Benduhn, F., Schallock, J., Lawrence, M. G. (2016): Early growth dynamical implications for the steerability of stratospheric solar radiation management via sulfur aerosol particles. - *Geophysical Research Letters*, *43*, 18, p. 9956-9963.

DOI: http://doi.org/10.1002/2016GL070701

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Geophysical Research Letters

RESEARCH LETTER

10.1002/2016GL070701

Early growth dynamical implications for the steerability of stratospheric solar radiation management via sulfur aerosol particles

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Key Points:

- There are five sufficient growth dynamical preconditions for the steerability of stratospheric SRM
- Logistics condition the steerability via the initial concentration of the injected material
- The fulfillment of all preconditions will require careful optimisation of the main injection parameters

Supporting Information:

- Supporting Information S1

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Citation:
Benduhn, F., J. Schallock, and M. G. Lawrence (2016), Early growth dynamical implications for the steerability of stratospheric solar radiation management via sulfur aerosol particles, *Geophys. Res. Lett.*, *43*, 9956–9963, doi:10.1002/2016GL070701.

Received 2 AUG 2016
Accepted 9 SEP 2016
Accepted article online 10 SEP 2016
Published online 23 SEP 2016

Abstract Aerosol growth dynamics may have implications for the steerability of stratospheric solar radiation management via sulfur particles. This paper derives a set of critical initial growth conditions that are analyzed as a function of two key parameters: the initial concentration of the injected sulfuric acid and its dilution rate with the surrounding air. Based upon this analysis, early aerosol growth dynamical regimes may be defined and classified in terms of their likelihood to serve as candidates for the controlled generation of a radiatively effective aerosol. Our results indicate that the regime that fulfills all critical conditions would require that airplane turbines be used to provide sufficient turbulence. The regime's parameter space is narrow and related to steep gradients, thus pointing to potential fine tuning requirements. More research, development, and testing would be required to refine our findings and determine their global-scale implications.

1. Introduction

Various so-called solar radiation management (SRM) techniques have been proposed for intervening in the climate system by increasing the planetary albedo, thereby partially counteracting global warming. The production of an artificial aerosol layer in the stratosphere via the local release of sulfur or other types of aerosols, known as stratospheric solar radiation management (SSRM), is currently perceived as one of the most promising candidate for climate interventions [*RoySoc*, 2009; *National Academy of Sciences*, 2015; *European Transdisciplinary Assessment of Climate Engineering*, 2015]. Model studies suggest that injecting a sufficient amount of sulfate could reduce the global average temperature by an amount that is comparable but opposite to the expected global warming due to the ongoing emissions of anthropogenic greenhouse gases [*Rasch et al.*, 2008; *Heckendorn et al.*, 2009; *Tilmes et al.*, 2009; *Niemeier et al.*, 2011]. While it seems likely that aerosol microphysics will eventually limit the potential of SSRM to counteract global warming [*Niemeier and Timmreck*, 2015], it should be considered that particle growth dynamics may also have an influence on determining the characteristics of the formed aerosol early on in the growth phase [*Turco and Yu*, 1997]. Depending on the actual implementation of a technique that is based on sulfur particles, particle formation and a substantial fraction of their growth would take place in the vicinity of the injecting device [*Pierce et al.*, 2010]. However, in global models aerosol processes are computed on the grid box scale, which is numerous orders of magnitude larger than the length scale on which particle formation actually occurs. The simulation of particle processes with an inappropriate spatial resolution may result in a misrepresentation of the relationship between the injected sulfur mass and the amount of global average cooling that is achieved. *Pierce et al.* [2010] and *English et al.* [2012] have tried to increase the spatial resolution of global models via the use of embedded plume models, which would provide a more realistic aerosol input function for application in such models.

This study reexamines key technical elements early on in the aerosol growth process that affect the steerability of SSRM and further defines and explores the circumstances under which its steerability may be improved during this initial period.

2. Steerability

The steerability of SSRM refers to the capacity to spatially and temporally control its effects both in nature and scale. The potential to achieve a desired degree of steerability would be reduced if it were not possible to technically control particle size and number concentration. If these parameters were determined exclusively by the ambient properties of the atmosphere, a degree of steerability of SSRM would only be realizable via the mass of sulfur released and its temporal and geographical localization. Active control over the size and particle number produced would open up an entirely different dimension of possibilities.

The following elements may be seen as important for the steerability of SSRM:

1. The chemical species that is released. This pertains to the choice of whether a precursor gas such as SO_2 or H_2S is injected or whether H_2SO_4 is released directly. *Rasch et al.* [2008a] and *Pierce et al.* [2010] have pointed to sulfuric acid as the most likely chemical species through which one might use technical means during the injection to steer the aerosol size and number concentration that will eventually be obtained on the global scale. The release of a precursor gas would restrict the prospect to achieve control over the size and number of the geoengineered particles, as the chemical conversion requires considerable time, such that particle formation would solely be conditioned by the ambient properties of the atmosphere wherever the respective precursor species is oxidized to sulfuric acid.
2. Aerosol microphysics. Self-coagulation among liquid sulfate particles appears to be inevitable. Collisions among liquid particles would only become inefficient if a chemical composition were chosen to increase surface tension well above that of sulfuric acid. Self-coagulation need not be detrimental to the steerability of SSRM in as much as it is predictable and used as part of a process that builds particles of the desired size. *Turco and Yu* [1997] have shown that particle growth in a jet plume is essentially steered by the initial concentration of the condensible and its dilution rate with the surrounding air. Above a certain threshold of initial concentration the nucleation rate plays no role in the aerosol size distribution that is eventually obtained. This is due to the self-limiting property of aerosol dynamics [see, e.g., *Friedlander and Wang*, 1966], which implies that as time progresses, the particle number concentration of any coagulating aerosol is increasingly independent of its initial values. As nucleation is a process that is particularly difficult to simulate with confidence, self-limited growth dynamics facilitate the accurate prediction of particle size and number.
3. Interaction with background particles. The coagulation of newly produced sulfate particles with older background particles also appears to be inevitable and to the degree it occurs decreases the ratio of cooling by SSRM versus injected sulfur mass, since it results in a smaller increase in particle surface area than the growth of new particles. However, it could be reduced to a minimum if it was technically feasible to ensure that the amount of interaction during the early growth period would be negligible. The early growth period may thus be defined as the time interval during which the coagulation rate with background particles is lower than self-coagulation among newly produced sulfate particles [*Pierce et al.*, 2010]. Past this point the interaction with local background particles is not distinguishable from self-coagulation, and further, interaction among aerosol particles may be taken into account in terms of the aerosol that is chosen to be produced as a result of the early growth period. As we neglect background particles in our early growth simulations, we take the direct approach of defining the distinguishability via the similarity of number concentration.
4. Logistics. SSRM would involve the release of a considerable mass of sulfur into the stratosphere if it is to have a significant global impact. Current estimates to counteract global warming typically range between a few and 45 Mt S yr^{-1}, depending on the degree of global cooling to be achieved and the multiple factors that would determine the actual efficiency of SSRM (see, e.g., *Niemeier and Timmreck* [2015], *Schmidt et al.* [2012], *English et al.* [2012], and other references mentioned above). Planes (or more precisely fighter jets) appear to be one of the most realistic options to inject such a huge mass at high altitude [*Rasch et al.*, 2008a]. Based on payload and other plane specific considerations, the number of planes required may be quite low [*McClellan et al.*, 2012]. However, as will be shown herein, steerability considerations might also have an impact on the logistic requirements of SSRM.

The four conditioning elements mentioned in this section—injected species, aerosol microphysics, interaction with background particles and logistic requirements—will now be discussed in terms of their combined implications for SSRM steerability. This analysis should allow us to derive a number of preconditions for the achievement of steerability, especially in terms of the optimum particle size, the predictability of aerosol growth, and the interdependence of aerosol properties and logistics.

3. Constraining an Optimal Injection Scenario

SSRM is based on the backscattering of incoming shortwave radiation by injected or secondarily produced aerosol particles. Whereas a single sulfate particle's scattering efficiency increases with size, for SSRM this simple relationship is complicated by several factors: (1) Sulfate particles beyond a radius of the order of 1 μm have a substantial absorptivity of longwave radiation [*Lacis et al.*, 1992], thus inducing a heating effect that tends to be larger than their cooling potential, which is an undesirable feature for a technique aimed at counterbalancing global warming; (2) On the other hand, the shortwave scattering efficiency of a population of submicron particles increases monotonically with their total surface area (see, e.g., *Kokhanovsky and Zege* [1997]), such that a larger number of smaller particles tend to scatter more efficiently than a smaller number of larger particles; (3) Larger particles have a shorter lifetime, as sedimentation acts as a limiting factor to their lifetime. Sedimentation is the primary removal process of sulfate particles from the stratosphere for particles with a radius exceeding $r \approx 0.8$ μm and should therefore limit local heating effects via longwave absorption [*Benduhn and Lawrence*, 2013]. Still, combined shortwave and longwave absorption and indirect effects via the increase of scattered radiation are not negligible and may result in a significant perturbation of stratospheric circulation and chemistry [*Pitari et al.*, 2014]. Exclusively focusing on the shortwave scattering potential, and adjusting for global average sedimentation and transport losses as a function of particle size as described in *Benduhn and Lawrence* [2013], the optimal particle size can be estimated to be $r \approx 0.25$ μm (see Figure S1 in the supporting information).

In order to limit undesirable particle number reduction via the scavenging of immature particles through larger background particles, the early growth period should be as short as possible. Long-term particle growth and number reduction via self-coagulation may then be estimated as follows. Global aerosol model studies suggest that a typical concentration of radiatively relevant particles for stratospheric SRM would be of the order of 10–100 cm^{-3} depending on location, altitude, injected mass, and injection scenario [*Heckendorn et al.*, 2009; *English et al.*, 2012]. If only transport and sedimentation are considered, the lifetime of these particles decreases with size and is of the order of 1–3 years for particles of $r = 0.1$–1 μm. Coagulation and condensation render the particles larger, such that their lifetime should not exceed 3 years. A size of 1 μm should not be readily reached, as particles should take too long to grow to this size [*Benduhn and Lawrence*, 2013]. For a self-limited aerosol, the number concentration, *c*, of the particles is only a function of the coagulation kernel, *k*, and time, *t*. As a first approximation, it may then be shown that $c = k^{-1} t^{-1}$ [*Benduhn and Lawrence*, 2013]. For an assumed lifetime of $t = 2$ years and an effective coagulation kernel $k = 10^{-15}$–$10^{-14}\ \text{m}^3\ \text{s}^{-1}$ one then finds a concentration at the end of the particles' lifetime of $c = 1.5$–$15\ \text{cm}^{-3}$, which is somewhat lower than the concentration computed by global models. Due to mixing in the stratosphere, number concentrations will not simply decline from the injection to the removal site but will have a more or less pronounced average character. If a "start" background value of 100 cm^{-3} is assumed and the "end" value would be typically 10 cm^{-3}, then particles would on average undergo about three self-coagulation events during their entire lifetime as background particles. As a first approximation, an average background particle would thus have undergone one to two coagulation events, which with the monodisperse approximation would correspond to a doubling to quadrupling in mass or a ~25–60% increase in size, if variations in aerosol number and mass due to sedimentation and condensation are neglected as a first approximation. *Pierce et al.* [2010] find a significantly larger size increase of the geoengineered particles after they are handed over from the plume to the global model. At a number-median handover size of $r = 0.095$ μm they find median particle size to be $r \approx 0.13$ μm at the equator right in the middle of the injection area, and $r \approx 0.18$ μm at 40°N, thus pointing to significant condensation of ambient H_2SO_4. A more accurate estimation of the average increase of particle size may thus be a roughly 100%.

As we found $r \approx 0.25$ μm to be an optimum particle size, as given by considerations of scattering properties and sedimentation and transport losses, an optimum size after early particle growth would be roughly $r \approx 0.125$ μm. This size is compatible with *Pierce et al.* [2010], who found the global aerosol burden to decrease as particles beyond $r = 0.095$–0.180 μm are injected. However, it needs to be considered here that our optimum size estimation is (1) more inclusive, as not only the burden but also the relationship between burden, size, and scattering efficiency is considered; (2) complementary, as we obtain this estimate with a rather simplified approach; and (3) essentially prospective (in opposition to prognostic), as we want to show that long-term background particle interaction could in principle be restricted, provided that appropriate technical optimization prerequisites are developed and implemented. The findings of *Pierce et al.* [2010] may thus be relativized as the technique is implemented, tested, and adapted for optimization and steerability considerations. Finally, it will turn out that particle size is not a limiting factor to SSRM steerability considerations, as particles of the entire potential optimum size range of $r \approx 0.10$–0.25 μm may be produced.

The use of H_2SO_4 instead of a precursor gas would increase steerability, as the geoengineered aerosol particles could grow to size within a relatively short time interval. The growth of sulfate particles from H_2SO_4 depends on the initial concentration, while it does not if a precursor gas is used. As will now be shown, the number of planes required may be a function of the initial concentration if H_2SO_4 is used. If particle dynamical and/or related steerability considerations were to recommend a low initial concentration, and logistic considerations were to disfavor the use of H_2SO_4, the (complementary) use of a precursor gas might be a benefit or even a precondition for SSRM with sulfate particles. Within this study we will therefore consider both the exclusive use of H_2SO_4 and the complementary injection of a precursor gas as possible options to achieve steerability of SSRM.

The number of planes required may be expressed as follows:

$$N = \max\left\{\frac{r_r}{r_c},\ 1\right\}\cdot\frac{M}{P_{\max}\cdot n},$$

where r_r is the distance required for the full release of the maximum payload of the plane at the release rate that is chosen in order to produce particles with target characteristics, r_c is the cruising range of the plane, M is the total mass to be released within a given time period, $P_{\max}$ is the maximum payload of the plane, and n is the number of flights that can be achieved by one plane within the given time period.

In case H_2SO_4 is used, the release distance depends on the initial concentration and may be calculated as follows:

$$r_r = \frac{P\{H_2SO_4\}}{S\cdot C},$$

where $P\{H_2SO_4\}$ is the mass of sulfuric acid to be released during one flight, S is the total injection surface area of the device(s) on board, and C is the initial mass concentration of the H_2SO_4 that is injected.

There is no aerosol growth dynamical prerequisit to release a precursor gas at a certain initial concentration, such that $r_r < r_c$. If H_2SO_4 or a mixture of H_2SO_4 and a precursor gas is released, growth dynamical requirements may limit the initial concentration of H_2SO_4 to the point that r_r has to exceeed r_c.

Figure 1 shows the number of planes required as a function of the initial concentration of H_2SO_4, assuming 1 Mt S yr^{-1} released exclusively as H_2SO_4, a release device(s) total injection surface of 1 m^2, and a payload and cruising range of 1 t and 1000 km, respectively. Furthermore, it is assumed that each plane is able to perform on average six flights daily and that pure H_2SO_4 may be released, as water vapor would be sufficiently abundant in the fuel combustion plume and the stratosphere for it not to act as a limiting factor to nucleation and particle growth. The lower limit of the initial concentration range considered was chosen for the nucleation rate to be high enough that self-limiting behavior may be assumed [*Turco and Yu*, 1997]. The higher limit was chosen such that $r_r < r_c$. It follows from Figure 1 that under the given circumstances, the number of planes would depend on the initial concentration whenever $C < 10^{16}$ molecules H_2SO_4 cm^{-3}, as the payload could not be released within the cruising range of the planes. The number of planes increases quickly below this concentration, to the extent that if for whatever reason such a concentration was chosen, a sufficient quantity of sulfur could not be realistically released without the injection of an additional amount of precursor gas for $C < 10^{15}$ cm^{-3}.

Sufficient, however not unique, preconditions to achieve steerability of SSRM on the basis of early growth dynamical considerations may thus be summarized as follows:

1. The evolution of a self-limited aerosol may be more readily predicted, as the number of variables that particle growth dynamics depend upon is reduced. Self-limiting behavior may be assumed for $C > 10^9$ molecules H_2SO_4 cm^{-3}.
2. At the end of the early growth period, particle size should be of the order of $r = 0.10–0.15$ μm, considering that the average long-term size increase for $c < 100$ particles cm^{-3} should be of the order of 100%, and that the optimum particle size should be $r \approx 0.25$ μm.
3. The use of H_2SO_4 is a prerequisite for the control of particle formation during the early growth period. However, for $C < 10^{15}$ molecules H_2SO_4 cm^{-3} the injection of a precursor gas alongside H_2SO_4 would become a necessity.
4. For full control over particle formation, particles should grow close to target size before they reach the background number concentration.

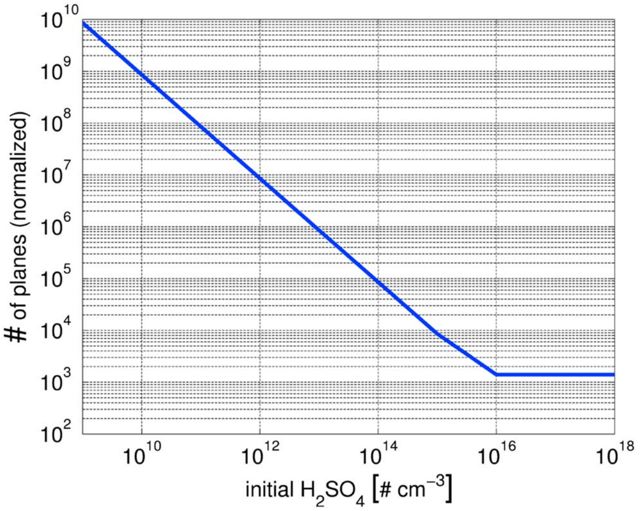

Figure 1. Number of planes required per Mt S a^{-1} to be injected globally as a function of the initial concentration of H_2SO_4. See text for applied normalizations and assumptions.

5. In order to limit the interaction with background particles, the early growth period should be as short as possible.

4. Numerical Simulations

Following *Turco and Yu* [1997] we will now explore whether the above preconditions can be fulfilled simultaneously as a function of the two main early growth dynamical parameters of self-limited aerosol growth, plume dilution rate, and initial H_2SO_4 concentration. The initial concentration range that will be considered is similar to the previous one for the estimation of the required number of planes. The plume dilution rate is associated with the local diffusivity of the air, assumed to be equal throughout the plume, and held constant during the length of the experiment, which is set to 12 h. The choice of the time interval of 12 h is sufficient for large-scale mixing effects to be nonnegligible, and aerosol growth may not be considered to be in the early growth period after this time interval, considering the typically predominant zonal component of stratospheric wind of the order of magnitude of 10 to 100 m s^{-1}. All effects of significant early growth control should thus take place in a shorter time interval. The diffusivity range will span from 10^{-2} m^2 s^{-1}, a value that is typical for ambient conditions in the stratosphere [*Brasseur and Solomon*, 2005], to 10^3 m^2 s^{-1}, which is representative for conditions in the vicinity of a plane turbine [*Schumann et al.*, 1998]. A constant value greater than the background diffusivity cannot realistically be maintained during the entire early growth period, such that none of these values is representative on its own. However, this study does not attempt to simulate aerosol plume growth realistically but to derive principles for SSRM steerability as a function of the two main early growth parameters.

The simulations were carried out with the aerosol microphysics model described in *Benduhn* [2008], linked to the parameterization of binary homogeneous nucleation of *Vehkamäki et al.* [2002], and extended for the representation of plume dilution. By definition, self-limited aerosol growth does not depend on the actual nucleation rate. Any assumption on the mechanism can be made, if the formalism that is used yields a sufficient amount of nucleated particles for self-limited growth to occur. Figure 2 shows the number-median particle radius at the point in time that the assumed background particle number concentration $c = 100$ cm^{-3} is reached, as well as the corresponding time interval. It is apparent that the maximum time interval of 12 h is exceeded for a large fraction of the considered parameter space. In this later circumstance the particle radius that is reached at this point in time is indicated.

The results of Figure 2 may be compared against the findings of *Pierce et al.* [2010] for the case of slow plume dilution described therein, at an initial cross section of 6 m^2 and injection rates of 3 and 30 kg S km^{-1}. This case is equivalent to a diffusivity coefficient of 10^{-2} m^2 s^{-1} at initial concentrations $C \approx 9 \cdot 10^{15}$ and $9 \cdot 10^{16}$ molecules H_2SO_4 cm^{-3}, denoted in Figure 2 by letters *A* and *B*, respectively. *Pierce et al.* [2010] found median radii of $r = 0.096$ and 0.250 μm, which compare to $r = 0.106$ and 0.846 μm at $C = 10^{16}$ and 10^{17} cm^{-3} in this study. The considerable dissimilarity at the higher concentration is basically due to a steep gradient at $C = 10^{17}$ cm^{-3}, as we find $C \approx 7 \cdot 10^{16}$ to yield particles of $r \approx 0.3$ μm.

Based on the findings of Figure 2, the following growth dynamical regimes (numbered on the figure) may be distinguished:

1. Particle size would exceed $r \approx 0.10$–0.15 μm before the end of the early growth period (see above).The combined effects of the size dependence of sedimentation and scattering efficiency would lower the cooling versus sulfur mass ratio.

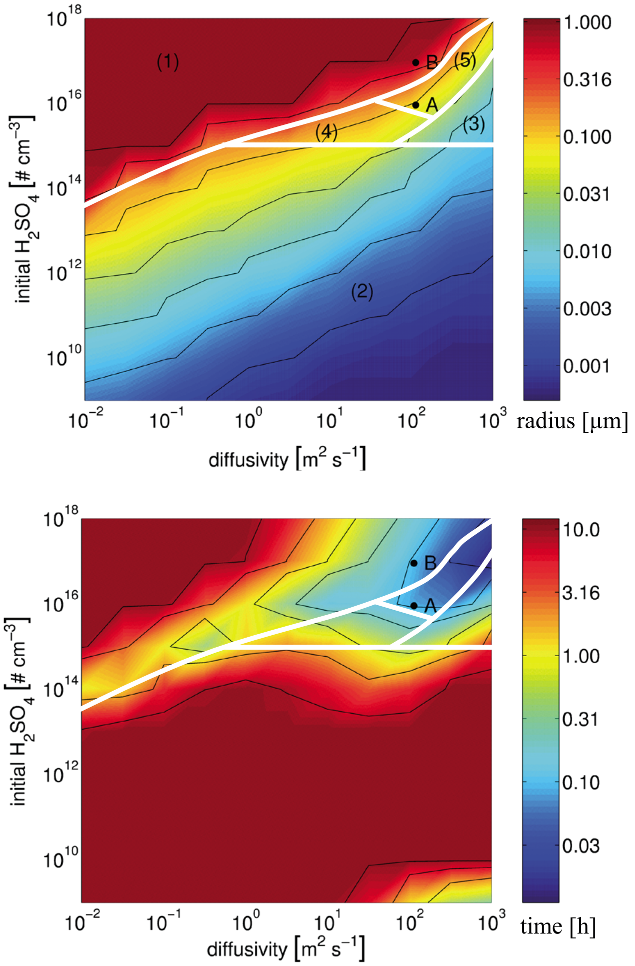

Figure 2. Number-median particle radius as background concentration is reached, or after a 12 h period if background concentration is not reached any earlier (top), and the corresponding time interval for particles to reach this size (bottom), both as a function of the air diffusivity coefficient and the initial concentration of H_2SO_4. Slow dilution experimental conditions of *Pierce et al.* [2010] are denoted by letters A and B. Characteristic growth dynamical regimes are delimited by white lines and the corresponding labels (1) to (5) are shown in Figure 2 (top). See text for further details.

2. Unless a complimentary precursor gas were used, the amount of sulfur ($M > 1\,\mathrm{Mt\,S\,a^{-1}}$) that could be released with a realistic order of magnitude of dedicated fighter jets ($N < 10{,}000$) would not be sufficient for SSRM to have the intended impact on global average temperature. Particles might eventually attain optimum size; however, for the larger fraction of the corresponding parameter space this is subject to uncertainty. As a result of their lengthy early growth period, substantial particle losses due to interaction with background aerosol might occur, and this might eventually imply a low ratio of cooling to injected sulfur mass.
3. The use of a precursor gas is not a necessity, and the early growth period is relatively short; however, the growth of the particles to size is uncertain as particles reach background concentration earlier. Considerable interaction with background particles is likely and efficient size control seems unlikely.
4. Particles definitely grow to size before they reach background concentration. However, their early growth takes a considerable amount of time, which might result in a considerable amount of interaction with background particles due to airmass mixing.
5. The initial growth period is very short; all criteria for steerability are fulfilled.

Table 1 summarizes the properties of the above growth regimes with respect to the five criteria for steerability of SSRM during the early growth period.

Table 1. Steerability Criteria Fulfilled for Each Growth Dynamical Regime (See Text)

Regime	Criterion				
	Self-Limiting	Radius	Species	Concentration	Time
1	Yes	No	Partially	Partially	Partially
2	Yes	Possibly	No	Possibly	No
3	Yes	Possibly	Yes	No	Possibly
4	Yes	Yes	Yes	Yes	No
5	Yes	Yes	Yes	Yes	Yes

5. Discussion and Conclusion

The exploration of the parameter space relevant to the early growth dynamics of an aerosol engineered for SSRM has allowed us to distinguish five characteristic regimes. Each of these regimes stands for a distinct level of fulfillment of the five criteria that were previously derived as sufficient to ensure the steerability of SSRM as given by early growth dynamical considerations.

In this analysis, regime (5) is the one in which all criteria are fulfilled. It is characterized by a relatively narrow parameter space as well as steep gradients for both particle size and the early growth period. This circumstance might translate into technical difficulties, since its implementation might prove to require a level of accuracy and adaptation to variable local conditions that is difficult to achieve. Regime (5) would require the input of an amount of turbulent energy that is typically found in the wake of a plane turbine. Planes have been considered to be a realistic option for injection, but open technical, logistic, and financial questions cast doubt on these assessments. For instance, the very high initial concentration of H_2SO_4 that corresponds to regime (5) might put limitations on the technical specifications and achievable working lifetime of the injection device. Technical development and experimentation would be required to determine the actuality and the persistency of these potential difficulties.

Particles within regime (4) should grow to size before they reach background concentration. Regime (4) would thus offer the advantage that no additional injection of a precursor gas would be required, although the logistic effort would generally be considerably larger than in regime (5). However, the particle initial growth period would be relatively long, such that the interaction with background particles may limit the steerability of this regime. Moreover, regime (4) would require a source of turbulent energy, which likewise to regime (5) puts a constraint on the choice of the injection method.

The interaction with background particles is likely to be considerable within regime (3), whose particles do not grow to size before they reach background concentration. The question of the steerability of regime (3) may thus remain unanswered until large-scale experimentation has been performed. Within parts of regime (2) the growth to full size appears likely. However, as it would require the additional input of a precursor gas, it is likely that its steerability is restricted and its optimization would also require extensive large-scale experimentation. Finally, due to size considerations, it appears likely that regime (1) would not be applicable for efficient implementation of SSRM.

There are several uncertainties which would need to be resolved if more refined results than the ones presented in this study were desired. In particular, some potentially relevant small-scale factors were not taken into consideration, such as the water vapor requirements for efficient particle nucleation. Also, the use of a 3-D plume model would help resolve the formation and dilution of the newly formed particles more accurately. Moreover, this study has aimed exclusively at determining early growth technical constraints on the steerability of SSRM. In doing so, it did not treat any long-term effects that result from the complex interactions of aerosol formation and growth with atmospheric circulation, chemical composition, and the energy balance, which make up the climate system as a whole. Long-term effects will influence the efficiency of SSRM, and steerability may be obtained with other means, such as the use of a precursor gas, and the spatial and temporal variation of the injected mass of sulfur. Still, the early growth constraints to steerability found in this study are principally self-contained to an extent that it seems unlikely that any of the above mentioned long-term interactions would have a relevant influence on their reality. In this respect, this study should provide a relevant contribution to the exploration and assessment of the idea to use SSRM as an efficient climate intervention technique.

Acknowledgments
The authors would like to thank the anonymous reviewers for their insightful comments and suggestions that have contributed to improve this paper. This work was supported by the EU Seventh Framework project IMPLICC. We note that there are no data sharing issues since all of the numerical information are provided in the figures and are the result of own modeling work.

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