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Trade-Offs between Sulfuric Acid Aerosol Precursors for Stratospheric Aerosol Geoengineering

James Franke,* Wake Smith, Angela Slavens, Frank N. Keutsch, Fangqun Yu, and David Keith

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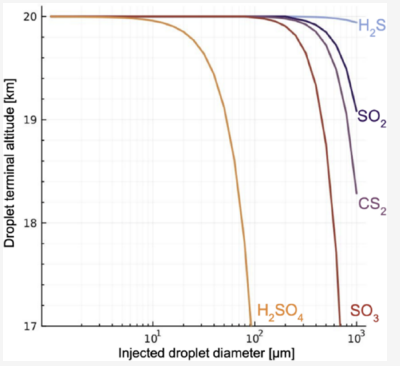
ABSTRACT: Solar geoengineering with sulfuric acid aerosols in the lower stratosphere might be used to reduce the impacts of climate change. Almost all studies have assumed sulfur is transported aloft as sulfur dioxide; several other sulfur compounds are plausible. We evaluated elemental sulfur (S_8), hydrogen sulfide (H_2S), sulfur dioxide (SO_2), carbon disulfide (CS_2), sulfur trioxide (SO_3), and sulfuric acid (H_2SO_4), examining the mechanisms of dispersal in liquid form, safety, and costs for materials supply and deployment with a 20 km altitude aircraft. Levelized, combined costs for material production and lofting at 100 ktonne/year scale range from $1140 to $5150/tonne-S. We find that the cost to loft H_2S is about half that of SO_2, but the production cost of H_2S is more uncertain and could be high enough to change this conclusion. The additional oxidation step required for H_2S adds <20% to the aerosol formation time scale, so H_2S and SO_2 should produce similar aerosol size distributions if new particle formation is negligible. H_2S and SO_2 have high and roughly equivalent toxicity. H_2S presents a further flammability risk that SO_2 does not. S_8 would be much safer than H_2S or SO_2, but it requires airborne combustor technology that is not yet developed. CS_2 is not preferred on any metric. SO_3 and H_2SO_4 are ∼4 times more expensive than H_2S to loft but offer additional control over the aerosol size distribution that will likely reduce some side effects of sulfuric acid aerosol geoengineering.



KEYWORDS: *sulfur, SAI, climate intervention, sulfate*

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INTRODUCTION

Stratospheric aerosol injection (SAI) is a potential climate intervention whereby reflective aerosol particles or their precursors would be intentionally deployed continuously in the lower stratosphere to cool Earth. Almost all SAI studies have assumed that gaseous SO_2 is carried aloft to the stratosphere and released, where it slowly oxidizes to SO_3 over several weeks[1] and then quickly hydrates to gaseous H_2SO_4. Gaseous H_2SO_4 in turn forms liquid sulfuric acid aerosols that persist in the stratosphere on the order of 1 year.[2]

SO_2 is highly toxic and cannot be feasibly carried aloft as gas in a pressure vessel due to the vessel wall thickness required to contain the pressure. Lofting potentially millions of tonnes of material to 20 km altitude yearly is not trivial. With plentiful oxygen and hydrogen already in the stratosphere, the limiting element—and that which sets the overall radiative forcing—is sulfur. Several other commercially relevant compounds may also serve as the source of sulfur for the same oxidation−hydration aerosol formation pathway. What these compounds have in common is that, due to their short tropospheric lifetimes, they cannot effectively be released in the troposphere and instead must be transported to the stratosphere. S_8, burned in situ to SO_2,[3] H_2S,[4] and CS_2 all oxidize at varying rates to SO_3 and then quickly hydrate to H_2SO_4. In contrast, SO_3 or H_2SO_4 do not require oxidation after vaporization and hence form sulfuric acid aerosol more directly and controllably.

Each sulfuric acid aerosol precursor has trade-offs in mass efficiency (the amount of sulfur per total compound mass), ease of distribution, cost, and safety. While some existing economic analyses of SAI implementation exist,[4−7] an analysis comparing different sulfuric acid precursor compounds has not yet been conducted. We consider six commercially relevant sulfur compounds here (listed in Table 1, with key material properties). Inclusion is limited to those compounds naturally occurring in the stratosphere except carbonyl sulfide (OCS) and dimethyl sulfide (DMS). OCS is very slow to be photolyzed such that the stratospheric lifetime is several years,[8] and although natural DMS emissions to the troposphere are substantial, industrial production is very small (and starts with H_2S as feedstock), and provides no weight benefit over SO_2. Here, we compare the deployment feasibility, safety, and economics of procurement and lofting each compound. Economic comparisons in this work are made per tonne of sulfur, as this is the relevant metric in terms of total amount of sulfuric acid aerosol formed in the stratosphere.

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Table 1. Key Sulfuric Acid Precursor Material Properties[a]

		slow-acting			fast-acting	
	$S_8 \rightarrow SO_2$	H_2S	SO_2	CS_2	SO_3	H_2SO_4
mass compound/mass sulfur	1	1.1	2	1.2	2.5	3
freezing point@1 bar (K)	392	187	200	162	290	283
boiling point@1 bar (K)	718	212	263	319	318	611
transport temperature (K)	392	211	262	293	308	293
vapor pressure@293 K (bar)		18	3.2	0.5	0.3	10^{-6}
vapor pressure@transport T (bar)	10^{-4}	1	1	0.5	0.7	10^{-6}
vapor pressure@216 K (bar)		1.2	0.08	0.03	~0.01	
ΔH_{va} (kJ/mol)	63.2	18.7	24.9	26.8	46.7	67.6
specific gravity, liquid	1.80	0.92	1.43	1.26	1.92	1.84

[a]Each compound would be carried aloft at the listed transport temperature (see text for rationale). Vapor pressure at 20 °C (293 K) is representative of ambient conditions at the airfield, and vapor pressure at 216 K is standard atmosphere for 20 km altitude in the tropics. Specific gravity of the liquid is reported at its transport temperature: either chilled, heated, or surface ambient (20 °C). ΔH_{va} is the latent heat of vaporization. Slow-acting gaseous precursors form sulfuric acid aerosols mainly by oxidizing and condensing on existing particles over several weeks or longer. Fast-acting compounds form aerosols in seconds when vaporized in the aircraft exhaust.

The only lofting technology mature enough for plausible implementation within a few decades is fixed-wing aircraft, so our study is limited to this deployment method.[5,9,10] Additionally, we focus only on sulfuric acid aerosols because they are comparatively well-studied in the stratosphere,[11] and existing sulfur emissions to the troposphere are large compared to proposed SAI levels.[8] We therefore do not include nonsulfur materials[12−14] or other solid particles, which, despite their potentially advantageous radiative and chemical properties,[15,16] present distinct engineering challenges.

MATERIALS AND METHODS

Sulfuric Acid Precursor Injection and Aerosol Formation

We divide sulfuric acid aerosol precursors into two categories: high-vapor-pressure compounds, which predominantly slowly oxidize and condense on existing background particles after being widely dispersed by stratospheric circulation, and low-vapor-pressure compounds, which can nucleate immediately to form new particles in the aircraft wake (Table 1).

Low-vapor-pressure, fast-condensing compounds offer the advantage that they can be used to generate new aerosols in the accumulation mode[17−19] that may steer the overall stratospheric aerosol size distribution closer to the optimum.[20] If the aerosol size distribution is closer to the optimum, then the radiative forcing efficacy, or the amount of radiative forcing per unit of injected sulfur, can be increased, which may reduce various side effects associated with SAI.[21] The disadvantage of these low-vapor-pressure compounds is that they require more mass per unit sulfur (i.e., higher total molecular weight), and they must be distributed more slowly to control particle size,[19] which increases the cost of aircraft operations. We consider two low-vapor-pressure compounds: H_2SO_4 and SO_3. SO_3 combines almost instantaneously with water to make H_2SO_4, and so we assume they will act quite similarly.

We consider three high-vapor-pressure compounds: SO_2, H_2S, and CS_2. Almost all prior research on SAI, including the modeling of the environmental impacts, has assumed that the compound released would be SO_2. Here we address the question: what differences might arise in using H_2S or CS_2, and to what extent can we assume that the prior research results hold with one of these alternate compounds?

The lifetimes of SO_2 and H_2S vary widely in the stratosphere, while the ratio between the two lifetimes is stable. Both are oxidized by OH, and variation in the oxidation time scale is dominated by variation in OH concentration. Across relevant stratospheric conditions (210−230 K and 50−100 hPa), the oxidation of H_2S is 7−10 times faster than that of SO_2.[22] The larger ratio corresponds to 50 hPa, due to the pressure dependence of the OH + SO_2 reaction.[22] The temperature dependence of the reactions is similar enough that it does not affect the relative lifetimes over relevant stratospheric temperatures.

An important determinant of the aerosol size distribution is the time it takes the emitted high-vapor-pressure gas to be transformed into low-vapor-pressure species that will condense to form aerosols. Considering the chain from H_2S to H_2SO_4 compared to SO_2 to H_2SO_4, the reaction sequence starting with H_2S is at most 20% slower. This, in turn, should result in a relatively small difference in size distribution if most of the added sulfate ends up on existing particles, as is the case with background sulfate emissions.[23] However, in some cases there can be considerable new particle formation from injection of a high-vapor-pressure gas in the first few days when plume concentrations are still high. In that case, H_2S may be less effective at making new particles than SO_2 because, while the overall reaction chain to H_2SO_4 is only ~20% slower, the time delay, plume dilution, and nonlinear dependence of nucleation rate on H_2SO_4 vapor concentration might reduce the opportunity for the injected sulfur to make new particles.

Finally, per Mt of injected H_2S, 0.5 Mt of H_2O are produced in its oxidation to SO_2, which represents a negligible perturbation of the annual water flux into the stratosphere, e.g., compared to the 150 Mt estimated to be injected by Hunga Tonga,[24] or the 250 Mt/year from convection.[25] Additionally, as injected H_2S ultimately turns into H_2SO_4 aerosols, it uses up that water, so there is no direct net water flux to the stratosphere.

The oxidation of CS_2 is more complicated. There is a branch in the reaction pathway wherein half of the sulfur ends up making OCS,[26] which has a much longer lifetime in the stratosphere.[27] The formation of aerosols from CS_2 would therefore be quite different from that of the SO_2 reference.

The overall forcing efficiency of each compound is largely determined by the resulting aerosol size distribution. We do not compare aerosol kinetics or formation sizes in detail here. For the remainder of the analysis, we will compare compounds within these categories (fast vs slow acting) to each other but not across categories except to note some relative economics of theoretical aerosol sizes in the discussion. Key material properties for each category of precursor can be found in Table 1.

Engineering Trade-Offs. Pressure vessels required to confine compressed gases during loading and ground operations, where temperatures may exceed 30 °C, could weigh much more than the gas itself (e.g., an example pressure vessel for H_2S would weigh nine tonnes for just over 1 tonne of compressed gas), making it economically improbable. We therefore assume that all sulfur compounds will be carried aloft in tanks as a liquid. This means that gases such as SO_2 and H_2S must be chilled cold enough so that their vapor pressure is <1 bar. We note also that standard engineering practice would dictate that chilled, liquefied gases be carried on aircraft in pressure containers rated for full vapor pressure given an insulation failure and subsequent warming. However, liquefied hydrogen airplanes are already being operated with an apparent relaxation of this requirement.[28] Special relief devices and/or certifications may be needed for this approach.

We assume H_2S and SO_2 are chilled and carried at temperatures ~211 and ~262 K, respectively. Industry standard practice would be

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to carry SO_3 slightly elevated at 308 K to avoid freezing in the presence of impurities and to carry S_8 molten above 393 K. H_2SO_4 and CS_2 can be carried as liquids at ambient surface temperature, assumed 20 °C (293 K). The aircraft climb time is approximately 30 min. We assume no additional mechanical cooling or heating is required to maintain transport temperature within tolerance. Some heat tracing may be required for distribution lines for S_8. Material vapor pressures at transport temperatures are shown in Table 1.

Precursor liquids, stored in a stratospheric aircraft cruising at an altitude of 20 km, must be pumped from their tanks and sprayed either directly into ambient air or into jet exhaust. The cost of delivering material to the stratosphere is minimized if the material can be released quickly because a longer cruise burns more fuel, reduces payload, and reduces the number of cycles an aircraft can complete per day (see the Lofting Cost section). However, a droplet released into the stratosphere will fall as it evaporates, so spraying too quickly may result in material lost to sedimentation. Evaporation rates are different for various compounds (see the next section for details).

Very rapid dispersal could theoretically be achieved by pumping through a large-diameter hose, which would produce relatively large droplets, perhaps on the order of 1 mm after a breakup in the airstream. We assume that there is no limit to the rate at which slow oxidizing precursors could be dispersed, assuming that droplet evaporation can be assured. That is, we assume that there is no advantage to spreading gaseous SO_2, H_2S, or CS_2 over a longer flight path because any of these gases will have time for substantial mixing before they are oxidized and can form aerosols. Smaller droplets will evaporate more quickly but are more difficult to produce quickly because they require specific spray nozzles with very low flow rates, thus either necessitating a longer cruise time or additional weight and complexity owing to a large array of nozzles and associated pumps.

Rapid dispersal is not possible, even given fast evaporation, if the goal is new sulfuric acid aerosols produced by SO_3 or H_2SO_4 directly in the aircraft wake, where faster injections lead to larger (less efficient) aerosols.[19] This is a separate, higher-level trade-off that we do not examine comprehensively, in which the radiative advantage of optimum-sized aerosols is weighed against the added cost of flying long flight tracks. In the next sections, we compute how far a droplet falls before it evaporates, as a function of the initial droplet size.

Vaporization of Droplets. As injected liquid droplets fall, they quickly cool as they evaporate, and slow down as they shrink. We estimate evaporation and sedimentation rates for different droplet-starting sizes based on vapor pressure and Stokes' law. Larger initial droplets will take longer to evaporate and thus fall further in the limit, several kilometers and out of the stratosphere. We do not account for large droplet breakup in the airstream, secondary coagulation, or recondensation on remaining droplets, all of which would require a full 3D CFD simulation, which is beyond the scope of this analysis. This assumption holds best for low injection rates, where particle density is low. Evaporative mass transfer rate (Q_m) is estimated from vapor pressure by eq 1.

$$Q_m = MKA\left[\frac{P_{sat}}{TR} - \frac{P_{inf}}{T_{inf}R}\right] \quad (1)$$

where M is the molar mass, K is the mass transfer coefficient, A is the droplet surface area, P_{sat} is the saturation vapor pressure at the current temperature (T) of the droplet—estimated with Clausius−Clapeyron, R is the universal gas constant, and P_{inf} is the background partial pressure of the compound, and T_{inf} is the temperature of the medium. The mass transfer coefficient for a spherical droplet in flow can be estimated with the Sherwood number (eq 2), which in turn can be estimated with the Reynolds (Re) and Schmidt (eq 3) numbers, all of which are dimensionless.

$$Sh = 2 + 0.6Re^{1/2}Sc^{1/3} \quad (2)$$

$$Sc = \frac{\nu}{D} \quad (3)$$

where ν is the kinematic viscosity, and D is the mass diffusivity of the medium. The background partial pressure of each compound in the stratosphere is effectively zero, and the mass diffusivity of air is scaled from the near-surface value with pressure to 20 km conditions of 0.05 bar. Terminal settling velocity for each droplet is based on Stokes' law adjusted for Cunningham slip.

Only H_2S has a sufficiently high vapor pressure at stratospheric temperatures to be naively sprayed quickly at almost any droplet size (Figure 1). SO_2 and CS_2 droplets will evaporate quickly if sprayed

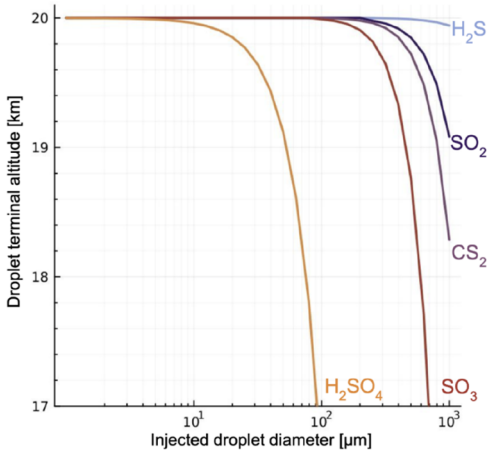

Figure 1. Evaporation/sedimentation dynamics for precursor compounds as a function of injected droplet diameter. Droplets are sprayed with no additional heating and assuming no secondary coagulation. Droplet terminal altitude is that at which evaporation is complete. Evaporation rates are estimated based on vapor pressure at the current droplet temperature (estimated from Clausius−Clapeyron). Sedimentation velocity is estimated with Stokes' law, corrected for Cunningham slip. SO_3 is assumed not to hydrate until evaporation. H_2SO_4 must be sprayed in very small droplets to persist in the stratosphere without active vaporization.

below 500 μm diameter. H_2S, SO_2, and CS_2 can be vented directly out the rear of the aircraft through an array of tubes and nozzles.

We assume here for simplicity that SO_3 droplets do not hydrate until vaporized (which would only slow down evaporation) and that vapor pressure can be scaled to 216 K with Clausius−Clapeyron. We are not aware of any published data on vapor pressures of SO_3 at temperatures below 333 K.[29] Detailed modeling of the SO_3 hydration and freezing processes after injection is outside the scope.

H_2SO_4 particles will not persist near 20 km unless injected at droplet sizes well below ~20 μm diameter (Figure 1). Spraying droplets at 200 μm is straightforward with commercially available nozzles; spraying quickly below 20 μm is not (see Supporting Information Section S3). We therefore assume an additional heat source is needed to actively vaporize H_2SO_4 and SO_3. Water and impurities in commercial H_2SO_4 will change evaporation dynamics slightly,[30] but not enough to change this conclusion. The obvious—and free—choice of heat source is the aircraft engine exhaust, which produces roughly 6500 kW of available heat, enough to vaporize an upper bound of 10 kg/s of H_2SO_4. (Assuming 100:1 air-to-fuel ratio, 6:5 water-to-fuel ratio, and a cruise burn of 0.16 kg/s.[19] See Lofting Cost section below for payload/injection/flight time trade-off.) Vaporizing H_2SO_4 or SO_3 in the aircraft exhaust stream would require engine modifications.

However, the aircraft exhaust stream only provides 0.2 kg/s of H_2O—enough water to instantly hydrate ~0.5 kg/s of SO_3. The remaining balance of SO_3 must disperse and be hydrated by the environment, which consists of only ~3−4 ppm of H_2O at 20 km in the tropics.[31] H_2O depletion from a 1 Mtonne-S/year (full-scale) SO_3 injection would amount to 0.2% of the background stratospheric water influx from convection,[25] but this in turn would be counteracted

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Table 2. Key Material Safety Metrics from the U.S. Environmental Protection Agency (EPA), National Institute for Occupational Safety and Health, and National Fire Protection Association (NFPA)[b]

	S_8	H_2S	SO_2	CS_2	SO_3[a]	H_2SO_4
AEGL-2, 1 h (ppmv)	NA	27	0.8	160	2.4	2.4
AEGL-3, 1 h (ppmv)	NA	50	30	480	40	40
LC50, 1 h (ppmv)	NA	712	2520	12,500 (4 h)	347	347
NFPA Health	2	4	3	3	4	3
NFPA Fire	1	4	0	4	0	0

[a]Exposure to SO_3 vapor is in the form of H_2SO_4 aerosols, so the same values are used for AEGL and LC50. [b]Acute exposure guideline levels: AEGL-2 is the concentration level at which serious and irreversible harm is likely, and AEGL-3 is the life-threatening level. Lethal concentration (LC50) is the lethal level to 50% of the test subjects after one hour. Larger numbers are better for concentrations.

by a stratospheric warming-induced water vapor feedback. Without detailed plume modeling, it is impossible to say that all SO_3 will form aerosols instantly when only partially hydrated in the aircraft exhaust. (One could also carry the requisite liquefied hydrogen, which would still be lighter overall than H_2SO_4, or new hydrogen aircraft engine technology may be another option to supply additional exhaust H_2O.)

Onboard Production of SO_2 or SO_3 from S_8. Molten S_8, even if sprayed in very small droplets, will not oxidize under stratospheric conditions. S_8 must be actively combusted to produce SO_2. Once combusted, SO_2 gas persists in the stratosphere and forms aerosols over several weeks. One possibility is to adapt a Brayton cycle gas turbine engine design to combust S_8 aloft, in which molten S_8 would be injected into the combustion chamber with air compressed to roughly 1 bar, at a ratio of 0.17 kg S/kg air.[3] Combustion enthalpy would generate enough work to compress the intake air (from 0.05 bar[3]). One challenge with this configuration is the quick atomization of molten sulfur to ensure complete combustion. Existing sulfur burners consist of large refractory chambers (not easily carried on an aircraft) for this purpose.

Two alternative sulfur-burner configurations may be possible. Solid S_8 powder may be directly combusted, drawing from the technical legacy of coal burners. This could relax the liquid atomization challenge but would pose separate material-handling challenges. It may also be possible to burn sulfur in an afterburner configuration in which atomized molten sulfur is sprayed into the hot exhaust stream of the engine. This afterburner configuration is simple (aside from the sulfur atomization) and provides additional thrust with low additional flown weight, but it may be difficult to achieve complete combustion due to the limited ability to control mixing and residence time. Comprehensive engineering analysis is required for all three proposed sulfur combustion methods.

Finally, it is theoretically possible to produce SO_3 on board from SO_2 with the addition of a catalytic converter. However, secondary in situ conversion to SO_3 is a much harder technical challenge than sulfur combustion,[3] so we assume that if SO_3 is the injected material, it must be produced on the ground and carried aloft as a liquid and vaporized in the aircraft exhaust as described above.

Material Safety

We focus on the acute safety implications of an accident with an SAI aircraft and ignore the chronic, environmental impacts of sulfate material released by such an accident because the amount released would be tiny compared to the chronic impacts of sulfate materials deliberately released by industry or SAI.[32] All sulfur compounds analyzed here are common in industry, so material safety is widely known, and industry-standard safe handling practices are well-established. However, these materials are not transported on a large scale via aircraft. We therefore consider safety aspects specific to aircraft operations, including leakage during normal operations and under accidental tank rupture. While property or equipment might be compromised in an accident, we focus here on risks to persons directly involved in SAI operations or those exposed in a crash.

Leakage or Spillage. While all sulfur compounds pose skin contact risks if spilled, the most extensive hazard is through inhalation of vapors. SO_2 and H_2S are highly toxic, with nearly zero acceptable levels of exposure. Since they act in different pathways—H_2S is a neurotoxin and SO_2 forms sulfuric acid in the lungs—their lethal concentration (LC50, median lethal dose) and acute exposure guideline levels (AEGL-3, life-threatening, EPA) levels differ with respect to which compound is more dangerous (Table 2). CS_2 is a flammable liquid at room temperature, and, while less acutely toxic than SO_2 or H_2S, would require similar safety precautions in practice because very low vapor concentrations are still deadly. Both SO_3 and H_2SO_4 are liquid at surface temperatures and have very similar hazard profiles, with a slightly higher vapor pressure for SO_3 (Table 1). SO_3 also poses a higher secondary fire risk because it acts as a powerful desiccant on organics and will react violently with water. SO_3 readily reacts with moisture in air to form H_2SO_4; thus, the risk of exposure to SO_3 is most likely inhalation of H_2SO_4 vapor. Elemental sulfur is by far the safest of the compounds considered, but note that S_8 can contain trace H_2S, which will slowly off-gas, so some precautions are required.

Molten sulfur is regularly transported in large quantities, and the risks of loading it on aircraft would be analogous to the risks of loading aviation kerosene. The risks of loading SO_3 and H_2SO_4 onto an aircraft would be higher than those of aviation kerosene and would require special moisture control and ventilation safety measures—as are common for high-concentration oleum and sulfuric acid in industry. The risks of transporting and loading either SO_2 or H_2S as a chilled liquid are sufficiently high that both would require extensive training, protective gear, special equipment to reduce and ideally eliminate leakage volumes, and safety monitors to detect leaks if they occur. All of these measures are common in the petrochemical industry for H_2S and SO_2, but SAI operations would likely need to occur in a segregated area of the airport, and would take place after fueling, maintenance, and ground inspections.

Tank Rupture. Airport personnel or the public could be exposed in the case of a ground collision, runway excursion, rejected takeoff, or crash that ruptures the material tank. We estimate the spatial extent of the hazard due to a crash using the U.S. Environmental Protection Agency (EPA) Areal Locations of Hazardous Atmospheres (ALOHA) software. We assume H_2S and SO_2 vaporize instantly upon crash. This is a conservative assumption because it takes considerable enthalpy to vaporize chilled liquid H_2S and SO_2, so the actual release rates would be lower than we assume and the plumes correspondingly smaller and less concentrated. S_8 is assumed to burn within 1 h of crashing, and H_2SO_4 and SO_3 are vaporized by jet fuel burning within 1 h, both of which are also very conservative assumptions. See supplemental sulfur report Section S4.5 for detailed modeling assumptions. An example toxicity threat extent for H_2S is shown in Figure 2, and summary hazard zone extents for airborne toxicity, blast, and flammability risk are listed in Table 3. Hyper-local distribution in a crash of liquid payload and fire propagation is beyond the scope of this study.

The most important difference between the tank rupture risk of H_2S and SO_2 is that H_2S is flammable. Aircraft or emergency response crews with breathing apparatus could be protected from the toxicity of H_2S or SO_2 but not the fire risk of H_2S. However, the aircraft is already carrying fuel, which poses significant fire risk. H_2S has lower risk than Jet A with respect to combustion enthalpy and autoignition temperature, but higher risk with respect to spark energy and vapor pressure. H_2S poses less fire risk than hydrogen and is likely comparable to liquefied natural gas—the handling of both of which is

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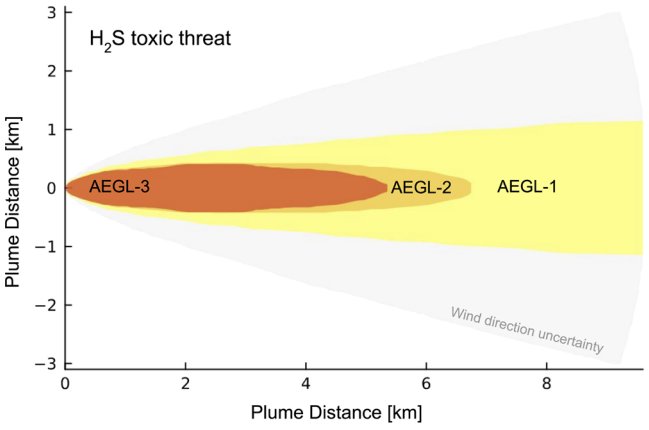

Figure 2. Example catastrophic crash zone hazard extent (km) from the EPA's ALOHA dispersion analysis program for toxicity for instantaneous H_2S release. The crash is assumed to occur over an urban or vegetated environment and to use weather conditions near Houston, TX: 3.3 m/s wind speed, 31.7 °C temperature, 50% RH, and no atmospheric inversion (see the supplemental sulfur report, Section S4.5, for more modeling assumptions). Crashes over open water (not shown) result in slightly larger hazard areas but presumably far less human risk. *x*-axis trimmed to 9.7 km (6 miles).

Table 3. Summarizes Threat Extents (Length of Zone in km in the Direction of Wind) for Each of the Compounds Based on the Same Crash Condition from Figure 2[a]

	S_8	H_2S	SO_2	CS_2	SO_3	H_2SO_4
toxic threat (AEGL-3)	1.6	5.1	4.4	1.2	0.6	0.6
flammable	local	0.8	N/A	0.7	local	N/A
blast force threat	N/A	0.3	N/A	0.4	N/A	N/A

[a]Larger number is worse. Analysis from EPA's ALOHA dispersion analysis program. SO_3 is not flammable but acts as a strong desiccant on organics.

now relatively common in industry and even being applied as fuel for experimental aircraft.[28,33] See supplemental Table S1 for material flammability metrics.

SO_3 and H_2SO_4 pose a significantly lower catastrophic crash risk than H_2S and SO_2, because their impact would be more localized, and neither poses a direct fire risk. S_8, if ignited by the crash, would pose a toxicity risk greater than 1 km downwind of the crash under the crash assumptions used here.

In practice, overall, H_2S carries a greater risk than SO_2, but the difference is unlikely to significantly change hazard management plans. This is because, while H_2S poses blast and fire hazards, both are much more localized than the toxicity (AEGL-3) hazard, so an unprotected person within the blast or fire risk zone for H_2S would be exposed to such high gas concentrations that they would be unlikely to survive, implying that the added fire risk is irrelevant except to those wearing specialized breathing apparatus. This judgment is reinforced by the fact that in the 216 H_2S-related workplace deaths in America since 1984, only two are due to fire (OHSA Database[34]).

For all sulfur payload options, engineering teams would have to devise strategies and procedures that sharply minimize the risk that aircraft could crash into populated areas. This could restrict the choice of airports from which such missions could be flown, such that SAI deployment flights would not mix on the taxiways and runways with passenger flights. The certification path to permit flight crews to routinely operate these aircraft in the manner described is unknown and beyond the scope of this paper. Comprehensive safety analyses must be conducted by specialized chemical safety experts. The relative cost of such safety measures is beyond the scope of this analysis but is likely a small percentage of operational expense.

Economics

Material Acquisition. More than 95% of global sulfur supply is produced from H_2S separated from "sour" gas or produced in the petroleum refining process via hydrodesulfurization.[35] H_2S is not typically transported because of its toxicity. Instead, it is either converted on-site to elemental sulfur or disposed of via acid gas injection. Elemental sulfur is widely available on the commercial market for ~$100/tonne (can vary substantially based on market dynamics) and global market production is currently approximately 85 Mt/year.[36] Production costs for the other sulfur compounds are very strongly dependent on the size of the operation. For a small-scale SAI deployment, we will assume >100 ktonne-S/year (to approximately double the background sulfur flux to the stratosphere, not including volcanoes[8]) at a single site; however, a moderate-forcing SAI campaign could require an order of magnitude more material and at least 2 sites for north−south hemispheric balance.

Most commercial sulfur is currently used to make H_2SO_4 in a process that entails burning to make SO_2, then oxidizing with a

Table 4. Central Estimates of Material Acquisition and Lofting Costs[a]

	S_8	H_2S		SO_2	CS_2	SO_3	H_2SO_4	
injection rate (kg material/s)	22	40		40	40	4	4.8	0.1
cruise time (s)	580	351		355	355	3202	2722	22,791
sulfur efficiency (kg material/kg S)	1	1.1		2	1.2	2.5	3	
lofted sulfur (kg/sortie)	12,760	12,760		7100	11,830	5120	4360	760
sorties (#/aircraft/year)	2130	2130		2130	2130	1650	1650	660
lofted sulfur (ktonne/aircraft/year)	29.5	29.5		16.4	27.3	8.5	7.2	0.5
fleet size (# aircraft)	4	4		7	4	12	14	200
lofting rate (ktonne S/year)	118	118		115	109	102	101	100
fleet cost (M$/year)	140	140		245	140	420	490	7000
lofting cost (M$/100 ktonne-S)	119	119		213	128	412	485	7000
material cost (M$/100 ktonne-S)	10	−5	440	15	80	27.5	30	
total (M$/year/100 ktonne-S)	129	114	559	228	208	440	515	7030

[a]Injection rates are given in kg material per second, not in kg sulfur per second. The injection rate for S_8 is calculated as the optimum based on combustor mass scaling. The injection rates for the high-vapor-pressure compounds are set at 40 kg/s (less than ~10 min cruise at 20 km for the assumed payload) because 7 sorties are the maximum possible per aircraft per day even for a sub-10 min cruise time. The injection rates for H_2SO_4 (and for SO_3) are set as the maximum rate (1.6 kg S/s, 4.8 kg H_2SO_4/s) and the rate that produces ~0.1 μm diameter aerosols (0.1 kg/s) based on using the engine configuration of Yu et al., 2024. The yearly lofting rate is based on whole aircraft at full utilization, which results in the indicated lofting cost. The 100 ktonne-S yearly lofting cost represents the fleet cost normalized to a lofting rate of 100 ktonne-S/year (i.e., not accounting for whole aircraft).

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catalyst to SO_3, and finally reacting it with water. Commercial H_2SO_4 is widely available at approximately $300/tonne-S (varies based on market dynamics). A 100 ktonne-S/year SAI operation at a single site would require approximately 1000 tonnes per day (TPD) of sulfuric acid—a medium-sized plant in commercial terms. We therefore assume SAI operations could either purchase H_2SO_4 on the commercial market or produce H_2SO_4 on-site at market cost. Next, we assume that a dedicated plant constructed on site could produce SO_2 and SO_3 at a cost between the sulfur feedstock ($100/tonne-S) and finished sulfuric acid ($300/tonne-S). We can therefore estimate $150/tonne-S for SO_2 and $275/tonne-S for SO_3 (because the oxidation process from SO_2 to SO_3 is the costlier step).

H_2S has a much smaller commercial market, and so its production cost is much less certain. We could not find a modern study of H_2S production economics at the >100 kt-S/year scale relevant here. We therefore provide three cost estimates for H_2S. First, on the high end, currently operating small-scale (<50 TPD) H_2S production facilities using the Unitel-Bechtel[37] reaction process have market prices on the order of $4400/tonne-S (Randhava, personal communication, October 23, 2024).

Second, we can crudely estimate on-site production of H_2S on a 300 TPD production line by analogy with the widely used Claus sulfur recovery process in which H_2S is converted into S_8. Several industry sources suggest that the Claus process[38] can produce S_8 from zero-cost H_2S at ~$150/tonne.[39] We assume that the levelized capital and operations and maintenance (O&M) costs of an H_2S production plant are also ~$150/tonne-S—where O&M excludes the cost of input sulfur and hydrogen. We additionally assume $1500/tonne of H_2 (~$90/tonne of H_2S), which is less than the current cost of large-scale production from cracked natural gas and more than the $1/kg target of the DOE's "Hydrogen Shot" for 2031.[40] Under these assumptions, the levelized cost of H_2S is $340/tonne-S. This estimate turns on our assumption that the levelized capital and O&M costs of H_2S production are the same as those of Claus process plants for making sulfur from H_2S. The Unitel H_2S process has fewer thermal steps than the Claus process and no catalytic conversion, so there is some basis for our assumption that capital costs might scale.

Finally, if SAI operations are co-located adjacent to a sizable sour gas or refinery site, the cost to source H_2S for SAI would be zero or even slightly negative, but impurities—mostly water and CO_2—could be substantial, so an additional purification step would probably be required. Here we assume negative $50/tonne-S and apply a 10% penalty to lofting cost for impurities. Because of H_2S conversion costs, commercial S_8 produced from H_2S is often sold at a loss. Large gas operations exist at latitudes suitable for a tropical SAI operation (e.g., Jerun 3N, Natuna 4N, Lake Maracaibo 9N, Cabo Delgado 12S, to name a few). Assumed material costs are summarized in Table 4.

Lofting Cost. Lofting costs are calculated for each sulfur compound based on purpose-built, fixed-wing aircraft deployment in the tropics at 20 km altitude (using existing aircraft engine technology like that of SAIL-01[6,41]). The aircraft's useful payload is assumed to be 15 tonnes (20 tonnes total carrying capacity minus 5 tonnes of fixed fuel for ascent-descent and emergency diversion). The useful payload is then further divided among variable fuel, tank, and insulation mass; injection/combustion equipment; and sulfate precursor material. We assume an additional 5% of lofted material mass for the tank and structural supports, and 1−3% for insulation, depending on the transport temperature requirements for the different materials. (H_2SO_4 and S_8 must also be insulated to prevent freezing in the hold during ascent and cruise.) Pump and power are negligible for the injection rates (and maximum injection droplet size) listed in Table 4. Combustor mass for in situ sulfur combustion is estimated based on the method of Smith et al.[3]

Slower injection rates lead to longer cruises, which require more fuel and reduce the sulfur payload and the number of sorties one aircraft can complete per day. See Supporting Information Section S3.2 for cruise time calculation details. The total amount of sulfur lofted by one plane in one year is calculated based on ascent and descent of 30 min each, a ground refuel/reload/preflight interval of 80 min between sorties, 16 min to taxi out and 5 min to taxi in, and 4 h per day of both scheduled and unscheduled maintenance.

Cruise time is an additional factor in the above and is variable based on material injection rates. For example, a 30 min cruise at 20 km would result in a total flight time of 90 min, block time (total time in motion) of 111 min, and total time per cycle (including ground time) of 191 min. A 30 min cruise would enable 6 cycles to be accomplished in a 20-h operational day, whereas either shorter or longer cruise legs would lead to different cycle capacities per day. Aircraft operate 330 days per year at an assumed total cost of $35 M per year per plane (given these operating assumptions). The 100 ktonne-S lofting cost is representative of the fleet size and lofting rate for that fleet (Table 4). See Supporting Information Section S3 for full calculation details.

DISCUSSION

For the slow-acting gaseous precursors, H_2S is almost twice as cost-effective to loft as SO_2 and only marginally less safe. Both are very hazardous compared to the other sulfur compounds. H_2S should form aerosols in a similar size distribution as SO_2, and although H_2S production costs are uncertain at scale, we see no compelling reason to favor SO_2 over H_2S for SAI if H_2S can be produced below $1100/tonne-S. Use of H_2S will reduce the number of flights by a factor of ~2 compared to the use of SO_2, so it would halve the non-SAI environmental impacts of aircraft operations. Halving the number of required flights means that the overall risk of using H_2S may be smaller than the risk of SO_2 even though the H_2S risk is larger for a single flight with equal payload. We note that several experts had the impression that H_2S was substantially riskier than SO_2, while differences in acute toxicity are small; there are important differences that need further examination.

The combined safety and weight-efficiency of S_8 are promising, though further engineering would be required to clarify the optimal method of combusting it at altitude. Combustion rates and associated equipment mass lead to longer cruise times and a reduced payload such that S_8 is probably more expensive to loft than H_2S per tonne-S. However, if light, reliable airborne combustors could be developed, the safety trade-off would clearly favor S_8 over H_2S even at higher lofting costs.

CS_2 is essentially ruled out because of its sulfuric acid formation pathway. Half of CS_2 oxidizes to OCS,[8] which persists for a long time in the stratosphere or is transported out before forming sulfuric acid aerosols.[27] Any resultant aerosols would be ultimately hard to track and attribute, making small-scale test deployments impossible to evaluate. Furthermore, it does not provide a substantial safety benefit over H_2S or SO_2.

Of the fast-acting options, SO_3 is more efficient than H_2SO_4 in terms of mass, and while no plume modeling of SO_3 injection has yet been conducted, it will act like H_2SO_4, given sufficiently fast hydration. SO_3 is only slightly more dangerous than H_2SO_4 but presents some stability issues during transport and loading that could be reduced if produced on site. The S-mass efficiency increase for SO_3 over H_2SO_4 leads to a 15% reduction in cost per tonne-S with only moderate safety and ease of handling downsides.

Injecting SO_3 or H_2SO_4 at 0.1 kg/s to produce optimum-sized aerosols[19] is ~14× as expensive per kg S or ~4× as expensive per watt of forcing than the high-end modeled injection rate of 4−4.8 kg/s. (See supplemental Figure S3, dependent on aircraft engine configuration and plume dilution rate. A larger aircraft engine than simulated in Yu et al.[19] could allow for higher injection rates and thus more competitive cost

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performance, given full evaporation under technological constraints.) However, minimizing the total sulfur lofted may be desirable at significantly higher cost to reduce ozone depletion,[20,42,43] stratospheric heating and associated circulation changes,[44] ecosystem impacts of photosynthetically active radiation changes or sulfur deposition,[45] sky whitening,[46] or air quality impacts on human health.[47] These cost-environmental impact trade-offs should be evaluated in future work.

H_2SO_4 is four times the cost of H_2S to loft per tonne-S at the maximum injection rate for each compound. H_2S would need to form persistent aerosols at around 1 μm (~0.25 W/m^2 MtS[17]) to be equal to H_2SO_4 from a pure dollars/forcing perspective (not kg/forcing or accounting for background aerosol size distribution steering). Additional plume and aerosol kinetic modeling is needed to make an apples-to-apples radiative determination across these precursor types. Laboratory chamber and real-world data at 20 km conditions are needed to validate simulations.

Fast-acting compounds, while possibly too expensive for a full-scale deployment at the slowest injection rates, could make up a portion of the overall program alongside H_2S or SO_2 to help steer the entire aerosol size distribution toward the optimum.[20] Future plume-in-grid modeling work could find an optimal mix of implementation strategies.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.est.5c15172.

Additional calculation details and sulfur trade study report (PDF)

■ AUTHOR INFORMATION

Corresponding Author

James Franke − *Department of the Geophysical Sciences, University of Chicago, Chicago, Illinois 60637, United States; Climate Systems Engineering Initiative, University of Chicago, Chicago, Illinois 60637, United States;* orcid.org/0000-0001-8598-750X; Email: jfranke@uchicago.edu

Authors

Wake Smith − *Yale School of the Environment, New Haven, Connecticut 06520, United States; Mossavar-Rahmani Center for Business & Government, Harvard Kennedy School, Cambridge, Massachusetts 02138, United States*

Angela Slavens − *UniverSUL Consulting, Westwood Hills, Kansas 66205, United States*

Frank N. Keutsch − *John A. Paulson School of Engineering and Applied Sciences, Department of Chemistry and Chemical Biology, and Department of Earth and Planetary Sciences, Harvard University, Cambridge, Massachusetts 02134, United States;* orcid.org/0000-0002-1442-6200

Fangqun Yu − *Atmospheric Sciences Research Center, State University of New York, Albany, New York 12226, United States;* orcid.org/0000-0001-8862-4835

David Keith − *Department of the Geophysical Sciences, University of Chicago, Chicago, Illinois 60637, United States; Climate Systems Engineering Initiative, University of Chicago, Chicago, Illinois 60637, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.est.5c15172

Notes

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