\mathrm{Tg\,yr^{-1}}$ was chosen because it results in an all-sky radiative forcing of about -1 $\mathrm{W\,m^{-2}}$ (Vattioni et al., 2025), a reasonable magnitude for an SAI scenario aiming to offset some effects of anthropogenic climate change."

Comment: Line 78: Briefly explain here why the tropical latitudes from 30°N to 30°S are chosen as injection site. It may seem obvious to the authors, but it would improve the text for readers that are non-specialists.

Reply: Thank you for pointing this out. We added the following to the manuscript:
"These tropical and subtropical latitudes were chosen because material injected into this region is efficiently distributed globally by the Brewer–Dobson circulation, leading to a more even spread across both hemispheres."

Comment: Line 85: What is the spin-up period? Is it the period necessary to reach steady-state?

Reply: Thank you for the question. We rephrased it at follows:
"The extinction coefficients represent yearly means averaged over a 15-year period in the quasi steady-state phase, following a 5-year spin-up period required for the simulation to transition from the initial reference conditions to quasi steady-state, during which the stratospheric aerosol burden builds up under continuous injection."

Comment: Section 2.2.2 (Retrieval): it is not clear from the text if random simulated noise was added to the transmittance, before converting to the logarithm (measurement vector y) for every individual retrieval. Please specify.

Reply: We rephrased it at follows:
"...with y as measurement vector, containing the logarithms of the transmission values at 565 nm for all tangent heights (no random noise realisations were added to y), ...

Comment: Again, Section 2.2.2: the measurement vector **y** contains the logarithm of the transmittance. Therefore, the Beer-Lambert law says that **y** is linearly related to the aerosol extinction coefficients: **y=K x**, with **K** the matrix containing the path segment lengths in the atmosphere. In Eqn. 1, the terms $\mathbf{F(}x_a\mathbf{)}$ and **-K***x_a disappear. The merit function (Eqn. 2) is then simply $||Kx - y||^2 + ||x - xa||^2$, which is the standard expression for optimal estimation. Can you clarify why the equations are unnecessarily complicated? (perhaps I'm missing something).

Reply: Thank you for the comment. We agree that, for the retrieval of extinction coefficients from the logarithm of the transmission, the Beer–Lambert law results in a linear expression. We presented the equations in the general (linearised, iterative) optimal estimation form because this is the formulation implemented in the SCIATRAN retrieval algorithm (Rozanov et al., 2011), where the inverse problem is formulated in logarithmic form and solved with a Gauss-Newton iterative approach.

Comment: Eqn. 5: is it valid to add the variances due to T and P (constant air density) to the variances due to T only and again P only? Isn't this overestimating the error?

Reply: The "temperature and pressure (constant air density)" perturbation varies T and p jointly such that the air density is held fixed, thereby isolating only the effect of the temperature dependence of the ozone absorption cross section. In contrast, the individual "temperature" and "pressure" perturbations are applied, so that they isolate only the effect of the resulting change in air density.
We clarified this in Table 3 as follows: "Temperature and pressure (constant air density, effect on

ozone absorption cross section)". Furthermore, we added: "Note that temperature and pressure are treated as statistically independent error sources, whereas in reality temperature and pressure are physically coupled. This may lead to a slight overestimation of the total error."

Comment: Figs. 2 and 5: I think the figure would be clearer to interpret if you would plot the altitude of the tropopause on every plot as a horizontal line. It really is the stratosphere that is of interest in this paper.

Reply: Thank you, added. The tropopause heights shown now in Figs. 2 and 5 are taken from the SOCOL-AERv2 reference simulation. For the SAOD calculations (Figs. 3, 6) and the natural variability analysis (Sect. 3.3), SAGE II tropopause heights (2000–2004) were used, to ensure consistency with the SAGE II-based natural variability estimate.

Comment: Figs. 3 and 8: how was the stratospheric optical depth obtained? Integration from the tropopause up to the maximum available altitude in the data? Please specify.

Reply: Yes, correct. The stratospheric optical depth is obtained by integrating the extinction coefficient from the tropopause up to the maximum available altitude in the respective dataset. The latitude-dependent tropopause heights are taken from SAGE II data (2000 – 2004), consistent with the natural variability analysis. For the SOCOL-AERv2 data, the upper integration limit is 27 km, as described in the Methodology section. For the SAGE II data, the same upper limit of 27 km is applied, to ensure consistency with the SOCOL-AERv2-based results. We have clarified this in the manuscript as follows:
"...The stratospheric optical depth is obtained by integrating the extinction coefficient from the tropopause up to 27 km, consistent with the upper limit of the SOCOL-AERv2 data (see Sect. 2.1)."

Comment: Line 202: It is stated that at 75°S, detectable SAI signals are present above about 20 km. In Fig. 5, I observe detection (background outside the retrieval error) from 10 km to 24 km, which does not correspond with the text. Please correct.

Reply: Corrected, thank you. This referred to the comparison with the alumina injection, where detection above 20 km was not possible. But we have now adjusted this (change is printed in bold black):
"Compared with the extinction coefficient profiles based on the injection of alumina (compare Fig. 2), the extinction coefficients for calcite are slightly higher, but still of the same order of magnitude. Consequently, the corresponding profiles deviate more distinctly from the background profiles. In particular, at 75° S (panel (b)), this leads to detectable signals above ≈ 20 km **(here up to 24 km)**, where the background profile lies outside the error range of the retrieved profile."

Comment: Technical comments, corrections and suggestions
It would be good to specify in the abstract that conclusions were obtained for stratospheric aerosol extinction at the specific wavelength of 565 nm. For example, Line 7: "Retrievals of 565 nm aerosol

extinction demonstrate that, under the assumptions made, ..."

Reply: Thank you for this suggestion. We have revised the corresponding sentence in the abstract accordingly: "Vertically resolved retrievals of the stratospheric aerosol extinction coefficient at 565 nm demonstrate that...".

Comment: Line 26: change 'probably' − > 'potentially', and '... above a certain size ...' − > '... above a certain particle size ..."

Reply: Thank you for your comment. We have amended the first part. In the second part, 'size' refers to the injection rate. To make this clearer, we have reworded it as follows: "...can potentially be used to detect possible future SAI deployments above a certain injection rate."

Comment: Line 36: change '... start-ups may see an option to earn money with SAI (e. g. "Make sunsets" company)' − > '... start-ups may find SAI to be profitable (e. g. the "Make sunsets" company)'.

Reply: Changed, thank you.

Comment: The entire paragraph from line 76 to 81 contains some redundancy (aluminia or calcite, radius of 240 nm are mentioned several times). Could you remove this redundancy?

Reply: Rephrased as follows: "Following Vattioni et al. (2025), simulations were conducted with a continuous injection of 5 Tg yr^{-1} of mono-disperse alumina or calcite particles with a radius of 240 nm, close to the optimal radius for backscattering solar radiation (Dykema et al., 2016), injected at 50 hPa ($\approx$ 20 km) and distributed homogeneously across all model grid boxes ($\approx$ 325 km $\times$ 325 km $\times$ 1.5 km) between 30°N and 30°S. These tropical and subtropical latitudes were chosen because material injected into this region is efficiently distributed globally by the Brewer–Dobson circulation, leading to a more even spread across both hemispheres. The particles were assumed to be fractal aggregates composed of 240 nm spherical monomers, further details on the model simulations are given in Vattioni et al. (2025).".

Comment: It is important to make a distinction in mathematical notation between scalars and vectors/matrices/multidimensional objects in general. One possibility is to write multidimensional objects in boldface, and scalars in plain roman. Eqn. 1 then is: $\mathbf{y} = \mathbf{F}(x_a) + \mathbf{K}(\mathbf{x}\text{-}x_a)$. Please do this everywhere in the text.

Reply: In accordance with the ACP guidelines, we have decided to make the following changes:
- Matrices boldface
- Vectors boldface italic

Comment: Line 109: change text to: "... the logarithms of the transmission values at 565 nm for all tangent heights, . . . ”

Reply: Changed, thank you.

Comment: Line 122: change text to “. . . and T(TH) as the transmission value . . . ”, in order for the notation to be conform with Eqn. 4.

Reply: Changed.

Comment: Line 128: change text to “. . . the error estimation on the extinction was . . . ”

Reply: Changed to ”..the error estimation of the aerosol extinction coefficient was performed as follows:...”.

Comment: Eqn. 6: This is a scalar equation; it can be written in a better way as $r_i = (x_i - x_{ref,i})/x_{ref,i}$, with i the index indicating the tangent heigh index.

Reply: We have rewritten Eq. (6) as suggested. With i denoting the altitude index of the retrieved extinction coefficient profile, and x_i, $x_{\mathrm{ref},i}$ the retrieved extinction coefficients at altitude i for the modified and reference settings.

Comment: Table 4, second row, first column: change to “A priori”.

Reply: Changed.

Comment: Table 4, 4^{th} row, second column: change to “Extinction coefficient profile with SAI (alumina or calcite), retrieved with SCIATRAN.”

Reply: Changed, thank you.

Comment: Fig. 3, Fig.6, caption: change to “. . . optical depth (565 nm) as function of latitude . . . .”. Similar for Fig. 8.

Reply: Changed in the captions and in the corresponding text.

Comment: Line 201: change to “. . . the corresponding profiles deviate more distinctly . . . ”

Reply: Changed.

Comment: Line 205: change “pannel” to “panel” (twice).

Reply: Corrected, thank you.

Comment: Fig. 5, caption: change “apriori” to “a priori”

Reply: Changed in all relevant captions.

Comment: Line 247: change to “For both injection scenarios (compare Figs. 3, 6), this is the case.”

Reply: Changed, thank you.

Replies to comments by reviewer 2

Comment: This paper shows the results of a study on the detection of geoengineering with solid particles at a rate of 5 Tg per year using a simulation of satellite solar occultation measurements and aerosol extinction retrievals on SOCOL-AERv2 model output. The detection criteria considers both measurement uncertainty and natural variability during a "near-background" period. The study is interesting and relevant, and fits well within the scope of AMT. However, there are a few shortcomings in the design of the study and the assumptions in the simulations that need to be addressed:

Reply: We thank the reviewer for his/her constructive and helpful comments. We tried to answer every comment in an appropriate way.

Comment: Line 2.2.2: What is the rational for a 30% a priori variance? Are the covariance terms (off-diagonal) assumed to be zero? How sensitive are the results to this assumption? Similarly, the assumed instrument SNR seems to be a critical element of this study, and the sensitivity to this assumption needs to be addressed. For example, how similar is this value to the current standard satellite occultation measurements, for example from SAGE II, or SAGE III/ISS, or even SCIAMACHY? The authors also need to compare their retrieval uncertainty to the uncertainties reported by a typical (real) occultation instrument.

Reply: Thank you for the comments. The a priori variance of 30% was chosen as a 'loose' constraint. This is because, on the one hand, it must allow the retrieved state sufficient freedom to deviate from the a priori (background) profile, which is necessary as the SAI signal considered here differs from the background by up to several orders of magnitude. On the other hand, the a priori covariance ensures regularisation of the inverse problem, thereby preventing unphysical, oscillating solutions. The value of 30% thus reflects a standard methodological choice balancing these two requirements.
The off-diagonal elements of the a priori covariance matrix are not assumed to be zero. The off-diagonal elements of the a priori covariance matrix are defined as:

$$S_a^{i,j} = \sigma^2 \exp\left(-\frac{|z_i - z_j|}{r_c}\right) \tag{1}$$

where σ is the a priori variance, z_i, z_j are the altitudes corresponding to element (i,j) of the covariance matrix, and r_c is the correlation radius, which was set to 3.3 km in this study.
The maximum SNR of 1000 (at TH = 60 km) used in this study is based on Meyer et al. (2005), as cited in the manuscript, who report SNRs for SCIAMACHY solar occultation measurements of above 1000 throughout the visible and near-infrared range for a sufficiently high TH. The value used here therefore represents a conservative choice relative to the SNR reported for a real solar occultation instrument rather than an optimistic assumption. Please also note, that we used a TH-dependent SNR, as described in the manuscript.
To put the retrieval uncertainties into context with real satellite solar occultation instruments, we compare them to reported uncertainties for SAGE III/ISS. Wrana et al. (2021) report an extinction measurement uncertainty at 20 km altitude of 5.66% at 520 nm, averaged over the period from June 2017 to December 2019 (SAGE III/ISS Level 2 solar aerosol product). The retrieval uncertainties

obtained in our retrievals at the injection altitude ($\approx 20\,\mathrm{km}$) range from 3 – 7% for alumina and 2 – 4% for calcite, which is of the same order of magnitude as the SAGE III/ISS uncertainties.

We added the following to the manuscript:
- The a priori variance of 30 % allows the retrieval state sufficient freedom to deviate from the a priori and ensures regularisation of the inverse problem, thereby preventing unphysical, oscillating solutions.

- The off-diagonal elements of the a priori covariance matrix are defined as follows:

$$S_a^{i,j} = \sigma^2 \exp\left(-\frac{|z_i - z_j|}{r_c}\right) \tag{2}$$

where σ is the a priori variance, z_i, z_j are the altitudes corresponding to element (i,j) of the covariance matrix, and r_c is the correlation radius, which is set to 3.3 km in this study.

- The estimated retrieval uncertainties at the injection altitude of $\approx 20\,\mathrm{km}$ (3 – 7 % for alumina and 2 – 4% for calcite) are comparable to the uncertainties reported for the SAGE III/ISS Level 2 solar aerosol product. Wrana et al. (2021) reported an extinction measurement uncertainty of 5.66 % at 20 km altitude for 520 nm, averaged over June 2017 – December 2019.

Comment: Section 3.1: At several points, the authors mention the "relevant altitude range". This needs to be more thoroughly supported with at least some discussion of a tropopause definition with justification. It is mentioned later in the discussion around Fig 3, that SAGE II tropopause altitudes were used for the optical depth calculation but that this limits the analysis to 55 S to 55 N. This seems to be an unneeded limitation – tropopause altitudes are readily available from a large number of sources, including almost certainly from the SOCOL-AERv2 model output.

Reply: Thank you for the comments. In line with reviewer 1 we added the tropopause heights to the Figs. 2 and 5. The tropopause heights shown now in Figs. 2 and 5 are taken from the SOCOL-AERv2 reference simulation. For the SAOD calculations (Figs. 3, 6) and the natural variability analysis (Sect. 3.3), SAGE II tropopause heights (2000–2004) were used, to ensure consistency with the SAGE II-based natural variability estimate. Furthermore, we added: "The tropopause heights from SAGE II were chosen to ensure consistency with the following discussion on the natural variability, which is likewise based on the SAGE II data.".

Comment: Section 3.3: What is the justification for using only "near-background" levels of stratospheric aerosol to characterize natural variability? Stratospheric aerosol levels have been highly variable over the last two decades with perturbations from both minor to moderately sized volcanic eruptions and increasingly frequent forest fire smoke.

Reply: We agree that stratospheric aerosol levels have been highly variable over the last two decades. The SAGE II period 2000–2004 was chosen because it represents one of the closest approximations to near-background conditions. We do not claim that this period is entirely free of variability. The

standard deviation shown in Fig. 8 already illustrates the year-to-year variability even within this comparatively quiet period.
We consider the analysis of the detectability under such near-background conditions a necessary baseline case, as it represents the most conservative scenario for detectability. Characterising detectability against a volcanically perturbed background would raise a different question, since a major volcanic eruption would likely lead to an adaptation or temporary suspension of the SAI deployment strategy (e.g. Quaglia et al., 2024). In this case, "background" and "signal" could no longer be clearly separated, and disentangling the volcanic perturbation, an adapted SAI signal, and residual natural variability would require a dedicated analysis beyond the present study.
Furthermore, we note that the injection rate of 5 Tg per year considered here corresponds to an all-sky radiative forcing on the order of -1 W m^{-2} (now added to the manuscript), i.e. a large-scale climate intervention that would require coordinated, large-scale deployment rather than a small-scale or clandestine application. Such a coordinated deployment can plausibly be expected to be actively monitored and adapted in response to volcanic eruptions, further supporting the argument above that a volcanically perturbed background would need to be considered jointly with a correspondingly adapted SAI strategy, rather than as an independent source of natural variability against an unchanged SAI signal.

We added the following to the manuscript: "Near-background conditions were chosen, as this represents the most conservative case for detectability. A background disturbed by volcanic activity, for example, would very likely require an SAI strategy adapted to the volcanic eruption, which would mean that the background and the signal would no longer be clearly distinguishable and a separate analysis would be required.".

Quaglia, I., Visioni, D., Bednarz, E. M., MacMartin, D. G., & Kravitz, B. (2024). The potential of Stratospheric Aerosol Injection to reduce the climatic risks of explosive volcanic eruptions. Geophysical Research Letters, 51, e2023GL107702. https://doi.org/10.1029/2023GL107702.

Comment: Section 4: The conclusions from this study are somewhat limited due to the small scope of the study: only one injection scenario (5 Tg per year) and one set of instrument/retrieval parameters. It would be much for useful if the authors could identify a limit of detection in terms of both injection rate and instrument SNR (for example).

Reply: The injection rate of 5 Tg per year was not chosen arbitrarily. This injection rate results in an all-sky radiative forcing on the order of -1 W m^{-2} (now added to the manuscript), representing a physically motivated, policy-relevant reference scenario for solid-particle SAI. Extending the analysis to a range of injection rates would require additional SOCOL-AERv2 simulations with the corresponding solid-particle microphysics at each rate, which are currently not available and are beyond the scope of the present study.
We agree that detectability is specific to the assumed instrument performance, and our conclusions apply to an idealised instrument with characteristics comparable to current solar occultation instruments, as described in Sect. 2.2.1 and 2.2.2. We note that the manuscript already addresses this

point: "When interpreting the results presented here, the underlying assumptions must be taken into account. It is not the aim of this study to make general statements about the detection capabilities of all satellite solar occultation instruments, but rather to assess the detectability of SAI scenarios based on solid particles within a consistent simulation framework based on typical satellite solar occultation measurements." Furthermore: "It is worth noting that at significantly higher injection rates than the 5 Tg yr^{-1} considered here, the zero-transmission problem, i.e. very low transmissions from the perspective of the satellite solar occultation instrument, could become relevant for solar occultation retrievals, potentially limiting the detectable range of SAI scenarios (Lange et al., 2026). Within the injection rates examined in this study, however, this effect is not expected to play a role.". We consider a systematic sensitivity analysis across injection rates and instrument SNRs as a valuable direction for future work.

Comment: Technical corrections:
Line 71: Are these really "Mie" calculations? Better to say something like "Scattering cross-section calculations…"

Reply: Changed to "The scattering and absorption cross sections for the solid particles were calculated using a semi-empirical code..."

Comment: Lines 82=85: At this point the discussion shifts from discussing Vattioni et al., 2025, to the current work and it's somewhat confusing. For example, what does "Based on" and "are analysed" mean in this context?

Reply: Changed to "The SOCOL-AERv2 model output provided by Vattioni et al. (2025) is used to derive the vertical profiles of the extinction coefficients, covering altitudes from 10 to 27 km, for alumina and calcite in the 440 – 690 nm wavelength band. In the present study, these profiles are calculated as zonal means at latitudes ranging from 85° S to 85° N in 10° steps.".

Comment: Line 97: Does the vertical field of view, quoted in Table 1, account for the forward scattering of the aerosol into the "instrument" line of sight? If not, how is this relevant?

Reply: Thank you for the question. The vertical field of view does not account for forward scattering of radiation into the "instrument's" line of sight. As described in the manuscript (Sect. 2.2.1), the transmission calculations are performed in the transmission modelling mode (solar occultation mode), in which only the directly transmitted solar radiation through the spherical Earth's atmosphere is simulated, including the effects of atmospheric refraction. Under the injection rates considered here (5 Tg per year), the resulting increase in aerosol extinction does not lead to highly reduced transmission values, such that the directly transmitted solar radiation can be expected to dominate over the forward-scattered contribution.

Comment: Figs 2,3,5,6: The "total error" is potentially confusing. The plot actually shows the range of uncertainty about the retrieved profile, not the total error. Please consider changing to shading on the plot and a different terminology.

Reply: Thank you, we've reworded it to 'retrieval uncertainty range'.