



RESEARCH ARTICLE

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Key Points:

- This study describes a method of converting sulfur to sulfur oxides onboard an aircraft to form stratospheric sulfate aerosols for solar geoengineering
- Onboard conversion could roughly halve the mass transported to the stratosphere for a given radiative forcing, substantially reducing costs
- Direct costs of deployment are likely much smaller than indirect costs and should play a minor role in decisions about geoengineering

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Production of Sulfates Onboard an Aircraft: Implications for the Cost and Feasibility of Stratospheric Solar Geoengineering

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Abstract Injection of sulfate aerosols into the stratosphere, a form of solar geoengineering, has been proposed as a means to reduce some climatic changes by decreasing net anthropogenic radiative forcing. The cost and technical feasibility of forming aerosols with the appropriate size distribution are uncertain. We examine the possibility of producing the relevant sulfur species, SO₂ or SO₃, by in situ conversion from elemental sulfur onboard an aircraft. We provide a first-order engineering analysis of an open cycle chemical plant for in situ sulfur to sulfate conversion using a Brayton cycle combustor and a catalytic converter. We find that such a plant could have sufficiently low mass that the overall requirement for mass transport to the lower stratosphere may be reduced by roughly a factor of 2. All else equal, this suggests that—for a given radiative forcing—the cost of delivering sulfate aerosols may be nearly halved. Beyond reducing cost, the use of elemental sulfur reduces operational health and safety risks and should therefore reduce environmental side effects associated with delivery. Reduction in cost is not necessarily beneficial as it reduces practical barriers to deployment, increasing the urgency of questions concerning the efficacy, risks, and governance of solar geoengineering.

1. Introduction

Solar geoengineering, or solar radiation management (SRM), is a deliberate intervention to increase the Earth's albedo on a global scale. It has the potential to partially offset the radiative forcing caused by long-lived greenhouse gases, thereby reducing some of the risks of climate change (Crutzen, 2006; Keith & Dowlatabadi, 1992; National Research Council, 2015). There are, however, physical and social/political risks inherent in SRM, which are an area of serious concern and of active research (Schäfer et al., 2015; Shepherd, 2009). Solar geoengineering has gained interest in recent years given the cost and slow rate of mitigation, as well as uncertainties surrounding abrupt climate change (Parson, 2017). One method of implementing SRM is to inject aerosol into the lower stratosphere, thereby scattering a small additional quantity of incident solar radiation back to space. Several different types of aerosols have been proposed, including sulfates, titania, calcium carbonate, and engineered nanoparticles (Dykema et al., 2016; Keith, 2010). Most analysis of SRM has focused on increasing the concentration of sulfate aerosols in the lower stratosphere and midstratosphere through organized research efforts such as the Geoengineering Model Intercomparison Project (Kravitz et al., 2013). The most important questions about solar geoengineering concern its efficacy, risks, and governance. However, the cost and feasibility of delivering sulfates to the stratosphere remain uncertain (Davidson et al., 2012; Keith, 2013; Moriyama et al., 2016; Niemeier et al., 2011; Robock et al., 2009). While the direct cost and technical complexity are relevant to economic and political analysis of the barriers to deployment, it is crucial to recognize that these direct costs may be small compared to the costs of monitoring or to the scale of the benefits and harms of any deployment.

Almost all previous research on sulfate aerosol geoengineering has assumed that sulfur was delivered as SO₂, though a few studies have considered using H₂S to reduce the total mass lofted (Robock et al., 2009). A schematic of several options for dispersing sulfates is shown in Figure 1. Pierce et al. (2010) suggested that release of SO₃ or H₂SO₄ in an aircraft wake could create smaller particles, increasing the radiative forcing for a given mass of sulfur by pushing the aerosol size distribution closer to the light scattering optimum. We examine the possibility of converting elemental sulfur to SO₂ or SO₃ onboard an aircraft. If the mass of conversion equipment is low compared to the mass of sulfur, then such a conversion system could reduce the mass lofted by a factor of 2–3 compared to a base case in which SO₂ or H₂SO₄ were lofted directly. We conduct a preliminary

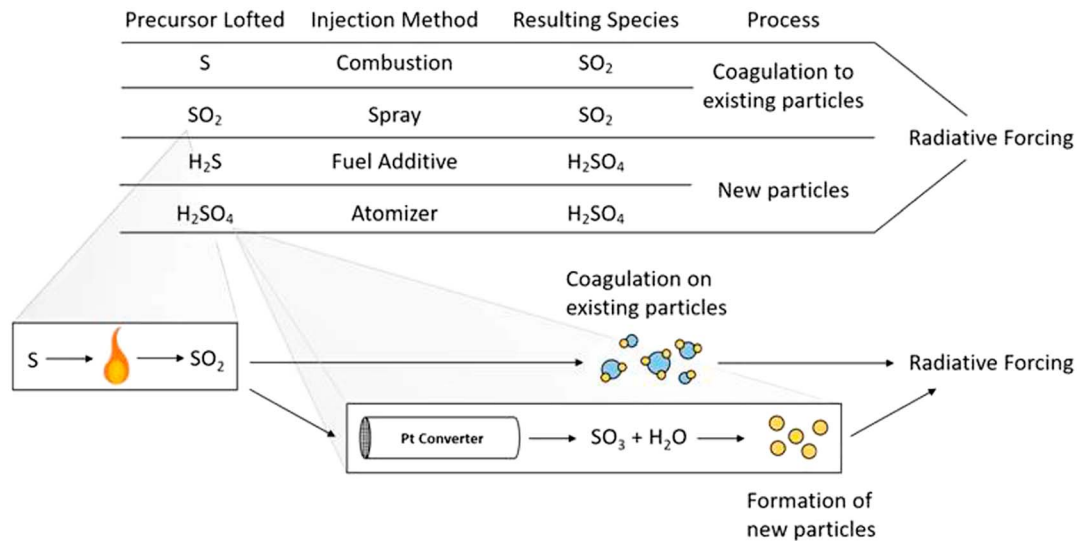


Figure 1. Diagram of different possible sulfate precursors, injection methods, and particle properties, ultimately resulting in radiative forcing.

engineering feasibility assessment for developing a compact, low-mass chemical plant capable of converting elemental sulfur to SO₂ or SO₃ onboard an aircraft flying in the lower stratosphere.

The remainder of this paper is structured as follows. Section 2 provides context for this engineering study with a brief literature review. Section 3 describes the engineering assessment, and section 4 assesses implications for estimating the cost of sulfate aerosol geoengineering.

2. Materials and Methods

In this section, we review three inter-related topics: (1) introducing sulfate aerosol precursors to the lower stratosphere and resulting radiative forcing efficiency, (2) the chemistry and toxicity of various sulfur compounds, and (3) technology for lofting materials to relevant altitudes.

2.1. Introducing Sulfate Aerosols Into the Lower Stratosphere Resulting in Radiative Forcing

Models of stratospheric sulfate aerosols have been developed over decades to understand the impact of volcanic sulfur. The radiative forcing of stratospheric sulfate aerosols is strongly dependent on their size (Rasch, Crutzen, & Coleman, 2008). Below the optimal size for Mie scattering, $\sim 0.4\text{-}\mu\text{m}$ diameter, the scattering efficiency per unit mass drops sharply, whereas larger particles are both optically inefficient scatterers and have shorter stratospheric lifetimes due to accelerated settling velocity.

Most analyses of sulfate aerosol geoengineering assume that the injected species would be SO₂, as is the case for volcanoes. SO₂ is a gas under stratospheric conditions and therefore does not directly form aerosols. Instead, SO₂ oxidizes to SO₃ over several weeks and then quickly reacts with H₂O to form H₂SO₄ (McElroy, 2002). Estimates of the radiative forcing from a continuous injection of SO₂ into the tropical lower stratosphere (the most efficient location for injection) suggest that a total injection rate, on an annual basis, of $20\text{--}40 \times 10^6$ metric tons of SO₂ (denoted Mt-SO₂/year hereafter) would be needed to produce global radiative forcing of -4 W/m^2 , a benchmark level that roughly corresponded to the CO₂ doubling often used in modeling studies. Although -4 W/m^2 represents a useful benchmark for model evaluation, this is equivalent to the *full* forcing at approximately year 2030 of the Representative Concentration Pathway 8.5 scenario and 2055 of the Representative Concentration Pathway 6.0 scenario (Moss et al., 2010). In contrast, engineering analyses of potential costs typically evaluate forcings of -1 to -2 W/m^2 , reflecting a perspective that SRM is an imperfect and uncertain complement to conventional mitigation. We define the radiative efficacy for sulfur E as radiative forcing divided by the injection rate of the injected sulfate species (W/m^2)/(Mt/year).

For injection of SO₂, E decreases with increasing injection rate because an increased fraction of the additional sulfur adds to the size of existing aerosols, making them too large (Heckendorn et al., 2009; Rasch, Tilmes,

et al., 2008). Unlike SO_2 , emission of SO_3 or H_2SO_4 into stratospheric plumes is followed by gas-phase and heterogeneous processes that produce new accumulation mode particles, keeping the overall particle size distribution closer the optimum (National Research Council, 1998). Applying this principle, an alternative proposed by Pierce et al. (2010) is to release sulfuric acid (H_2SO_4) into an aircraft wake where the rapid condensation of H_2SO_4 forms new aerosols, allowing the particle size distribution to be tuned by adjusting the injection rate. Findings from both in situ stratospheric observations and microphysical modeling have shown inadvertent formation of H_2SO_4 in high-altitude plumes during subsonic flight and provide supporting evidence for the mechanisms underlying this proposed approach (Fahey et al., 1995; National Aeronautics and Space Administration, 1997). The specific assumptions of Pierce et al. (2010) have been critically reexamined and confirmed in large measure by Benduhn et al. (2016). Using an aircraft plume model coupled to a 2-D global model, Pierce et al. (2010) found that the H_2SO_4 injection scheme would decrease mean particle size, which could significantly increase E .

To produce a radiative forcing of -4 W/m^2 , for example, the injection rates for H_2SO_4 and SO_2 are roughly 30 Mt- H_2SO_4 /year and 40 Mt- SO_2 /year, respectively. These injection rates are relevant to the computation of E associated with the delivery method proposed in Pierce et al. (2010), because sulfur constitutes 50% the mass of SO_2 but only 33% the mass of H_2SO_4 . For this reason, using H_2SO_4 results in a twofold reduction in sulfur mass per unit radiative forcing. This result depends on aerosol formation in the aircraft wake (Benduhn et al., 2016; Pierce et al., 2010) and will not occur if H_2SO_4 is initially distributed evenly over large volumes (English et al., 2012).

An important condition of the Pierce et al. (2010) proposal is that the linear injection rate, sulfur per kilometer of aircraft track, must be limited to produce an optimal size distribution for light scattering. This requires aircraft to cruise in the stratosphere, whereas for release of SO_2 , there is no obvious limit to injection rate since the gas requires weeks of mixing time before oxidizing to form particles. Pierce et al. (2010) found that a release rate of 1–10 kg S/km as H_2SO_4 would eventually form aerosols with $\sim 0.30\text{-}\mu\text{m}$ diameter after stratospheric dilution. According to this study, fluctuations in injection rate of $\pm 10\%$ will result $\pm 5\%$ fluctuations in median particle radius. For this injection method, the difference in median particle radius between strong and weak aircraft wake turbulence is more significant, in the range of 20–30%.

2.2. Chemistry and Toxicity of Sulfur Compounds Relevant to Solar Geoengineering

We compare the characteristics and risks of three sulfur compounds that might be transported to the stratosphere in aircraft: elemental sulfur, SO_2 , and H_2S . Molten sulfur is commonly transported in insulated tanks at around 150°C (d'Aquin & Fell, 2007), has density of 1.8 kg/L, and vapor pressure below 1 mbar, presenting a moderate exposure and fire risk. SO_2 is a gas at standard atmospheric conditions, and while it can be compressed, it would likely be transported as a chilled liquid onboard an aircraft. At a temperature of -10°C , SO_2 has a density of 1.5 kg/L and a vapor pressure of 1 bar. Hydrogen sulfide (H_2S) gas could similarly be transported as a liquid either at high pressure (17 bar at 21°C) or at low (-60°C at 1 bar) temperatures.

In practice, the degree of risk management measures would vary significantly depending upon the chemical product lofted. In the event of an accident or crash, SO_2 and H_2S stored as liquefied gases would vaporize and disperse rapidly, posing an immediate health hazard over a significant area. Both gases are acute airborne toxins, and H_2S gas would also pose a fire hazard. Accidents involving liquid S would have lower health, safety, and containment risks because of its much lower vapor pressure and the fact that it solidifies as it cools.

2.3. Technology for Lofting Materials to the Lower Stratosphere

A variety of methods have been suggested for delivering materials to the lower stratosphere for geoengineering. The use of naval rifles to loft artillery shells containing sulfates was suggested in a U.S. National Academy study (National Research Council, 1992), and rockets were considered in Katz (2010). More recently, fixed tower chimney designs were examined, though tethered balloons might be cheaper (Davidson et al., 2012).

While it is possible that these methods may prove useful, aircraft provide a technologically mature option to loft relevant quantities of material to these altitudes (Blackstock et al., 2009). McClellan et al. (2010) report the results of a consulting study by Aurora Flight Sciences, which used industrial aviation costing methods to

evaluate a range of aircraft options. We highlight two options from McClellan et al. (2010). First, a fleet of around 45 reengineered business jets could deliver 1 Mt/year to a maximum altitude of 18 km at a cost of about 2.15 USD/kg, with upfront acquisition and development investments of 3.2bUSD. Second, for large mass delivery rates or higher altitudes, a fleet of new purpose build aircraft could be developed that would deliver 5 Mt/year to 24 km at a cost of about 0.6 USD/kg after a larger \$13.5bUSD initial investment. While these aircraft would be new designs, they would not require novel technology for airframes or engines, consistent with analysis in a recent Delft University study (Design Synthesis Exercise Group 2, 2016). We use the reengineered aircraft and flight criteria from the Aurora study as a baseline in section 3. Note that a new analysis by Wake Smith shows that the McClellan et al. (2010) assumptions about flight altitude for reengineered business jets are inaccurate and that a new airframe design would be required (Smith, 2018).

McClellan et al. (2010) use similar cost estimating methods to reexamine the cost of rocket, gun, or tethered balloons, finding that costs for rocket and guns were 10–100 times greater than those of aircraft, so we ignore them here. Tethered balloons might compete with aircraft but pose a greater technical risk. However, the economics of tethered balloons depends on high dispersal rates per balloon, which would produce much higher local concentrations of the dispersed species than aircraft that release material continuously along a flight track. A high delivery rate does not matter for SO₂ or other species that are emitted as gases and slowly convert to a condensable form, but it will not work for H₂SO₄ or similar condensable species or solid particles for which aircraft are likely the only feasible means of delivery. We therefore confine our assessment to aircraft.

3. Engineering Analysis

For an in situ conversion system to provide a mass benefit, the combined weight of elemental sulfur and the conversion plant along with any fuel or consumables required must be less than the mass of the product (SO₂ or SO₃). The lighter the system, the better its efficiency relative to directly lofting the product. The size and mass of a chemical plant will increase with throughput, which we express as the conversion rate of elemental sulfur to products in units of kg/s. The required conversion rate is determined, in turn, by the total mass of product divided by the duration of the “cruise” segment of the flight during which the sulfur compounds are released. For a fixed target mass injected per flight, the shorter the cruise duration, the higher the required conversion rate and more massive the converter.

A convenient figure of merit in determining the advantage of onboard conversion is the mass of conversion equipment divided by the conversion rate, which we call the payback time, p , the time required for the conversion system to process its own mass.

The fractional mass of a complete system relative to dispersed sulfur species, f , is the sum of the in situ conversion system and feedstock mass divided by the mass of the delivered product. A system that lofts SO₂ and releases it without in situ conversion would have f_{SO_2} slightly larger than 1 because the mass of SO₂ and the container would be slightly larger than mass of SO₂ alone. This fraction is determined by the molar mass ratio of payload to product, r , along with the flight duration, D , and p , as follows:

$$f = r \left(1 + \frac{p}{D} \right) \quad (1)$$

Note that this expression for f is equivalent to $r \times (\text{mass of elemental S} + \text{mass of device}) / (\text{mass of elemental S})$. Ignoring the container mass, if the payload was elemental sulfur and the product was SO₂, then $r = 0.5$ since SO₂ has twice the molecular mass of S. If, for example, the payback time was 10³ s and the flight duration was 10⁴ s, then the conversion equipment mass is 10% that of sulfur, and $f_{\text{SO}_2} = 0.55$. That is, the total payload mass is only 55% of what it would have been in the base case where SO₂ was transported to the stratosphere. If, however, the payback time was 10⁴ s so that $(p/D) = 1$, then the mass of processing equipment would be equal to the mass of sulfur and f would be unity, and the in situ conversion system would provide no advantage with regard to mass.

A chemical plant for producing the relevant sulfates, SO₂, SO₃, or H₂SO₄, in situ would necessitate different equipment depending upon the target chemical product. For SO₂, a compressor must pressurize ambient airflow and feed it to a combustion chamber for reaction with molten sulfur. Producing SO₃ is more difficult as it requires the burner outflow to be cooled with a heat exchanger before entering a catalytic converter,

where the SO_2 present in the inflow gas oxidizes as it passes over a catalyst. H_2SO_4 is formed rapidly when SO_3 mixes with water vapor from the engine exhaust in the aircraft wake, so we focus on SO_2 and SO_3 . Our objective is to produce a rough estimate of total mass of a system that produces SO_2 or SO_3 at a given rate for a given duration, where that unit comprises the feedstock and any necessary conversion equipment.

For this scaling and feasibility analysis we focus on the ratio of chemical throughput relative to conversion system mass and energy requirements. As a reference scale, we follow the McClellan et al. (2010) analysis of a reengineered Gulfstream G550/G650 delivering a payload of 10,000 kg H_2SO_4 per sortie at 0.85 Mach over a cruise of 3 hr (10,800 s) at an altitude of 20 km (0.05 bar atmospheric pressure) and -60°C . This is equivalent to an injection rate of 0.3 kg S/s, which we use as a reference scale production rate. The mass of chemical processing equipment typically increases with throughput as a power law with an exponent of about 0.6 (Caduff et al., 2011), so although we do not address the scale trade-off here, larger aircraft payloads will allow conversion systems with lower payback time.

3.1. Sulfur to Sulfur Dioxide

Industrial sulfur burners are prohibitively heavy for use in an aircraft. We propose a reengineered jet engine combustor design as a suitably low-mass alternative and assume that this is configured as a conventional fuel-burning turbine engine (Brayton thermodynamic cycle; Farokhi, 2014), burning sulfur as fuel. A basic compatibility assessment provides evidence that aero-derivative combustion chambers could serve as a lightweight substitute for the purposes of in situ sulfur combustion.

The combustion of sulfur to form sulfur dioxide is a strongly exothermic reaction with a stoichiometric air-to-fuel ratio of 4.3:1 by mass and an adiabatic flame temperature of roughly 1900°C (Cameo Chemicals, 2015; Hasenberg & Bender, 2008). Sulfur melts at 119°C , and its flash point is 207°C , with autoignition occurring at 232°C (ConocoPhillips, 2016). Commercially available sulfur burners were designed as a component of industrial sulfuric acid plants, and their design has evolved to minimize capital and operating costs rather than equipment mass or volume. The two main types of commercial sulfur burners are spiral and cyclone flame burners. Compact cyclone flame models have payback time of about 5,000 s, and therefore, assuming linear scaling of mass and processing rate, the burner mass exceed that of sulfur burned during a 3-hr flight (NORAM International AB, 2014).

Use of ambient pressure would eliminate the need for a compressor, but it would require a much larger combustor, and while operating a combustor at 0.05 bar is possible, we were unable to find a reliable engineering analog that would achieve complete combustion under ambient stratospheric conditions. Given the need for pressurized product (SO_2 -rich air) in the case where the desired final product is SO_3 (section 3.2) we simply assume that the combustor must operate at 1 bar absolute pressure rather than exploring the mass trade-offs for sulfur combustion over a wide range of operating pressures.

The fact that this process requires compressed air and generates power from fuel combustion suggests that the system is not dissimilar from gas turbine engines consisting of a compressor that draws outside air into the engine, a combustion chamber that burns fuel in air, and an expander that extracts usable work from hot, high pressure air. If a turbine was designed for high resistance to sulfidic corrosion, the compression and combustion components could be simplified to an open Brayton cycle engine powered exclusively by sulfur combustion. We first analyze the thermodynamics and then discuss sulfur compatibility and mass estimates.

Nominal 0.05 bar air pressure at cruise altitude requires a pressure ratio of roughly 20:1 to reach 1 bar. Together, pressure ratio and inflow air mass determine the total compressor work required for a given fuel flux. A combustion chamber sized exactly for the air-to-fuel ratio would be as small and light as possible, but may result in incomplete combustion of sulfur. However, complete combustion would produce a combustor exit gas temperature of approximately 1900°C , exceeding the upper bound of temperature-rated limits for modern turbines (Kroschwitz & Grant-Howe, 1997). As described below, we assume 1500°C to be a reasonable upper bound for operating temperatures.

We model the system as an ideal open Brayton cycle as shown in Figure 2. A turboshaft engine for helicopters provides a useful design reference. We use the ideal gas law for compression and expansion, with fixed heat capacities of $c_p = 1.01 \text{ kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ for inlet gas and $c_p = 1.16 \text{ kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ for exhaust to account for the different gas compositions and elevated temperatures (Chase, 1998; Lemmon et al., 2000).

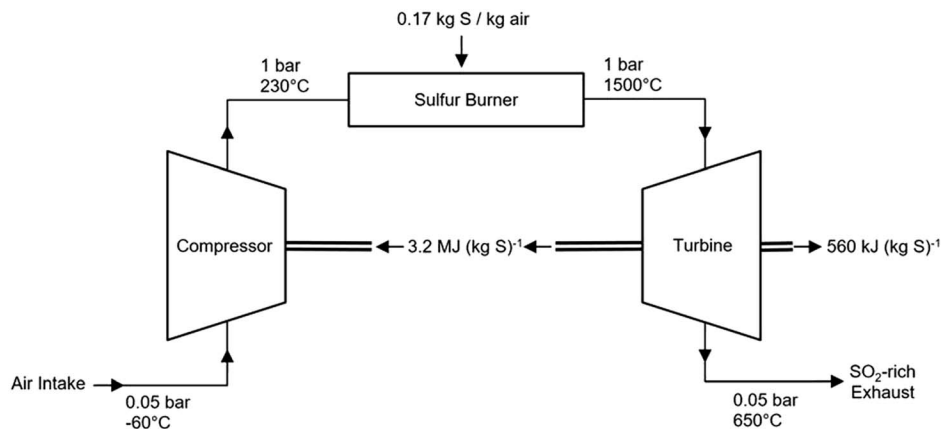


Figure 2. Process flow diagram of proposed SO_2 production system. This is a simple Brayton Cycle engine burning sulfur. Temperature and pressure are shown at each stage; energy flows are normalized by the mass flow of sulfur; for example, the work delivered is 560 kJ for each kilogram of S consumed, or $560 \text{ kJ} \cdot (\text{kg S})^{-1}$.

We assume that the compressor and expander each have efficiency of 55% (Nkoi et al., 2013). We believe that this is a conservative (low) estimate given existing turbojet engines (Gray, 1978; Koff, 2004; Wilson & Korakianitis, 2014).

We adjust the air-to-fuel ratio, increasing excess air to reduce temperatures to a specific maximum air temperature for the expander inlet. For a turbine inlet temperature of 1500°C , the air-to-fuel mass ratio is 6:1 (140% theoretical air). This air-to-fuel ratio requires a flow rate of 2.1 kg S/s to produce an injection rate of 0.3 kg S/s . Under these conditions, the turbine generates more work than the compressor requires, and therefore, the system produces $560 \text{ kJ} \cdot \text{kg}^{-1} \text{ S}$ of net work. Should this air-to-fuel ratio result in incomplete combustion of sulfur, the power generated by the turbine provides a generous margin to increase airflow without ancillary power.

A survey of existing turbine materials indicate that the sulfur combustion concept described here is feasible, but would require customized materials and further research and development in turbine materials technology. Titanium alloy turbine blades containing nickel, cobalt, lead, and tungsten have demonstrated considerable resistance to sulfidation, though turbine blades in commercial circulation are not specialized for such high concentrations of sulfur oxides (ASM International, 1997). While an additional coating or advanced composite material might increase the cost of the turbine, the exact details of such modifications are beyond the scope of this analysis and therefore account for no change in mass owing to such modifications.

We now compare characteristics of kerosene and sulfur combustion to evaluate whether a lightweight combustor could be designed to process sulfur. Aircraft engine combustors are designed for kerosene-based jet fuel; hence, the design has been engineered for combustion properties different than those of sulfur. Sulfur combustion produces a 9 MJ/kg enthalpy change, which is 4.6 times less than the 43 MJ/kg typical of jet fuel such as Jet A-1 (Chase et al., 1985; Edwards, 2002). However, the stoichiometric air-to-fuel mass ratio (100% theoretical air) for jet fuel is roughly 15:1, about 3.5 times higher than the 4.3:1 ratio required for sulfur. The enthalpy change of combustion per unit air mass is therefore quite similar, with sulfur combustion producing about 75% the heat produced by kerosene for the same airflow when both are burned at their stoichiometric air-to-fuel ratios.

For this analysis, we assume that an aircraft combustor could be redesigned for sulfur by keeping the airflow and geometry constant and increasing the fuel flow by a factor of 3.5. Sulfur has a lower vapor pressure than kerosene, raising questions as to whether such designs would burn sulfur efficiently. A related consideration is whether the velocity of combustor inflow air would need to be reduced to ensure flame stability.

Annular combustors used in aeronautics applications are designed to provide uniform flame distribution with low pressure drop for high-mass and volume-specific combustion rates (Mattingly, 2006; Zuyev & Skubachevskii, 2013). As a guide to the difference in fuel characteristics, a comparison of sulfur to various gas turbine fuels is provided in Table 1. Many modern gas turbines used for stationary power applications are closely derived from aircraft turbine engines and operate using residual fuel oils with viscosity and

Table 1
Fuel Properties and Descriptions, Including Heavy Residual Oils Commonly Used in Marine Engines

Fuel type		Flash point (°C)	Autoignition temp (°C)	Viscosity (m ² /s × 10 ⁻⁵)
Kerosene	Blended light distillate hydrocarbon fuel	38–74 °C	229 °C	0.5 (150 °C)
Sulfur	Elemental sulfur preheated to molten S	207 °C	232 °C	8 (150 °C)
Bunker B	No. 4 Distillate obtained from heavy gas cut used as commercial heating oil in burners without preheating	55 °C	263 °C	5–24 (100 °C)
	No. 5 Light Residual oil distilled to exceed the heating value of fuel oil No. 4 while maintaining similar viscosity without preheating	55 °C	NS	5–9 (100 °C)
	No. 5 Heavy Residual industrial heating oil obtained from heavy gas oil cut; requires preheating to 77–104 °C for proper atomization	55 °C	NS	9–15 (100 °C)
Bunker C	No. 6 Least valuable cuts of crude with significant impurities form a high-viscosity residual fuel; preheating to 104–127 °C is advisable	66 °C	407 °C	15–50 (100 °C)

Note. Given that residual fuel oils are a blend, temperature of minimum viscosity is not constant, impacting combustion chamber size and preheating. Autoignition temperature is not well defined for fuel oil No. 5 and is therefore not specified (NS) (American Society for Testing and Materials, 1986; Cameo Chemicals, 2015; Curl & O'Donnell, 1977; d'Aquin & Fell, 2007; Perry et al., 1963).

autoignition temperatures similar to sulfur. These precedents suggest that sulfur could be used with minimal modifications to fuel injector designs in existing aero-derivative gas turbines.

Small aero-derivative turboshaft engines such as the Pratt & Whitney Canada PT6 can serve as a benchmark for estimating mass. The PT6A-68 has 5.3 kg/s compressor mass flow, pressure ratio of 11:1, generating 1,365 kW of shaft power, and weighs 251 kg (Badger et al., 1994). We use a two-step power law scaling to derive a mass estimate. First, we scale the mass flow with a power law exponent of 0.9 to obtain an updated shaft power estimate of 590 W for 2.1 kg/s flow, and then scale the shaft power with an exponent of 0.7 to obtain a mass estimate of 140 kg (Caduff et al., 2011). There are competing adjustments to this mass estimate. It is likely an overestimate because our system has a lower maximum operating pressure, reducing structural requirements, and the lower power production could tolerate a lighter gearbox. However, considering the approximately 3 times higher fuel flow of sulfur versus kerosene, as well as the 3 times higher compression ratio required for our application, mass may instead increase (Leyes & Fleming, 1999). We judge that the overall uncertainty of this mass estimate is better than ±30%.

A system weighing 140 kg implies payback time of 470 s. Under these assumptions, the fractional mass of producing SO₂ using a sulfur-driven open Brayton cycle engine would be $f_{\text{SO}_2} = 0.52$, nearly twice as efficient as direct lofting. If SO₂ is the desired injection species, these design considerations provide strong support that such a system could be designed with minimal modifications to existing commercially available supplies.

3.2. Sulfur Dioxide to Sulfur Trioxide

Oxidation of SO₂ to SO₃ is exothermic with a reaction enthalpy of 98 kJ/mol, about one third that of S to SO₂. Although thermodynamically favorable, the reaction proceeds too slowly to be useful on timescales required for in situ production. The reaction is faster at higher temperatures, but the equilibrium shifts away from SO₃ toward SO₂. At 1 bar, an initial mixture of 8.2% SO₂ and 91.8% standard air by volume will have an equilibrium fraction of 67% SO₃ at 700 °C and 48% SO₃ at 900 °C (Näumann & Schulz, 2008). Efficient conversion of SO₂ to SO₃ therefore requires lower temperatures that are typical for formation of SO₂ and a catalyst is required to achieve practical conversion rates. Additional hardware enabling SO₃ production can be added to the SO₂ production system by adding a catalyst bed and thermal conditioning system. A complete system schematic is shown in Figure 3. Once SO₃ is produced, SO₃-rich gas is reheated to provide additional enthalpy during expansion.

Industrial sulfuric acid plants use multiple catalyst beds with intercoolers and gas recycle loops to achieve SO₂ → SO₃ conversion above 95%. It would be difficult for low-mass aircraft systems to achieve similarly high conversion fractions. We choose a single-pass design and target a 70% conversion fraction. As we discuss in section 3, this reduction in conversion fraction probably has little impact on the ability of SO₃/H₂SO₄ injection to alter aerosol size distribution.

Large industrial sulfuric acid production systems now use vanadium catalyst (Lloyd, 2011), but the literature suggests that platinum, which was used in earlier industrial applications (Wells & Fogg, 1920), has a higher

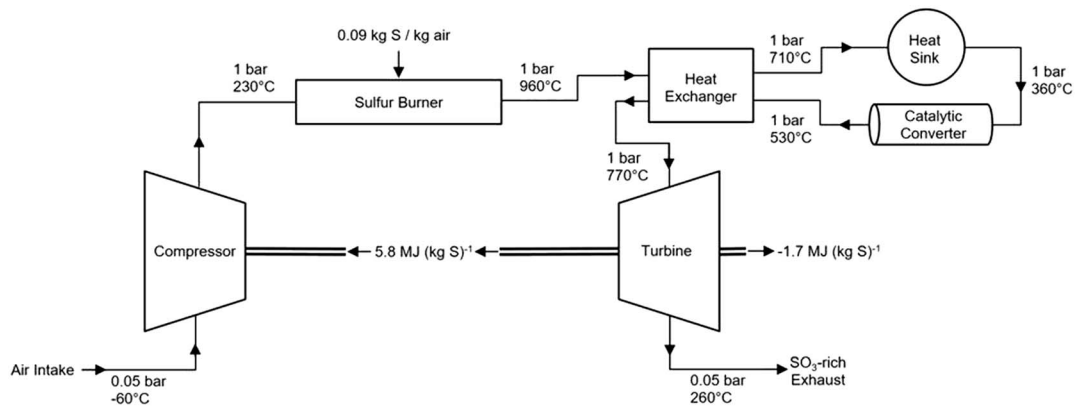


Figure 3. Process flow diagram of proposed SO_3 production system. The system is a Brayton cycle modified to include a counter flow heat exchanger and a catalyst bed for converting SO_2 to SO_3 . Temperature and pressure are shown at each stage; energy flows are normalized by the mass flow of sulfur; for example, the work deficit is -1.7 MJ for each kilogram of S consumed, or $-1.7 \text{ MJ} \cdot (\text{kg S})^{-1}$.

mass-specific catalysis rate and is therefore more suitable for low-mass systems (Koutsopoulos et al., 2006; Xue et al., 1996). We base our catalyst choice and converter configuration on recent experimental results from compact microchannel reactors for SO_2 oxidation. Benzinger et al. (2011) demonstrated single-pass conversions to SO_3 in a platinum-based reactor with titanium walls designed to remove the heat of reaction to increase the conversion fraction. Since such reactors are not commercially available, we size our design based on automotive ceramic monoliths with the same channel geometry ($0.2 \text{ mm} \times 0.2 \text{ mm}$ channels 80-mm long) and assume the same Pt/ TiO_2 coating, conversion efficiency, and $530 \text{ }^\circ\text{C}$ exit temperature (Corning Inc., 2016; Kikuchi et al., 1999). Applying existing ceramic monolith specifications to our reference scale, a conversion rate of 0.3 kg S/s would require a 1.6 m^3 monolith weighing 440 kg. We add 20% mass to account for converter shell and piping yielding $p = 1,740 \text{ s}$.

Oxidation of SO_2 to SO_3 requires additional oxygen, raising the required air-to-fuel ratio to a theoretical minimum of 6.4:1. Drawing from experimental data, a microconverter design described in Benzinger et al. (2011) suggests that the mass concentration of O_2 to SO_2 should be roughly 1:1 to achieve sufficiently rapid conversion. We therefore increase the sulfur air-to-fuel ratio to 11:1, almost double the 6:1 ratio used in section 3.1.

The additional excess air decreases the combustor exit temperature to $960 \text{ }^\circ\text{C}$, easing concerns regarding materials compatibility, but converter inlet gas temperature must still be reduced to reach the 70% conversion target. The heat removed from the combustor gases is used to heat the SO_3 -rich gas exiting the converter, increasing the work generated by the expander using a heat exchanger as shown in Figure 3. Theoretical research (Juhasz, 2010; Paeng et al., 2010) and development efforts (Yoshimura et al., 1995) suggest that a ceramic heat exchanger can meet these requirements. Based on this work in ceramic materials and supported by the upper temperature range of an existing system detailed in Banerjee et al. (2015), we develop a system as follows. Heat exchange would occur within a cross-flow heat exchanger (recuperator), followed by an air-to-air heat sink using outside air to dispose of additional waste heat prior to entering the converter.

For the purposes of this assessment, we ignore the modest pressure drop in the converter and heat exchangers such that air enters the turbine at 1 bar. Based on the production of SO_3 from SO_2 and temperature-independent heat capacity of $c_p = 1.06 \text{ kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$, the converter temperature increase would be over $160 \text{ }^\circ\text{C}$ for adiabatic first-pass conversion.

The thermal conductivity of the recuperator and heat sink designs shown in Figure 3 would cool the combustor exit gas from 960 to $360 \text{ }^\circ\text{C}$, the temperature required to yield a converter exit gas at $530 \text{ }^\circ\text{C}$ (Lienhard IV & Lienhard, 2008). Existing heat exchanger systems for turbine applications utilize silicon nitride, which have favorable thermal and mechanical properties for our operating temperature range and thermal gradients. We calculate that a silicon nitride shell-tube recuperator based on the design of Yoshimura et al. (1995) could achieve a heat transfer coefficient $U = 170\text{--}180 \text{ J} \cdot \text{s}^{-1} \cdot \text{m}^{-2} \cdot \text{K}$. Applying these parameters to the 0.3 kg S/s

combustion reference scale, the design would have surface area of 45 m², 0.1-mm tube thickness, effectiveness ϵ of roughly 60% and provide heat transfer Q of 1 MW, cooling the exhaust gas from 960 to 710 °C, and warming postconverter gas from 530 to 770 °C.

Recuperator exit gas must be cooled further to meet the 360 °C catalytic converter design requirement. Considering the abundance of −60 °C ambient air, potential use of highly conductive heat transfer mediums, and equipment integration options for meeting energy demand in other parts of the aircraft, we assume that required disposal of $Q = 1.2$ MW waste heat from 710 °C gas is achievable at a heat transfer coefficient of $U = 100 \text{ J} \cdot \text{s}^{-1} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ (Min et al., 2009; Sunden, 2005).

A recuperator with 45 m² surface area weighing roughly 155 kg could condition the temperature of air exiting the combustor. Based on the heat transfer coefficient of our design, a heat sink would require under 25 m², which we estimate would add 40 kg mass. Considering 30% additional mass for fittings and casing, the heat exchanger system would have total estimated mass of 250 kg ($p = 830$ s). Adding converter mass, the entire system would weigh 780 kg ($p = 2,570$ s). Increasing the air-to-fuel ratio requires the compressor and expander system to be enlarged to accommodate mass flow of 3.6 kg/s. Applying the same two-part power law scaling to the PT6A-68 for increased mass flow suggests a compressor and expander system weighing 190 kg.

At a turbine inlet temperature of 770 °C, the turbine lacks sufficient shaft work to power the compressor, and therefore, despite the heat exchanger, some ancillary power is required. For simplicity, we assume that supplemental power is supplied using a conventional kerosene-fueled turboshaft engine. Using the PT6A-68, scaled using the approach outlined above, could provide 1,670 kJ · kg^{−1} S compressor shaft work for our 0.3 kg S/s reference flow rate at a weight of 130 kg and fuel consumption of 0.039 kg/s. Such a design would require 420 kg of kerosene over the cruise duration, and therefore, the auxiliary compressor plus fuel have combined payback time of 1,820 s. The resulting payback times for the compressor and expander (630 s), converter (1,740 s), thermal conditioning system (830 s), and auxiliary power system (1,820 s) total 5,020 s. Given the molar mass ratio r of 0.4 for S/SO₃, the fractional mass of the SO₃ in situ production system is $f_{\text{SO}_3} = 0.59$.

3.3. Sulfur Trioxide and Sulfuric Acid

The onboard production of H₂SO₄ from SO₃ and H₂O would add significant mass and volume, reducing the overall mass benefit. In principle, this penalty could be avoided by lofting H₂ gas and producing H₂O in situ, but such a design would add complexity and mass even though the mass of H₂ is negligible.

When dispersed in the aircraft wake with sufficient water vapor, SO₃ reacts rapidly with H₂O to form H₂SO₄ vapor, so onboard conversion is likely unnecessary if sufficient water vapor is available. We estimate that the water vapor produced as by-product of fuel combustion in the engines would likely be sufficient. At a reference dispersal rate of 0.3 kg · s^{−1} S, the required water vapor production is 0.2 kg/s H₂O. McClellan et al. (2010) assumes a fuel flow rate of 1 kg/s for the dispersal aircraft, yielding ~2.5 kg/s excess water vapor, roughly 10 times the amount required for reaction with SO₃ (Penner et al., 1999). Using an in situ system such as that described, and assuming that the 30% residual SO₂ remaining from 70% SO₂ to SO₃ conversion eventually combines with other aerosols to form H₂SO₄, 10 tons could be produced from about 4,800 kg ($f_{\text{H}_2\text{SO}_4} = 0.44$) of hardware and sulfur.

4. Discussion

Our analysis suggests that if SO₂ or H₂SO₄ were ever transported to the stratosphere by aircraft and released at altitudes around 20 km in quantities relevant to solar geoengineering, it may be possible to reduce the overall aircraft payload by roughly a factor of 2 by transporting elemental sulfur in molten form, along with a chemical conversion system to produce either SO₃, which is rapidly converted to H₂SO₄ in the aircraft wake, or SO₂. In addition to reducing the costs and environmental impacts associated with aircraft operations, this would also reduce operational health and safety risks.

Independent from any reduction in the mass lofted to the stratosphere, the use of H₂SO₄ injection to control aerosol size distribution may reduce environmental impacts by reducing the mass of sulfur required for a given radiative forcing by a factor of 2 or more (Pierce et al., 2010). Reduction of total sulfur burden is important since most key environmental risks are proportional to the total amount of sulfur.

We estimated performance for a system that converts 70% of S into SO₃, which is then converted to H₂SO₄ in the aircraft wake, yet prior analysis assumed pure SO₃/H₂SO₄ (Pierce et al., 2010). The reason H₂SO₄ has been considered is that it can produce new aerosol particles, constraining the overall aerosol size distribution. The aerosol size distribution resulting from release of SO₂, which oxidizes to H₂SO₄ in situ, depends on the ambient aerosol size distribution. It is plausible that as long as more than about half of the sulfur mass is released as H₂SO₄, the eventual oxidation of the fraction of sulfur remaining as SO₂ will result in an aerosol size distribution roughly comparable to the distribution produced if all the sulfur mass were released as H₂SO₄. Initial unpublished numerical experiments with a stratospheric aerosol model support this hypothesis.

This engineering feasibility study is far from a final engineering design. Materials engineering challenges with the catalyst and sulfur tolerance of high-temperature components could reduce the performance compared to our estimate. Conversely, our system is not optimized, and there are multiple ways in which further engineering might yield lower mass designs. For example, we do not take advantage of the fact that the airflow into the converter would be pressurized by a ram effect on intake air, substantially decreasing the compressor work and mass.

The development of in situ conversion systems such as the one described would have implications for the cost of solar geoengineering. Here we consider only the direct operational costs of injecting aerosol precursors, and ignore the cost of the sulfur Brayton cycle hardware, as the cost would be negligible relative to other expenses such as the aircraft fleet. Other costs of solar geoengineering, such as monitoring, may contribute more to the overall operational cost. Moreover, other factors, such as the risk and benefits of solar geoengineering, will almost certainly be more important in any decisions about deployment.

We base our cost analysis on a recent comprehensive evaluation by Moriyama et al. (2016) that expressed direct costs as

$$F \left(\frac{L f}{E} \right) \quad (2)$$

where L is the amortized cost of delivering mass to the stratosphere in USD per kg or billions of USD per Mt, F is the radiative forcing (W/m²), and E is the radiative forcing efficacy (W/m²)/(Mt/year) defined in section 2.1. Here, as in Moriyama et al. (2016), the injection rate is the mass flux of the target sulfur species (e.g., SO₂ or H₂SO₄) measured in millions of metric tons per year. Finally, f is the total mass lofted divided by target species mass as defined in equation (1). Although elemental sulfur is somewhat cheaper than sulfate products, we ignore procurement cost of sulfur compounds, as the difference is not enough to significantly alter the annual cost of deployment.

Following Moriyama et al. (2016), and using results from Pierce et al. (2010) that include consistent, detailed microphysics for both SO₂ and H₂SO₄ from Figure 2 of their paper, representative estimates of E required to produce a radiative forcing of -2 W/m² are 0.108 ± 0.06 and exactly 0.193 (W/m²)/(Mt/year) for SO₂ and H₂SO₄, respectively. Using the f values for SO₂ ($f_{\text{SO}_2} = 0.52$) and H₂SO₄ ($f_{\text{H}_2\text{SO}_4} = 0.44$), the resulting estimates of the total lofted mass required to produce a radiative forcing of -2 W/m² are 9.6 and 4.6 Mt/year for SO₂ or H₂SO₄, respectively. While the values for radiative forcing efficacy and aircraft costs are uncertain, the costing relationship in equation (2) is linear in f , so that the total cost is reduced by roughly a factor of 2 by using in situ conversion when one ignores the nonlinear dependence of total cost with mass flux due the economies of scale for aircraft operations.

Moriyama et al. (2016) compiled cost estimates from a number of sources to derive a representative cost per unit of global radiative forcing of ~ 45 (bUSD/year)/(W/m²). If use of in situ conversion halved the lofted mass, costs would be reduced to ~ 22.5 (bUSD/year)/(W/m²), or roughly 45 bUSD/year to achieve a radiative forcing of -2 W/m². We note that the aircraft engineering results of McClellan et al. (2010), which form the basis for many of the cost estimates presented in Moriyama et al. (2016), have been broadly confirmed by a recent study from Delft University (Design Synthesis Exercise Group 2, 2016), increasing confidence in these cost estimates. An important caveat is that altitude is a major driver of costs, and therefore, altitudes significantly above 25 km would correspond to higher costs than those estimates presented, as engineering complexity and fleet size would increase.

Lower costs are not necessarily a benefit. Any technological advancements that significantly lower the financial burden or technological challenges of solar geoengineering also lower barriers impeding private or sovereign actors from implementing such measures unilaterally (Barrett, 2008; Schelling, 1996; Weitzman, 2015). Furthermore, one potential estimate of damages from solar geoengineering is in the range of 10–100bUSD (Horton et al., 2015). Low barriers to climate engineering pose not only an alarming geopolitical prospect but also concerns from an ethical, economic, and ecological perspective (Buck, 2012; Horton & Reynolds, 2016; Parson & Ernst, 2013).

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