Air quality impacts of ~~Stratospheric Aerosol Injections~~ stratospheric aerosol injections are likely small and mainly driven by changes in climate, not deposition

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Abstract. Stratospheric aerosol injection (SAI) is a proposed climate intervention ~~that could potentially reduce~~ method to offset future global warming through increased solar reflection in the stratosphere, but its broader environmental and public health implications are yet to be thoroughly explored. ~~Here, we~~ We use three large ensembles of fully coupled CESM2-WACCM6 simulations to assess changes in mortality attributable to fine particulate matter ($PM_{2.5}$) and ~~ozone~~ surface ozone exposure

5 (O_3) ~~using three large ensembles of fully coupled CESM2-WACCM6 simulations from the ARISE-SAI-1.5, ARISE-SAI-1.0 and SSP2-4.5 scenarios. In the ARISE-SAI-1.5 scenario, maintaining~~. Maintaining temperatures at 1.5~~degrees~~ °C above preindustrial levels through SAI ~~results in a modest~~ is projected to yield a modest 0.4% (ensemble range: -1.9% to +1.5%) reduction in pollution-related mortality ~~during 2060–2069 relative to SSP2-4.5, driven by a 1.26% decrease~~ relative to middle-of-the-road climate change scenario, reflecting a 1.3% (-2.3% to -0.6%) reduction in ozone-related deaths and a ~~0.86%~~ 0.9% (-0.4%

10 to +2.1%) increase in $PM_{2.5}$-related deaths. The spread among ensemble members underscores the influence of internal variability and highlights the importance of ensemble-based analyses when assessing the potential health impacts of climate intervention strategies. We find that global $PM_{2.5}$ mortality changes exhibit ~~almost no~~ little sensitivity to injected sulfate amounts, with the most variability driven by precipitation-mediated changes in non-sulfate $PM_{2.5}$ species (e.g., dust and secondary organic aerosols), whereas ozone-related mortality ~~are~~ is primarily driven by surface cooling and hemispheric

15 asymmetries in stratospheric-tropospheric exchange and ozone transport. ~~Overall, SAI~~ However, our results heavily reflect the specific forcing patterns of the SAI scenarios used; our estimates are also limited by model shortcomings, including omitting the effects of aerosols in the photolysis scheme - which might limit UV-driven changes and impact surface ozone rates - or not including nitrate aerosols. Within our framework, we find that SAI impacts on pollution-related mortality are modest ~~,~~ but regionally heterogeneous, and ~~much smaller in magnitude compared to~~ that the magnitude of the SAI-driven changes is smaller

20 than the improvements expected from near-term air quality policies ~~. Our finding that mortality impacts do not directly scale with SO₂ injection rates underscores the nonlinear and complex nature of atmospheric responses to SAI. Significant differences across ensemble members further emphasize the role of internal variability and the need for ensemble-based analysis when~~

~~evaluating potential health implications of climate intervention strategies.~~ planned or implemented within the same time frame.

25 1 Introduction

Stratospheric aerosol injection (SAI) is a proposed climate intervention strategy ~~to ameliorate~~ that could help ameliorate the effects of anthropogenic global warming. It involves ~~releasing reflective aerosols–usually sulfates–into the stratosphere~~ the release of precursors such as sulfur dioxide (SO_2), which would oxidize and create sulfate aerosols, into the stratosphere, in order to increase Earth's albedo and lower surface temperatures. This approach draws on the observed cooling effects of large

30 explosive volcanic eruptions (McCormick et al., 1995; Robock, 2000) and has been shown in climate model simulations to reduce global mean surface temperatures ~~relatively effectively~~ relative to scenarios without such intervention (Tilmes et al., 2018; Kravitz et al., 2015). However, despite its potential to offset some of the warming caused by greenhouse gases, SAI raises numerous questions ~~regarding~~ about its broader environmental, societal, and health-related consequences. One key concern is the impact of SAI on public health and air quality (Tracy et al., 2022). In terms of air quality, the main drivers of changes would

35 include the direct impacts of sulfate particles on surface fine particulate matter ($PM_{2.5}$), and changes in surface ozone exposure (O_3); the latter would be a function of changes in stratosphere-to-troposphere O_3 transport and in-situ changes in tropospheric ozone chemistry driven by the SAI-induced changes in temperatures and photolysis.

This study aims to assess the effects of SAI on air pollution mortality, particularly through changes in surface $PM_{2.5}$ and surface ozone (O_3), by using a fully-coupled modeling approach with the Community Earth System Model (CESM2) Whole

40 Atmosphere Climate-Chemistry Model (WACCM6)~~with comprehensive~~, which includes interactive aerosols and detailed representations of stratospheric and tropospheric chemistry. While using non-coupled model approaches allow one to better separate and quantify the contribution of single factors, a fully-coupled model allows for the simulation to include ~~their interaction~~ the interaction between aerosols, atmospheric composition and climate: what might be lost in precision in the diagnosis of changes can be gained in providing a more holistic picture of the overall expected change.

45 Previous studies have looked into the health impacts of SAI due to air quality changes (Eastham et al., 2018; Visioni et al., 2020; Moch et al., 2023; Harding et al., 2024). These efforts have either relied on more idealized modeling frameworks and/or simplified mortality estimation methods. In particular, both Eastham et al. (2018) and Moch et al. (2023) used chemical transport models (CTMs) to quantify global mortality effects from SAI, including ~~contributes~~ contributions from changes in air quality and UV-B exposure. While CTMs like GEOS-Chem have been widely applied to study air-pollution-related health

50 outcomes (Norman et al., 2025), they are fundamentally limited in capturing the dynamical and chemical feedbacks relevant to SAI. For example, in Eastham et al. (2018), the aerosol size distribution was prescribed offline assuming a fixed lognormal distribution centered at $0.16\ \mu$m. The use of a CTM also precludes accounting for interactive changes in stratosphere–troposphere exchange (STE), temperature-dependent tropospheric chemistry, and large-scale circulation responses to SAI. As a result, such models tend to predict spatially uniform decreases in stratospheric ozone and, consequently, reductions in tropospheric ozone

55 via STE, without accounting for compensating changes in transport or chemistry.

Harding et al. (2024) further used similar estimates as Eastham et al. (2018) and compared them against estimates of SAI impact on temperature-attributable mortality in the GFDL/FLOR model, in which the radiative forcing from geoengineering was simulated by reducing the solar constant. While solar dimming ~~provide~~ provides a simplified means of approximating the cooling effects of geoengineering, ~~they do~~ such approaches would not account for the spectrally dependent scattering and absorption properties of stratospheric aerosols, nor ~~do they~~ would it adequately capture the associated chemical and dynamical feedbacks, particularly those influencing ozone and STE (Visioni et al., 2021; Bednarz et al., 2022).

Finally, Xia et al. (2017) examined the impacts of SAI on tropospheric ozone through the use of a low-top version of CESM2, simulating SAI itself through prescribing an aerosol distribution (therefore with no changes in stratospheric aerosols settling and deposition) or through a solar constant reduction; they found that surface ozone generally decreases as a consequence of SAI, with some significant differences between solar dimming and SAI driven by changes in stratospheric ozone and STE, but did not quantify the ~~the~~ resulting health implications of changes in surface ozone on human exposure.

In this study, we use simulations from the Assessing Responses and Impacts of Solar intervention on the Earth system with Stratospheric Aerosol Injection (ARISE-SAI) experiment using CESM2-WACCM6 (Davis et al., 2023; Gettelman et al., 2019) , which simulates SAI with injections at four discrete latitudinal points (15°S, 15°N, 30°S and 30°N) ~~in order to maintain large scale~~ to maintain global mean surface temperatures at the 1.5°C (ARISE-SAI-1.5) or 1.0°C (ARISE-SAI-1.0) above preindustrial levels ~~(Davis et al., 2023)~~(Richter et al., 2022). This model includes ~~comprehensive atmospheric chemistry and~~ interactive aerosol processes, ~~allowing for a more detailed assessment of~~ whose evolution is simulated through the use of a modal approach (Liu et al., 2016), and a detailed representations of tropospheric and stratospheric chemistry (Emmons et al., 2020; Tilmes e , allowing us to assess how SAI influences air pollution and associated health risks ~~, as well as interactively simulating the coupling between SAI-induced~~ through coupled changes in atmospheric temperatures, transport, and chemistry. Compared to previous studies, our approach provides a more realistic representation of injection strategies and chemistry-climate interactions, improving estimates of pollution-driven mortality. While this provides an important advance beyond earlier studies, some limitations remain: for example, in CESM2(WACCM6) photolysis rates are calculated using lookup tables, taking into account the overhead ozone column and clouds but excluding the effects of aerosols, thereby reducing the effect SAI aerosols could have on tropospheric photochemistry and ozone. Our results should therefore be viewed as a further step toward understanding these interactions, with important knowledge gaps that future studies will need to address.

Another key contribution of this study is the explicit quantification of model internal variability in estimates of air pollution and associated health impacts. Modeled air pollutant concentrations are sensitive to changes in climate and dynamics which in turn are affected by model internal variability. This could be especially important when the changes in surface air pollution arise from climate system adjustments due to SAI rather than from changes in surface emissions. While this source of uncertainty is often underexplored in the literature (e.g., it cannot be easily assessed based on CTM results), our use of a 10-member ensemble of coupled simulations allows us to highlight its substantial influence on $PM_{2.5}$ ~~annd~~ and ozone concentrations, and the associated mortality outcomes. In the following sections, we evaluate the effects of SAI on surface air quality and associated health outcomes by analyzing changes in $PM_{2.5}$ and ozone exposure, estimating attributable mortality using epidemiological risk functions, and characterizing the spatial and ensemble variability in these impacts on global and regional scales.

2 Methods

2.1 ~~Simulations~~Model description

Simulations were conducted using the Community Earth System Model, version 2 with the WACCM, version 6 (CESM2(WACCM6); Gettelman et al. (2019); Davis et al. (2023)), a fully coupled ocean-atmosphere model with interactive tropospheric and stratospheric chemistry and aerosols. The model simulates aerosol formation and growth through an interactive, two-moment modal aerosol microphysics scheme (MAM4; Liu et al. (2016)), allowing sulfate aerosols to evolve over time based on the simulation of nucleation, coagulation, condensation and removal processes. However, ~~it is to be noted that~~ MAM4 uses assumptions of internal mixing for the size distribution of different species~~(,~~ whereas mass is tracked separately ~~)~~ (Visioni et al., 2022). While stratospheric and tropospheric chemistry are fully interactive, ~~tropospheric~~ photolysis rates are ~~prescribed and held constant at year-2000 levels (Kinnison et al., 2007). Sulfate~~ calculated using lookup tables, taking into account the overhead ozone column and clouds but excluding the effects of aerosols (Kinnison et al., 2007), thus excluding the direct effect of the aerosols on actinic fluxes (Michelangeli et al., 1992; Palancar et al., 2013). Simulated $PM_{2.5}$ components include sulfate (SO_4), secondary organic aerosols (SOA), primary organic matter (POM), salt, dust and black carbon (BC)~~are simulated and considered for calculating PM~~. However, the model does not include explicit ammonium or nitrate aerosol chemistry, which are standard in some regional air quality models (e.g., CMAQ) and can contribute significantly to PM2.5 in ammonia-rich regions (Nolte et al., 2018; Hancock et al., 2023). This omission may lead to an underestimate of absolute $PM_{2.5}$ ~~.~~

~~The~~ concentrations and associated health impacts in certain areas, but should not impact heavily our comparison of future trends when comparing SAI and non-SAI scenarios: while interactions between the formation of nitrate and sulfate aerosols are complex (Liu et al., 2020), recent observations have shown (Wen et al., 2023; Wei and Tahrin, 2024) that it is the absence of sulfate aerosols that favors fine particulate nitrate formation in some environment. However, our analysis emphasizes the differences between ARISE-SAI-1.5 and SSP2-4.5 ~~ensemble represents a control climate scenario following the~~ scenarios and so any systematic bias is applied consistently and is unlikely to materially affect our conclusions about the relative health impacts of SAI.

These limitations notwithstanding, the interactive chemistry–climate framework of WACCM allows us to capture coupled meteorological, chemical, and radiative feedbacks that are central to evaluating the air quality response to stratospheric aerosol injection (Tilmes et al., 2019). CESM2(WACCM6) has been evaluated against earlier model versions and observations–including NASA ATom aircraft profiles (Tilmes et al., 2019), Tropospheric Ozone Assessment Report (TOAR) surface ozone data (Emmons et al., 20...), and Measurements of Pollution in The Troposphere (MOPITT) carbon monoxide observations (Schwantes et al., 2020) –showing good agreement with ozonesonde data and seasonality of surface ozone, though with some regional spatial biases. Previous evaluations have also shown that WACCM reproduces the large-scale distributions of tropospheric ozone and key pollutants, as well as climatological patterns of aerosols, with skill comparable to other climate models (Griffiths et al., 2021; Hancock et al. ... These assessments further support the suitability of this model for investigating the relative changes in air quality under SAI.

2.2 Simulations

125 The baseline ensemble (i.e. without SAI) follows the Shared Socioeconomic Pathway 2 with ~~moderate~~ middle-of-the-road increases in greenhouse gas emissions, leading to a radiative forcing of 4.5 W/m^2 by 2100 ~~Fricko et al. (2017). The~~ (Fricko et al., 2017), and is hereafter referred to as SSP2-4.5. In the ARISE-SAI-1.5 ~~experiment (Richter et al., 2022) simulates the deployment of stratospheric aerosol injection (SAI) to limit global warming to approximately 1.5°C above preindustrial levels (Richter et al., 2022). In ARISE-SAI-1.5,~~ ensemble, under the same emission scenario, sulfur dioxide (SO$_2$) is injected annually at four fixed

130 latitudes (15°N, 15°S, 30°N, 30°S) at approximately 21.5 km of altitude starting in year 2035 ~~of SSP2-4.5~~, and run until 2070, with injection rates adjusted at the beginning of each year to offset continuing warming under the SSP2-4.5 emissions pathway ~~and~~, with the aim of maintain global mean surface temperatures and their large-scale gradients at the 1.5°C above preindustrial ~~levels~~ level (defined as the mean over 2020-2039 to ensure better consistency with other climate models, (Visioni et al., 2024)). The ARISE-SAI-1.0 simulations follow the same protocol, but SAI is used to cool by a further 0.5°C compared to the targets

135 in ARISE-SAI-1.5.

A 10-member ensemble is ~~used~~ produced for all three ~~ensembles~~ cases to account for internal climatic variability (Richter et al., 2022). For regional assessments of mortality and mortality-related factors, we will focus our analyses on the ARISE-SAI-1.5 case, whereas results from the ARISE-SAI-1.0 will be provided for global, temporal and injection-related analyses in order to highlight the ~~linerarity~~ linearity (or lack thereof) of the SAI response with the injection rates.

140

2.3 Calculation of exposure and mortality

~~We calculate~~

Here we describe how we calculated the impact on mortality rates attributed to changes in the simulated changes in ambient surface PM$_{2.5}$ and O$_3$ ~~calculations~~. All mortality estimates in future scenarios are calculated using the fixed 2020 population

145 distribution. ~~This~~: this approach isolates the effects of air quality changes by removing confounding influences from projected population growth or redistribution.

Mortality is estimated ~~as follows~~ using the health impact function (EPA, 2015):

$$M_{i,d,a,t} = \text{BMR}_{d,a,t} \times \text{P}_{i,a,2020} \times AF_{i,d,a,t} \qquad (1)$$

Where $M_{i,t}$ is the mortality for CESM grid i from disease d for age group a and year t; P is the number of population in

150 2020 with each age group a in grid i; BMR is the national base mortality rate for disease d, age group a and year t; AF is the attributable fraction which estimates the proportion of deaths in a population that can be attributed to a specific exposure to disease d or risk factor from epidemiological studies. For PM$_{2.5}$, we use the AF associated with noncommunicable diseases and lower respiratory infections (NCD+LRI). For ozone, we use the AF associated with cardiovascular and respiratory diseases. Whereas previous studies (Eastham et al., 2018) attributed PM$_{2.5}$ exposure to cardiovascular and respiratory diseases and ozone

155 exposure solely to respiratory diseases, here we attribute cardiovascular disease to ozone exposure ~~in this study~~, which aligns

with more recent epidemiological findings (Sun et al., 2024; Niu et al., 2022) and improves the completeness of ozone-related health impact assessments.

For PM$_{2.5}$, AF is calculated using the exposure–response function from the Global Exposure Mortality Model (GEMM; Burnett et al. (2018)), which provides improved estimates across a wide range of ambient PM$_{2.5}$ concentrations. GEMM is particularly effective in low-income and high-pollution regions where the older Integrated Exposure–Response (IER) functions tend to underperform due to limited observational data and less robust extrapolation at high exposure levels (Burnett et al., 2014, 2018; Burnett and Cohen, 2020):

$$AF_{i,d,a,t} = 1 - \frac{1}{RR_{i,d,a,t}}; \text{ where } RR_{i,d,a,t} = \exp^{\frac{\theta \times log(\frac{C_{i,t}}{\alpha+1})}{1+\exp^{-\frac{C_{i,t}-\mu}{v}}}} \text{ and } RR_{i,d,a,t} = 1 \text{ when } C_{i,t} < 2.4 \ \mu g/m^3 \tag{2}$$

Where C is the ambient PM$_{2.5}$ concentration; ($\mu g/m^3$); RR is the relative risk of morality at any concentration; θ, α, μ and v are ~~parameters specified~~ empirical coefficients from the GEMM which are specific for each age group.

For ozone-attributable mortality, we convert surface ozone to the ozone season maximum daily 8-hour average (OSMDA8; ppb) using hourly surface O$_3$ data for each experiment and each ensemble member. OSMDA8 calculates the highest 6-month rolling average daily 8-hour average ozone concentration ~~during the ozone season, which typically spans from spring through summer when ozone levels are at their peak~~, which reflects the highest average ozone concentration over a 6-month period. OSMDA8 is the metric used by the Global Burden of Disease (GBD) (Brauer et al., 2024) for quantifying the health effect from long-term ozone exposure and is used in the World Heath Organization's air quality guidelines (WHO, 2021, License: CCBY-NC-SA3.0IGO). To calculate the ozone-attributable risk fraction, we calculate the ~~AF~~ AF for cardiovascular and respiratory disease separately and then combine the associated mortality.

$$AF_{i,d,a,t} = 1 - \exp^{-\beta(X_{i,t}-X_{min})}; \text{ where } AF = 0 \text{ when } X_{i,t} < X_{min} \tag{3}$$

Where X represents the spatially and temporally resolved grid-cell level OSMDA8; X$_{min}$ represents the theoretical minimum risk exposure concentration and β represents a model-parameterized slope of the log-linear relationship between concentration and health from epidemiological studies. For chronic respiratory disease mortality, we apply a β of ln(1.06) per 10 ppb ozone (95% ~~CI~~ confidence interval (CI) 1.03-1.10) derived by GBD 2019 (Jerrett et al., 2009; Malashock et al., 2022; Murray et al., 2020). For cardiovascular disease mortality, we apply a β of ln(1.028) per 10 ppb ozone (95% CI 1.010-1.047) (Sun et al., 2024). A summary of the RR and disease d used to calculate mortality associated with PM$_{2.5}$ and O$_3$ is provided in Table 1.

Cause	Disease (d)	Minimum exposure concentration	Source
PM$_{2.5}$	Noncommunicable diseases & lower respiratory infections (NCD+LRI)	2.4 $\mu g/m^3$	Burnett et al. (2018)
Ozone	Cardiovascular diseases	40 ppb	Sun et al. (2024)
	Respiratory diseases	32.4 ppb	Malashock et al. (2022)

Table 1. Summary of risk functions used for estimating attributable mortality. Minimum exposure concentrations correspond to the theoretical minimum risk exposure levels for each pollutant-health outcome pair.

Our BMRs are drawn from the International Futures (IFs) health model, providing dynamic, age and disease-specific mortality projections consistent with policy interventions following the SSP2-4.5 scenario (Hughes et al., 2014). The IFs health model is a comprehensive, integrated modeling platform used to explore long-term global health dynamics. This represents a more realistic approach compared to the use of static BMRs in previous studies (Eastham et al., 2018).

Population (P) for each age group was calculated by using the global population density dataset based on Shared Socioeconomic Pathways (SSP) (Jones and O'Neill, 2020) and the ratio of the population for each age group to the total population retrieved from the SSP database developed by the International Institute for Applied Systems Analysis and the National Center for Atmospheric Research (Riahi et al., 2017; Samir and Lutz, 2017) for each country. The raster of nation-states was retrieved from the Gridded Population of the World, Version 4 (GPWv4): National Identifier Grid (Center for International Earth Science Information Network (CIESIN) - Columbia University, 2018) and is used to aggregate the calculated mortality to country-level mortality estimates. We further categorize the world into 21 regions following the GBD Study based on epidemiological similarities and geographic proximity.

Other studies estimating air pollution-related mortality have typically calculated mortality uncertainty based on the central intervals of the parameters used in the AF calculations (Peng et al., 2021; Eastham et al., 2018). However, less attention has been given to the uncertainty arising from internal model variability: this is important as internal variability can drive regional air quality differences (Fiore et al., 2015). Thus, our analysis account for uncertainty arising from climate ensemble spread, while applying central estimates for β (for ozone) and RR (for PM$_{2.5}$).

3 Results

3.1 Changes in health-related air pollutants

In Fig. 1, and in the subsequent mortality analysis, we present changes in surface PM$_{2.5}$, ozone, temperature (T_s), and total precipitation in three ways: (1) the 2060–2069 average from the ARISE-SAI-1.5 simulation minus 2030-2039 average from SSP2-4.5, illustrating the change under SAI implementation; (2) the 2060–2069 average from SSP2-4.5 minus 2030-2039 average from SSP2-4.5, representing changes under the SSP2-4.5 pathway without SAI; and (3) the difference between the 2060–2069 averages of the ARISE-SAI-1.5 simulation and SSP2-4.5, showing the direct impact of SAI by comparing a future with SAI to one without it. Particularly when looking at air quality impact, this three-way comparison is of particular relevance as we generally expect a reduction in surface pollutants in future scenarios independently of SAI implementation (Fricko et al., 2017; Hussain, 2025; Nazarenko et al., 2022), therefore a comparison just between the present day and future SAI scenario will almost always indicate improved air quality. Therefore, comparing also the same future periods (which have the same surface emissions) with and without SAI helps to isolate the direct SAI contribution to air quality.

Consistent with previous studies (Visioni et al., 2023), ARISE-SAI-1.5 exhibits an overall reduction in precipitation relative to the increase observed in SSP2-4.5 (Fig. 1f): these changes are due to both
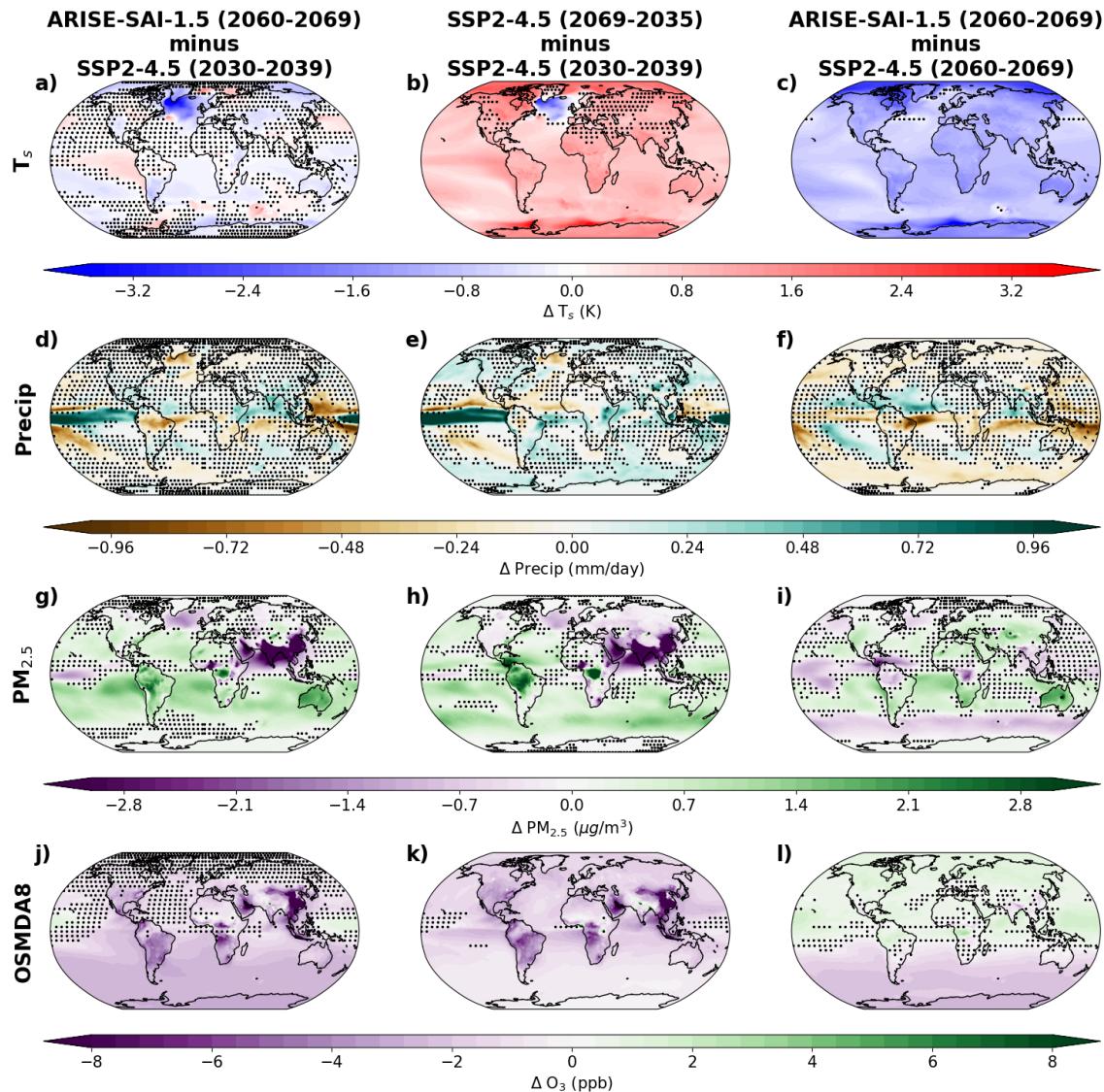
Figure 1. Spatial patterns of the changes in surface climate variables (surface temperature and precipitation)~~and~~, air quality ($PM_{2.5}$, and ozone exposure (OSMDA8) concentration) under the SAI scenario ~~,~~ (ARISE-SAI-1.5~~,~~) and the baseline scenario (SSP2-4.5) for the period 2060-2069 compared to 2030-2039. Each row represents changes in: (a-c) surface temperature (T_s, K), (d-f) precipitation (mm/day), (g-i) $PM_{2.5}$ concentration ($\mu g/m^3$), and (j-1) OSMDA8 (ppb). The stippling indicates areas where differences between ARISE-SAI-1.5 and SSP2-4.5 are not statistically significant ($p > 0.05$) based on a t-test performed across all 10 ensemble members. Columns indicate the difference between the SAI case and the reference period with same global temperatures (left), the difference between a warmer future and the reference period (center), and the difference between the SAI case and a warmed future following the same underlying emission scenario (right).

the avoidance of the temperature-related Clausius-Clapeyron increase expected under climate change, as well as to changes in the strength and position of the Intertropical Convergence Zone (ITCZ) and the Hadley circulation (Kravitz et al., 2017; Lee et al., 2020; Richter et al., 2022; Cheng et al., 2022) in the different scenarios. While some regions do not exhibit statistically significant changes in surface $PM_{2.5}$ relative to SSP2-4.5 (2030–2039), other areas - such areas–such as Central America and central Sub-Saharan Africa - do show Africa–show significant reductions. In these regions, $PM_{2.5}$ decreases coincide with increases in precipitation (Fig. 1d–f), suggesting that enhanced wet scavenging may play a role. However, the overall spatial pattern of $PM_{2.5}$ changes does not consistently align with precipitation trends (Fig. 1g, h and j), indicating that other processes, such as changes in circulation, vertical mixing, or aerosol-cloud interactions, may also contribute to changes in $PM_{2.5}$. Thus, while precipitation influences $PM_{2.5}$ in some regions, it does not fully explain the simulated patterns or their statistical significance.

Furthermore, Fig. 2 indicates that dust and secondary organic aerosols (SOA), rather than sulfate (SO_4), are the dominant contributors to total $PM_{2.5}$ concentrations across most regions in ARISE-SAI-1.5. In SSP2-4.5 (not shown), the spatial distribution of the dominant $PM_{2.5}$ species is broadly similar, with SO_4 not emerging as the dominant species across most regions, unless particularly pristine (Visioni et al., 2020), like at high latitudes, or already extremely polluted. While it is true that sulfate can still drive relative changes in $PM_{2.5}$ even when not dominant in absolute terms, our subsequent analysis of mortality (Section 3.2) shows that the changes in $PM_{2.5}$ concentrations and $PM_{2.5}$-related mortality are not driven by sulfate. Specifically, the spatial and temporal patterns of $PM_{2.5}$-related mortality changes align more closely with changes in non-sulfate species and are shaped by precipitation and circulation-driven effects such as wet scavenging and regional aerosol transport.

Lastly, Fig. 1j-l shows percent changes in surface ozone exposure. Interpreting these changes requires accounting for multiple mechanisms, including SAI-induced impacts on stratospheric ozone and its transport to the surface, and changes in ozone in-situ photochemical processing driven by changes in temperature and photolysis. We note that our study does not include tropospheric ozone changes caused by changes in photolysis, as in the troposphere photolysis rates are fixed at the present-day levels. SAI influences stratospheric ozone through multiple pathways, including alterations in heterogeneous chemical reactions on aerosol surfaces, modifications in photolysis rates due to changes in stratospheric radiationactinic flux from changes in the overhead ozone column and aerosol absorption and scattering, and dynamical changes in stratospheric circulation and temperature patterns that can impact ozone transport and distribution (Tilmes et al., 2009, 2022; Bednarz et al., 2023a). Injection strategy also plays a key role: in ARISE-SAI-1.5, SO_2 is injected primarily in the Southern Hemisphere (SH) during 2060–2069 to modulate restore hemispheric temperature gradients affected by the asymmetric warming in the underlying SSP2-4.5 simulations, resulting in an asymmetric stratospheric aerosol burden and consequently an asymmetric ozone response (Richter et al., 2022; Bednarz et al., 2023b). However, as discussed before, our study does not include tropospheric chemistry changes caused by direct aerosol-driven changes in photolysis. As a result, our analysis does not capture potential tropospheric ozone responses caused by aerosol scattering (Visioni et al., 2017a). A study by Bardeen et al. (2021) using a previous version of WACCM (WACCM4), but modified to include online Tropospheric Ultraviolet and Visible (TUV) model calculations, showed that the exclusion of aerosol optical depth from the TUV calculations only resulted in a small difference in the overall
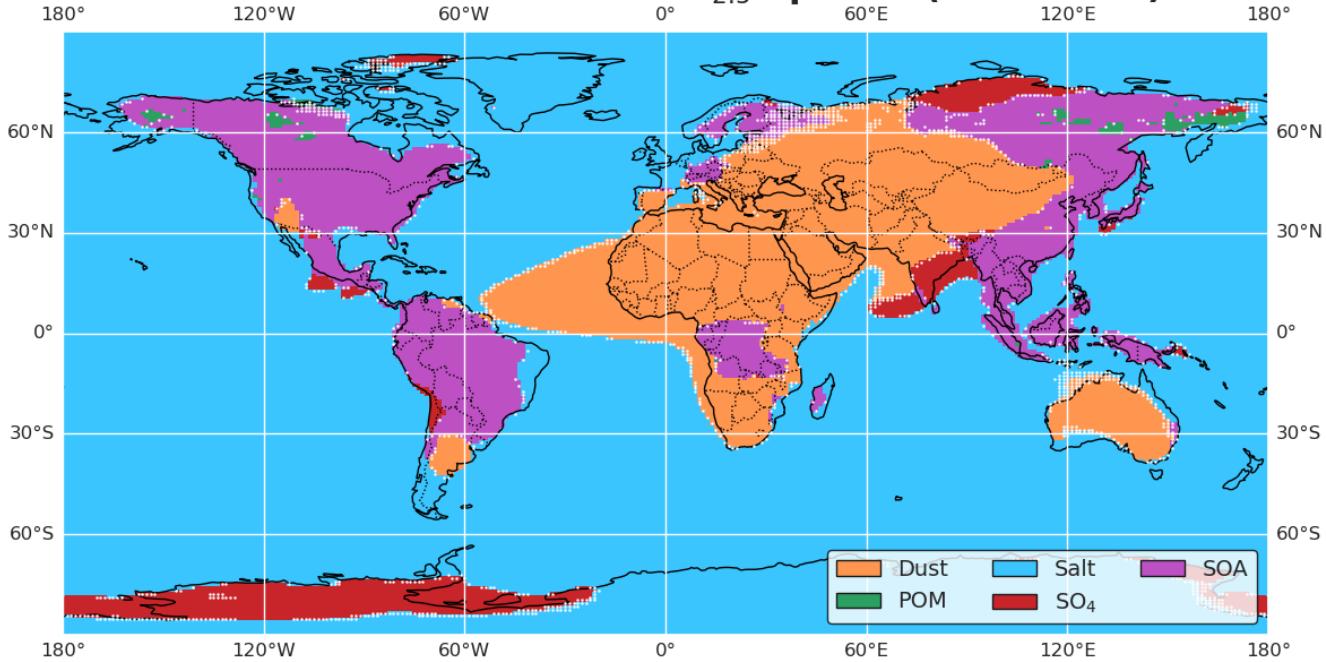
Figure 2. Map of the most prevalent PM$_{2.5}$ species (dust, primary organic matter (~~pom~~POM), salt, sulfate (SO$_4$), secondary organic aerosols (SOA) and black carbon (BC)) across grid cells, derived from ensemble model averages under the ARISE-SAI-1.5 scenario. Colors represent the dominant species at each location, determined by taking the fraction of the species to the total PM$_{2.5}$ concentration. ~~Note that black Black~~ carbon is not presented here because it does not dominate in any grid cell. White stippling ~~are~~ is over areas where fewer than 90% of ensemble members agree on the dominant species at a grid point.

ozone column changes due to minimal differences in the overall ozone loss rates, leading us to conclude that this shortcoming in our simulations is not likely to significantly impact our conclusions.

250   This stratospheric asymmetry propagates to the troposphere. Specifically, ozone concentrations decrease across much of the ~~Southern Hemisphere~~ SH troposphere, while increasing in the Northern Hemisphere (NH) (Fig. 11 ~~;~~ and Fig. 3f). These hemispheric differences arise from distinct underlying mechanisms. In the ~~Southern Hemisphere~~SH, the reduction in surface ozone is primarily driven by ~~the reductions in stratospheric ozone concentrations in the SH mid and high latitudes under~~ aerosol-driven catalytic ozone loss in the Antarctic stratosphere alongside any changes in polar vortex strength and large-scale

255 stratospheric transport (Bednarz et al., 2023b), and the resulting reduction in STE.

    In contrast, the ~~Northern Hemisphere~~ NH surface ozone increases are likely not driven by changes in STE. Although stratospheric ozone increases occur in the NH lower-to-mid stratosphere~~(Fig. A4)~~, this signal does not extend to the surface. Hence, the NH surface ozone changes likely reflect the SAI-induced changes in in-situ tropospheric chemical processing. In particular, ~~H2O levels decrease throughout~~ water vapor concentrations decrease in the troposphere in ARISE-SAI-1.5
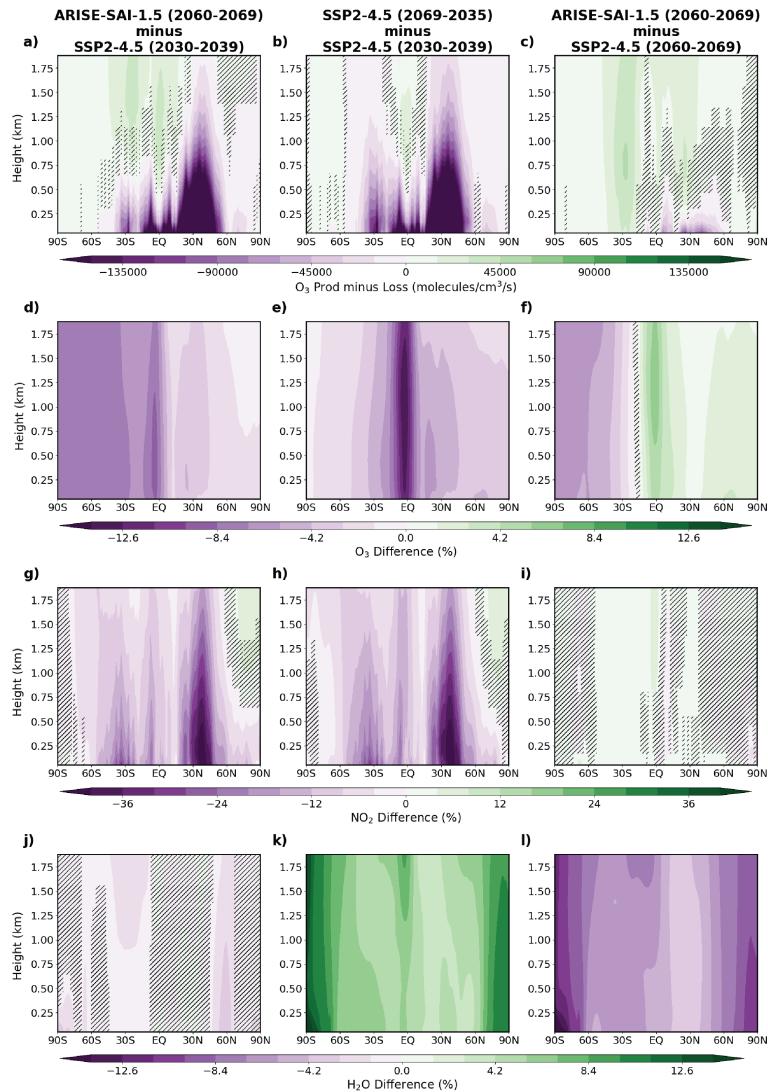
Figure 3. Zonal-mean percent changes in ozone chemical production minus loss rates, ozone concentrations, and NO_2 concentrations under ARISE-SAI-1.5 and SSP2-4.5 scenarios. Panels (a–c) show the difference in ozone production minus loss (molecules/cm^{-3}/s^{-1}): (a) ARISE-SAI-1.5 (2060–2069) minus SSP2-4.5 (2030–2039), (b) SSP2-4.5 (2060–2069) minus SSP2-4.5 (2030–2039), and (c) ARISE-SAI-1.5 (2060–2069) minus SSP2-4.5 (2060–2069). Panels (d–f) show the corresponding % differences in ozone concentrations (mol mol^{-1}) for the same scenario comparisons. Panels (g–i) show % differences in NO_2 concentrations(mol mol^{-1}), highlighting changes in a key ozone precursor and panels (j-l) show % differences in water vapor (H_2O) concentrations. In each panel, shading denotes the magnitude of the difference, and black contours indicate the absolute zonal-mean values from the first dataset of each comparison (e.g., The stippling indicates areas where differences between ARISE-SAI-1.5 or and SSP2-4.5 are not statistically significant (p>0.05) based on a t-test performed across all 10 ensemble members.

260 compared to SSP2-4.5 (Fig. 3l) as the result of large scale near-surface cooling (Fig. 1c). This reduces chemical ozone loss in the free-troposphere, as indicated by an increased net (i.e. production minus loss) photochemical ozone production (Fig. 3c). ~~Owing~~ Due to rapid tropospheric mixing timescales, the resulting NH ozone increases extend to the surface, even despite negative (particularly between 0 to 50°N) NH surface net production changes under SAI. The latter indicate suppressed in-situ photochemical ozone formation that occurs in a NO_x-rich region (~~see contours in~~ Fig. 3g ~~,h and Fig. ??~~and h) under decreased

265 OH ~~(Fig. A6)~~ and the resulting suppressed ~~RO2-NO2~~ RO_2-NO_2 cycling (despite a concurrent increase in NH surface NO_x which should otherwise enhance ozone production, Fig. 3i), consistent with previous work demonstrating that reductions in temperature and humidity can suppress photochemical ozone formation in NO_x-rich environments ~~Archibald et al. (2020); Rasmussen et al.~~ (Archibald et al., 2020; Rasmussen et al., 2013; Doherty et al., 2013; Zanis et al., 2022).

To further test this interpretation, we repeated our analyses in simulations with simulated SAI injections but no changes in

270 tropospheric anthropogenic emissions (i.e. in a preindustrial climate) and observed qualitatively similar ozone responses (not shown), reinforcing our finding that ~~the changes arises~~ these changes arise from stratospheric chemistry, transport, and in-situ oxidant perturbations, consistent with previous findings on SAI-driven ozone redistribution (e.g., Xia et al., 2017; Niemeier and Schmidt, 2017; Tilmes et al., 2009).

3.2 Calculation of the air pollution related mortality from PM$_{2.5}$ and ozone changes

275 This section presents the estimated mortality impacts of SAI under the ARISE-SAI-1.5 protocol, relative to SSP2-4.5. We ~~include the 35-year ARISE-SAI-1.0 simulation in Fig. 7 and 8.~~

~~We~~ first examine changes in PM$_{2.5}$-related mortality resulting from SAI, followed by an assessment of ozone-related mortality. Together with showing ensemble-averaged results, we also highlight in the following maps the large inter-ensemble and inter-ensemble spread when calculating mortality based on yearly model output. Local air quality is strongly dependent

280 on meteorological conditions (Liu et al., 2022; Jacob and Winner, 2009; Xu et al., 2020) such as precipitation rates, heatwaves and atmospheric ~~inversion~~inversions. Global warming itself has been postulated to strengthen many of ~~this~~ these conditions as well (Jacob and Winner, 2009). Therefore, ~~we deem important to place our obtained average estimates in this~~ it is important to interpret our estimates within this broader context.

Fig. 4 and 7a show the annual global deaths resulting from changes in PM$_{2.5}$ concentration and the average PM$_{2.5}$-related

285 deaths by country, respectively. We estimate that SAI leads to a reduction of ~151,~~750~~ 000 premature deaths from PM$_{2.5}$ under ARISE-SAI-1.5 (2060-2069), relative to SSP2-4.5 (2030-2039), with ensemble member estimates ranging from -140,~~245 to 000 to~~
-164,~~068.~~ 000. In comparison, SSP2-4.5 (2060-2069) results in a reduction of ~165,~~669~~ 000 premature deaths relative to 2030-2039 levels, with a range of -148,~~397~~ 000 to -177,~~296.~~ 000. This yields a net increase of ~~13,918~~ ~14,000 premature

290 deaths in ARISE-SAI-1.5 compared to SSP2-4.5 during 2060-2069, with an ensemble range of -7,~~468~~ 000 to +21,~~177. These estimatesand~~ 000. These estimates, along with the standard deviation shown in Fig. 4~~show that the standard deviation of mortality estimates highlights the large spread~~, illustrate the substantial variability in projected PM$_{2.5}$-related deaths.
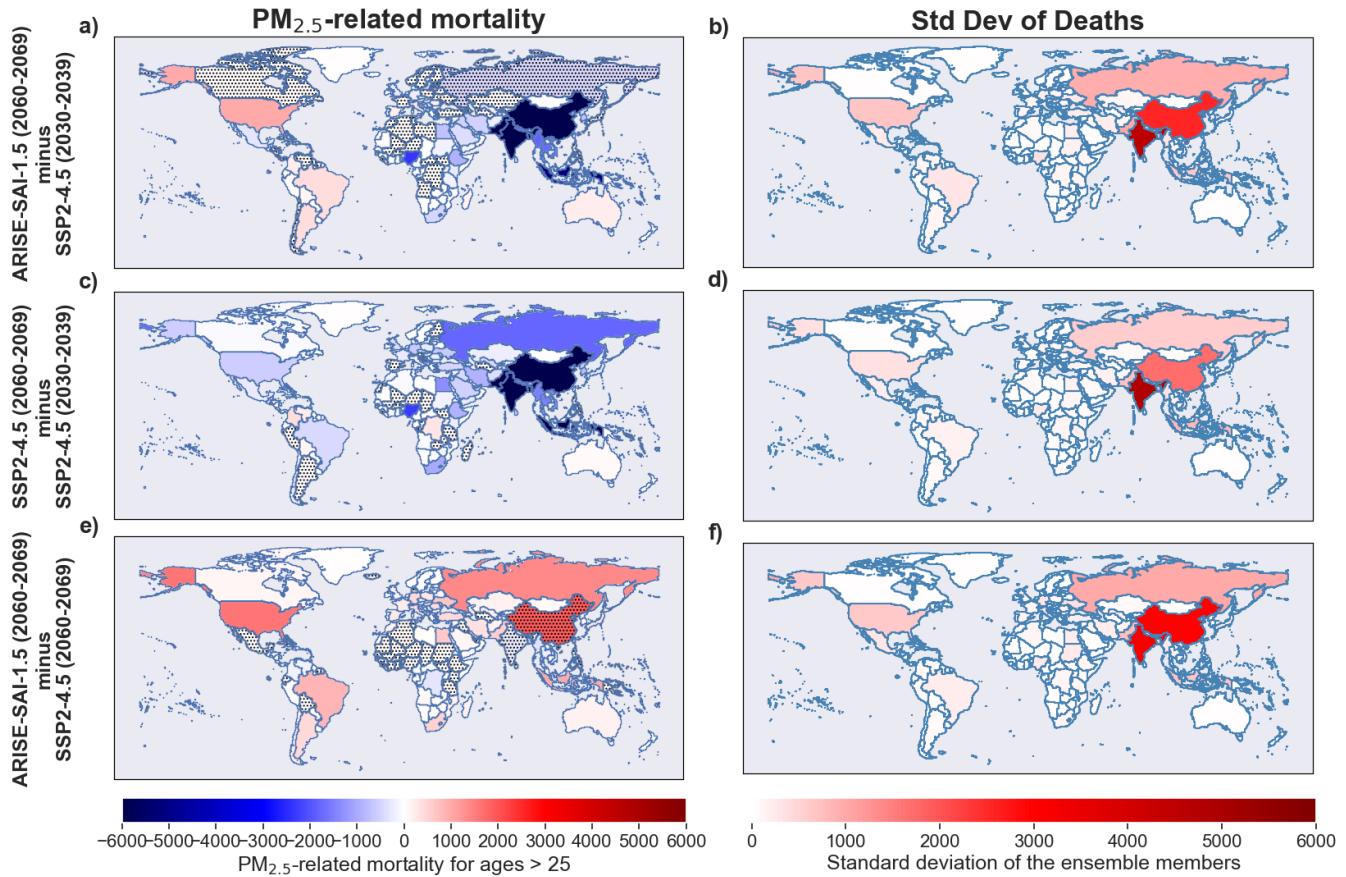
Figure 4. The ensemble-mean deaths from PM$_{2.5}$ for ages > 25. Stippling indicates countries where ~~changes~~ estimates of the PM$_{2.5}$-related mortality are not statistically significant ~~(p>0.05) based on a two-sided t-test~~ across ensemble members at the 95% confidence level.

The changes in PM$_{2.5}$-related mortality for each country in Fig. 4e are roughly consistent with the geographical changes in PM$_{2.5}$ shown in Fig. 1i. In Fig. 5a, we compute the ensemble-averaged global deaths resulting from SSP2-4.5 with added changes in individual PM$_{2.5}$ components between ARISE-SAI-1.5 and SSP2-4.5 to isolate the influence of each component on global mortality. Among the components, incorporating changes in the dust PM$_{2.5}$ produce the largest deviation from the unmodified SSP2-4.5 baseline. ~~Notably, the~~ The scenario with dust-only modifications results in fewer global deaths than the SSP2-4.5 baseline, which is likely due to the nonlinearity in the ozone-attributable risk function ~~(see Fig. ??b).~~ However, when changes in all PM$_{2.5}$ components are combined, the resulting mortality aligns with the increased PM2.5-related mortality observed in ARISE-SAI-1.5. For other components such as salt, BC, POM, SOA and SO$_4$, the resulting mortality estimates largely overlap the unmodified SSP2-4.5 baseline. In particular, the changes in global deaths attributable to SO$_4$ are ~~relatively small~~ small relative to other components, implying that sulfate-driven PM$_{2.5}$ mortality changes are modest compared to the total. Therefore, we conclude that SAI's contribution to PM$_{2.5}$–related mortality ~~appears small,~~ is small compared to the overall

changes projected due to future air quality policies (on the order of ∼1%, versus ∼10% from policy-driven improvements),

305   with internal variability among ensemble members and changes from other PM$_{2.5}$–related species ~~, potentially driven by precipitation changes,~~ playing a dominant role in driving uncertainty in our mortality estimates.
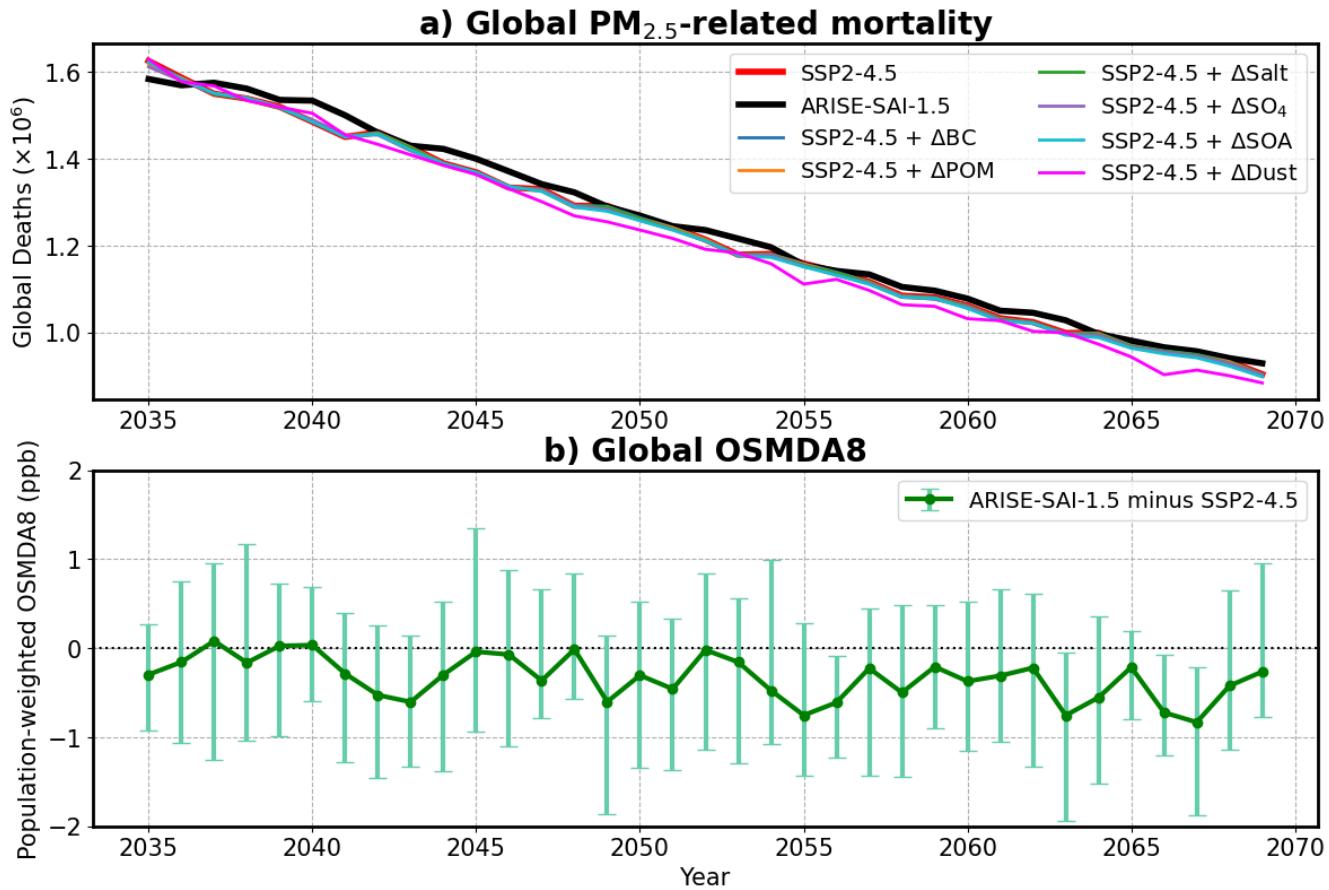
Figure 5. (a) Ensemble-averaged global PM$_{2.5}$-related mortality over time under the SSP2-4.5 and ARISE-SAI-1.5 scenarios, along with sensitivity simulations where changes in individual PM$_{2.5}$ components (ΔBC, ΔPOM, ΔSalt, ΔSO$_4$, ΔSOA, ΔDust) between ARISE-SAI-1.5 and SSP2-4.5 are added to the SSP2-4.5 baseline. (b) Time series of population-weighted global OSMDA8 (daily maximum 8-hour ozone) differences between ARISE-SAI-1.5 and SSP2-4.5, with error bars indicating ensemble spread~~(± 1 standard deviation)~~.

    For ozone-related mortality, Fig. ~~7b and 6~~ 6 and 7b show the annual global total deaths resulting from changes in ozone concentration and the average ozone-related deaths by country, respectively. We estimate that SAI leads to a reduction of ~~101,706~~ ∼102,000 premature deaths from ozone exposure under ARISE-SAI-1.5 (2060–2069), relative to SSP2-4.5 (2030–2039),

310   with an ensemble range of -91,~~413~~ 000 to -108,~~493.~~ 000. By comparison, SSP2-4.5 (2060-2069) results in an estimated reduction of ~~87,819~~ ∼89,000 premature deaths from ozone exposure relative to SSP2-4.5 for 2030–2039, with a range of ~~-76,583 to -96,917.~~ -77,000 to -97,000. The net difference between ARISE-SAI-1.5 and SSP2-4.5 during 2060-2069 is ∼-

~~13,887~~14,000, with a range of ~~-6,610 to -24,852. Fig. 6 shows that the standard deviation of the ozone-related mortality is smaller than that of PM$_{2.5}$-related mortality globally, indicating lower ensemble variability.~~ -7,000 to -25,000.

315 ~~The ensemble-mean deaths from ozone for ages > 25. Stippling indicates countries where changes are not statistically significant (p>0.05) based on a two-sided t-test across ensemble members.~~

The geographic distribution of ozone-related mortality changes, shown in Fig. 6, indicates that mortality reductions are concentrated primarily in Southeastern Asia. This spatial pattern aligns with the hemispheric asymmetry in the tropospheric ozone response observed in Fig. 3, where greater reductions in ozone concentrations occur in the ~~Southern Hemisphere~~ SH and
320 parts of Asia.

~~For both ozone and PM$_{2.5}$-related mortality, the range of total global deaths resulting from changes in air pollution illustrates the variability across ensemble members and highlights the importance of quantifying the internal variability within a model. The magnitude of these changes varies substantially across all ensemble members, emphasizing the role of internal variability in shaping the projected health impacts. Furthermore, the geographical distribution of the changes in ozone and~~ In Fig. 7b, the
325 evolution of global ozone-related deaths over time is consistent with the time series of global OSMDA8 (Fig. 5b). Overall, no clear long-term trend is evident in PM$_{2.5}$ ~~-related mortality reflect that the regional health benefits of SAI implementation are not uniform, but instead reflect underlying patterns in how surface air pollution responds to stratospheric aerosol perturbations. Both~~ and ozone-related mortality, as any underlying signal may be masked by the large ensemble variability in projected deaths (Fig. 7). Geographically, both ozone-and PM$_{2.5}$-related mortality changes exhibit substantial spatial variability, driven
330 by regional differences in how ozone and PM$_{2.5}$ concentrations respond to shifts in atmospheric chemistry, circulation, and precipitation patterns under SAI.

Figure 7 shows how global changes in mortality due to ozone and PM$_{2.5}$ evolve over time in our simulations. When aggregated globally, it is evident that the largest change in air-pollution related mortality is due to decreases in precursors and pollutants under the SSP2-4.5 scenario. ~~The differences~~ Differences between the futures with and without SAI, and those
335 between different amount of SAI cooling, are much smaller on a per-year basis, and in most cases within the range of variability for the ensemble estimates. This ~~clearly~~ demonstrates that the direct impact of deposited sulfate is ~~almost null, whereas climatic factors only minimally impact PM2.5~~ limited, and climatic factors minimally impact PM$_{2.5}$ changes under SAI. In contrast, ~~ozone-attributable mortality is~~ global ozone-related mortality is slightly lower in ARISE-SAI-1.0 than in ARISE-SAI-1.5, likely due to ~~enhanced cooling that reduced OH concentrations, thereby suppressing tropospheric ozone production~~larger
340 SAI-induced SH extra-tropical lower stratospheric ozone loss and the resulting reduction in ozone stratosphere-troposphere transport.

Figure 8 ~~presents~~ provides an alternative way of examining this relationship by plotting ensemble means ~~from ARISE-SAI-1.0 and ARISE-SAI-1.5 against their respective injection rates , rather than as a time evolution~~against injection rates in the SAI scenarios. For both simulations, PM$_{2.5}$-related mortality ~~exhibits a weakly positive trend~~ shows no clear linear scaling with
345 increasing injection rates. ~~However,~~, as substantial ensemble variability ~~, represented by the error bars, reflects considerable ensemble member spread in the mortality response. In contrast, ozone-attributable~~ and factors other than SAI affecting the evolution of mortality rates with time dominate the relationship. Ozone-attributable mortality remains consistently negative
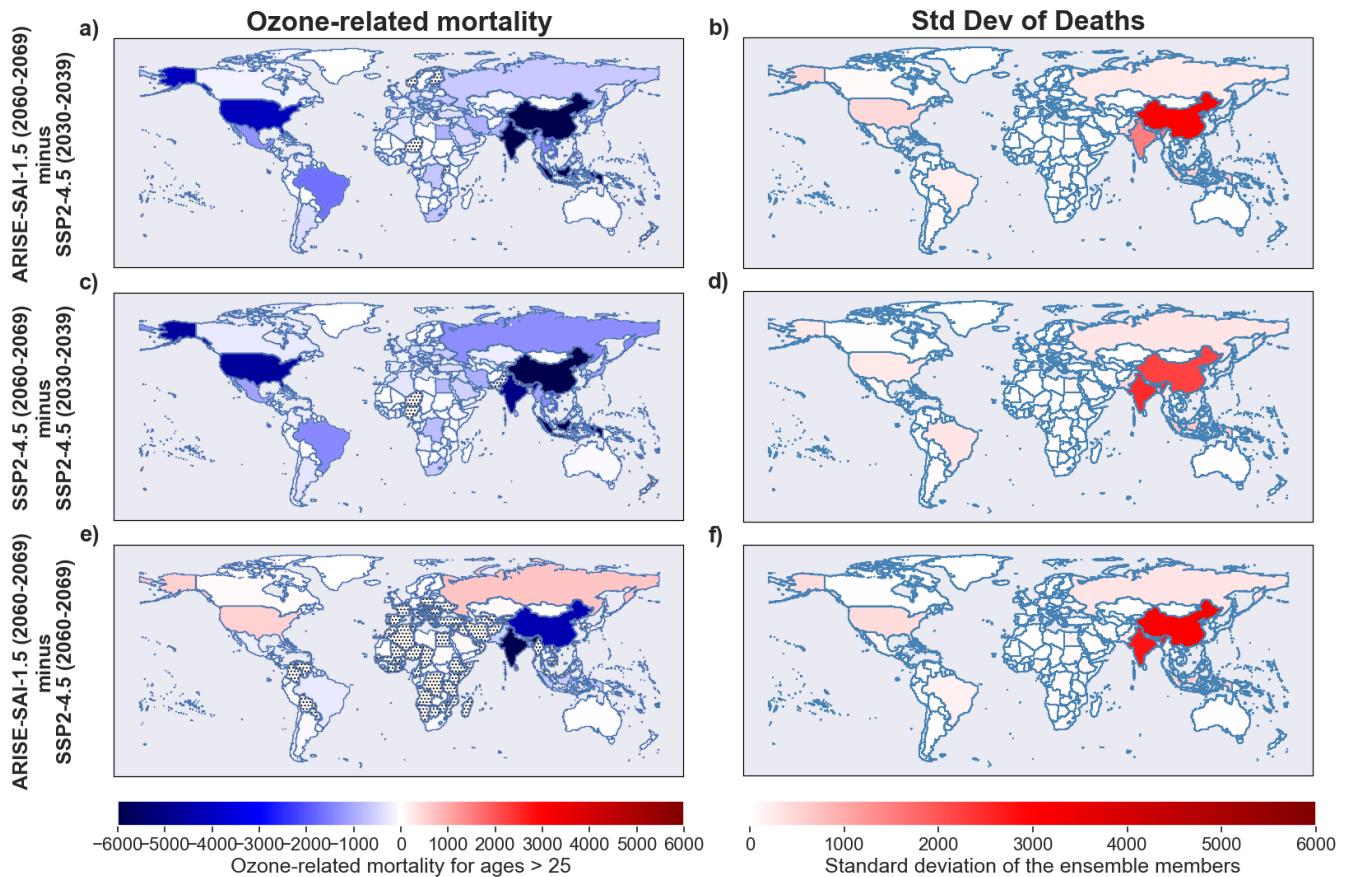
Figure 6. The ensemble-mean deaths from ozone for ages > 25. Stippling indicates countries where estimates of the ozone-related mortality are not statistically significant across ensemble members at the 95% confidence level.

across the entire injection range, indicating a reduction in ozone-related deaths under both ARISE-SAI-1.5 and ARISE-SAI-1.0. ~~These results reveal that pollution-driven mortality under SAI does not scale linearly or even monotonically with~~ (350) ~~the amount of SO₂ injected into the stratosphere. With the injection of SO₂ into the stratosphere, the resulting atmospheric responses involve complex feedbacks that we have tried to outline in this manuscript.~~

For $PM_{2.5}$-related mortality in particular, our component attribution analysis suggests that the primary driver of changes is not sulfate itself, but rather ~~arising~~ arises from changes in dust and secondary SOA concentrations ~~.~~ (Figs. 2 and 5a). Regional reductions in $PM_{2.5}$, particularly over Central America and central Sub-Saharan Africa, align with areas of increased (355) precipitation, highlighting the role of wet deposition and circulation-driven suppression of natural aerosol sources ~~.~~ (Fig. 1). However, the widespread lack of statistically significant precipitation or $PM_{2.5}$ changes across ensemble members suggests that internal variability and regional circulation shifts, rather than sulfate burden alone, govern the spatial and temporal patterns of $PM_{2.5}$-related health outcomes under SAI.
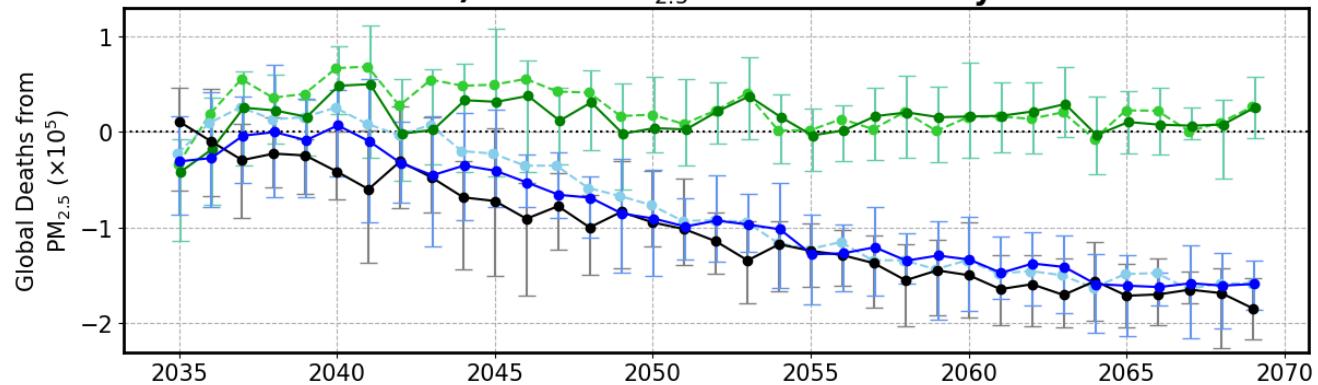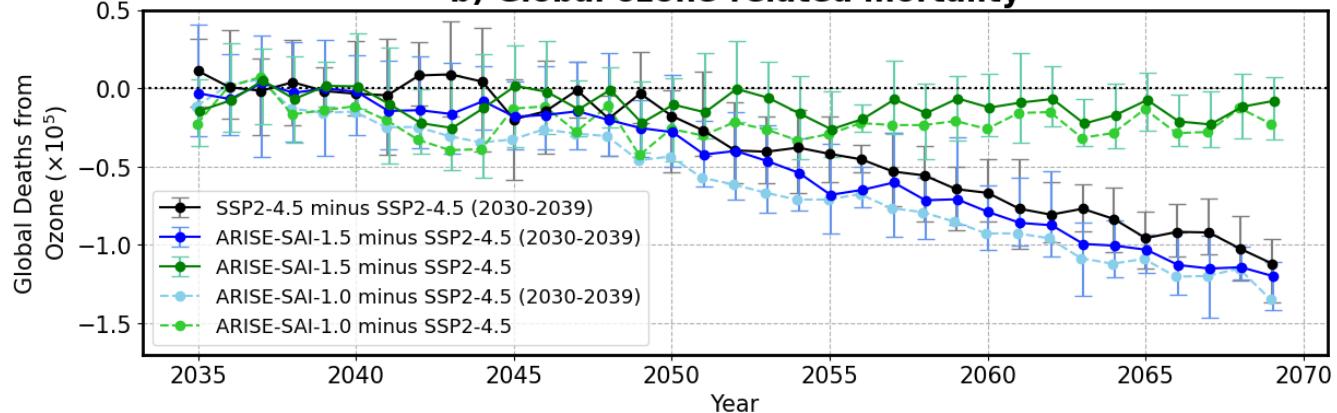
Figure 7. Global deaths from a) PM$_{2.5}$ and b) ozone evaluated as 1) ARISE-SAI-1.O minus SSP2-4.5, 2) ARISE-SAI-1.5 minus SSP2-4.5 (2030-2039), 3) ARISE-SAI-1.5 minus SSP2-4.5, 4) SSP2-4.5 minus SSP2-4.5 (2030-3029) and 5) ARISE-SAI-1.5 minus SSP2-4.5 (2030-2039). Error bars represent the full range of outcomes across the model ensemble, showing the minimum and maximum values.

For ozone, the mortality reductions appear more discernible. SO$_2$ is primarily injected in the ~~Southern Hemisphere~~SH, leading to decreased SH extra-tropical lower stratospheric ozone concentrations and the resulting reduction in SH surface ozone from reduced STE overwhelming any in-situ changes in tropospheric ozone chemistry there. In the ~~Northern Hemisphere~~NH, on the other hand, surface ozone increases due to the suppressed photochemical destruction under drier and colder troposphere ~~.~~(Fig. 3). These changes reflect the role of not only hemispheric asymmetries in sulfate burden alone but also those in STE and chemical processing arising from circulation changes and altered chemical regimes in shaping global ozone responses and associated health outcomes under SAI.

Taken together, these findings emphasize that air pollution-related health impacts under SAI are not governed mainly by the magnitude of SO$_2$ injected, but rather by the complex suite of dynamical, chemical, and aerosol responses in the Earth system–many of which are nonlinear and strongly influenced by internal variability. While our two large ensemble SAI simulations
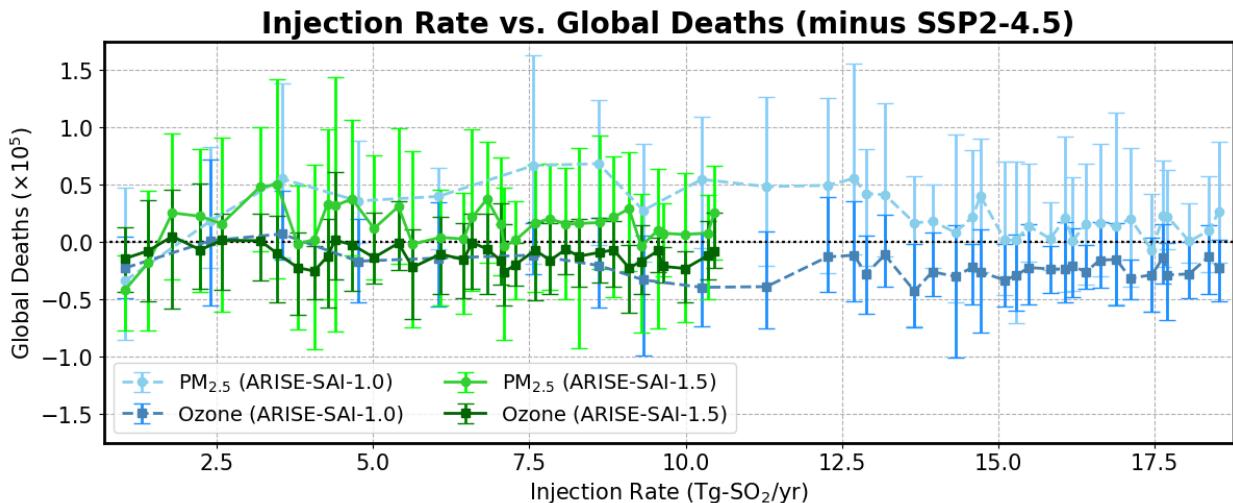
Figure 8. Global mortality differences for ARISE-SAI-1.5 minus SSP2-4.5 (shades of green) and ARISE-SAI-1.0 minus SSP2-4.5 (shades of blue) as a function of annual SO_2 injection rate (Tg-SO_2/yr), for $PM_{2.5}$-related deaths (green) and ozone-related deaths (green). Points represent ensemble means across years (2035–2069), with error bars indicating ensemble variability (± 1 standard deviation).

show no evidence for linear scaling with respect to injection rate, we acknowledge that longer simulations and additional

370   scenarios would be needed to more fully characterize how air quality related mortality is dependent on the SAI scenario under consideration.

3.3 ~~GBD~~ Global Burden of Disease super-region specific projections

Globally, ARISE-SAI-1.5 reduces total pollution-attributable mortality relative to a future without intervention (SSP2-4.5) by ~~0.41~~0.4%, driven by a ~~0.86~~0.9% increase in $PM_{2.5}$ and ~~1.26~~1.3% reduction in ozone-related deaths (Fig. 9a–b). However,

375   the direction and magnitude of health outcomes vary substantially across GBD super-regions. For instance, large percent increases in $PM_{2.5}$-related mortality occur in regions such as Central, Western and Eastern Europe. In contrast, regions like the Caribbean and Central Latin America exhibit reductions in $PM_{2.5}$-attributable ~~mortality—highlighting~~ mortality, highlighting the heterogeneous and sometimes adverse regional impacts of SAI.

For ozone-related mortality, the ensemble spread is also ~~large—both~~ large–both in magnitude and spatial ~~extent—especially~~

380   extent–especially in regions such as the Western and Eastern Sub-Saharan Africa and the Caribbean. Furthermore, while ~~BMR~~ national base mortality (cardiovascular, respiratory, and NCD+LRI baseline mortality rate) declines from 2030–2039 to 2060–2069 across all regions~~(see Appendix)~~, the magnitude of these changes is relatively small compared to the much larger shifts seen in air quality-related mortality.

In many regions, the ~~direction of the response varies across ensemble members, reflecting uncertainty~~ large ensemble spread

385   reflects uncertainties not only in the ~~scale~~ magnitude but also in the sign of the projected impact on air quality related mortality.
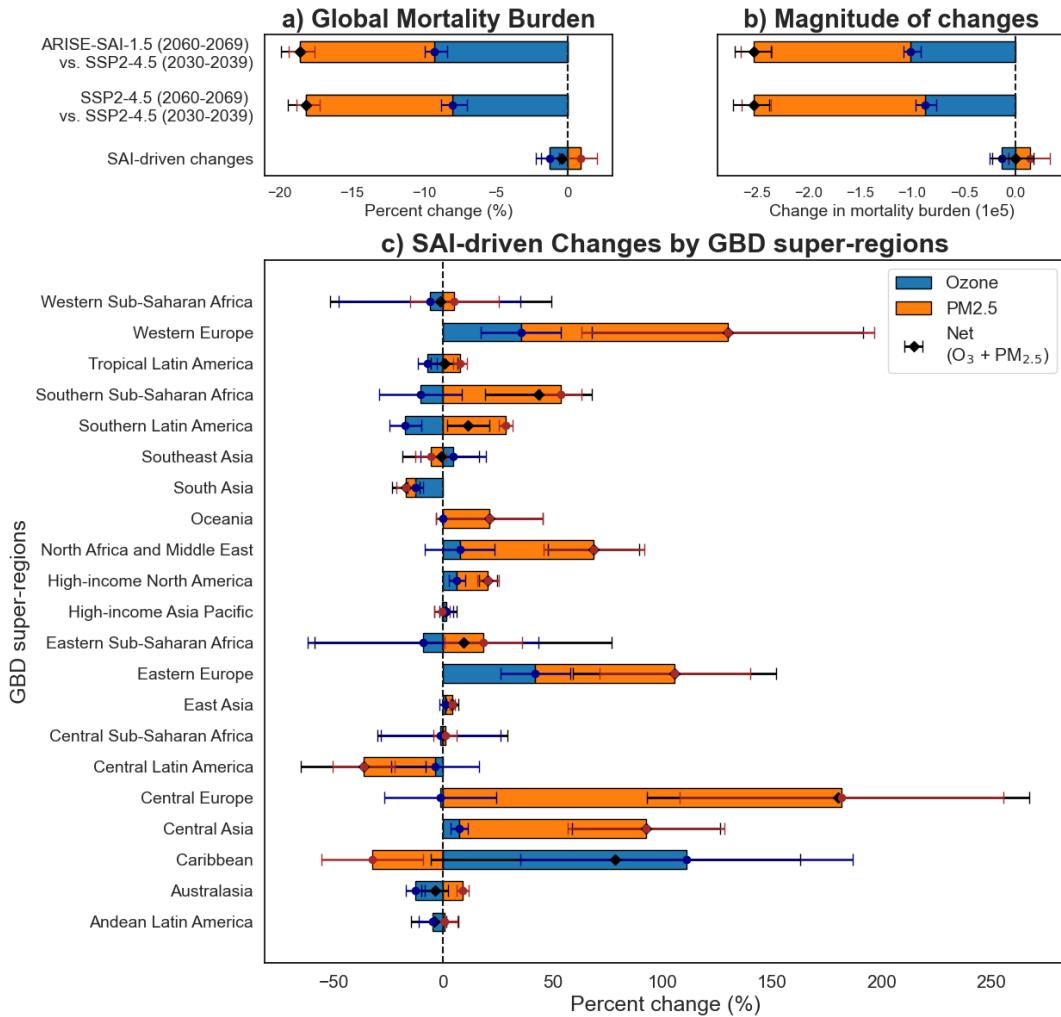
Figure 9. (a) Global percent change in mortality burden comparing ARISE-SAI-1.5 (2060–2069) and SSP2-4.5 (2030–2039), as well as SSP2-4.5 (2060–2069) and SSP2-4.5 (2030–2039). (b) Absolute global changes in mortality burden (in number of deaths). (c) Percent change in mortality burden by Global Burden of Disease (GBD) super-region between ARISE-SAI-1.5 (2060-2069) and SSP2-4.5 (2060-2069). Percent changes are calculated relative to baseline mortality rates. Positive values indicate an increase in mortality relative to the baseline, while negative values indicate reductions. Bars represent stacked contributions from ozone-related deaths (blue) and $PM_{2.5}$-related deaths (orange), with horizontal error bars indicating the ensemble spread (standard deviation) for each component and for the net total (black diamonds with error bars).

This spread arises from internal climate variability, which influences key drivers of air quality–such as atmospheric circulation, precipitation patterns, and chemical processing–and leads to diverging pollutant concentrations across ensemble members, even under identical forcing scenarios. These findings also highlight the spatial heterogeneity in health responses to SAI.

While global or hemispheric trends may point to a net decline in ozone-related mortality and an increase in PM$_{2.5}$-related mortality, such aggregates can mask substantial regional disparities. As a result, careful evaluation of region-specific trade-offs is critical when assessing the overall public health implications of SAI deployment.

4   Conclusions

This study evaluates the impacts of SAI on air quality–related mortality using a fully coupled climate model ensemble under the ARISE-SAI protocol. Unlike previous studies using CTMs (e.g., Eastham et al. 2018; Moch et al. 2023), which imposed stratospheric aerosols without capturing feedbacks on dynamics and transport, our use of ~~CESM2-WACCM6~~ CESM2(WACCM6) enables interactive coupling between aerosols, chemistry, and climate. ~~Our main finding is that the potential effects of SAI on surface air quality and associated mortality are relatively modest and largely fall within the range of internal model variability. In particular, pollution-related mortality are reduced by 18.2% in SSP2-4.5 compared to present day due to air quality policies, and by 18.6% in ARISE-SAI-1.5 due to the combination of air quality policies and SAI contributions, underscoring the importance of public policies that are having and will have large capacity to improve health outcomes (Vandyck et al., 2018).~~

We explore two potential sources of mortality: PM$_{2.5}$ and surface ozone exposure. PM$_{2.5}$ is affected both by direct deposition of sulfate from the stratosphere and by climatic conditions affecting other sources of particulates. Tropospheric O$_3$ changes from SAI can be driven by the combination of changes in stratospheric ozone and its transport to the troposphere, and by in-situ changes in tropospheric ozone chemistry driven by SAI-induced changes in ~~temperatures and UV photolysis. We note the~~ surface temperatures and photolysis. The latter are not fully considered in our study, as tropospheric ozone changes caused by direct aerosol impacts on actinic fluxes and photolysis rates are ~~fixed in the model at the present day levels~~ not included (although photolysis rates would still be affected indirectly by aerosol-driven changes to stratospheric ozone column above, and by cloud changes).

We find that the direct contribution of sulfate aerosols to PM$_{2.5}$-related mortality is minimal, primarily because much of the injected sulfate is transported poleward ~~, deposited at midlatitudes~~ and deposited at mid-latitudes, leading to a relatively diffuse and spatially uniform distribution. Furthermore, a portion of sulfate particles exceed the PM$_{2.5}$ size threshold and therefore does not contribute to fine particulate mass. Subsequently, the total mass of sulfate aerosols reaching the surface is insufficient to meaningfully alter concentration thresholds associated with mortality outcomes. Instead, regional changes in PM$_{2.5}$ concentrations and the corresponding health impacts are mainly driven by shifts in precipitation patterns and/or circulation, which affect the wet removal of non-sulfate species such as dust and secondary organic aerosols, consistent with Eastham et al. (2018).

Likewise, we find that ozone-related mortality is projected to maintain its decrease globally due to changes in pollutant sources even under SAI; but, when comparing the two future scenarios, the SAI impact result in a change in the spatial pattern reflecting a hemispheric asymmetry in the tropospheric ozone response, leading to a slight increase in surface ozone in the ~~Northern Hemisphere~~ NH and a decrease in the ~~Southern Hemisphere.~~

SH. However, some uncertainties related to the specific evolution of surface ozone remain, particularly due to the absence of ~~interactive photolysis in the troposphere~~the direct aerosol effect on the photolysis rates, which could lead to an underestimation of chemical feedbacks ~~involving ozone and hydroxyl radicals. In reality, stratospheric aerosols alter the spectral distribution of solar radiation, affecting both the amount and quality of light reaching the lower atmosphere. While SAI increases aerosol optical depth and reduces shortwave radiation (∼400–850 nm), catalytic ozone depletion allows greater transmission of ultraviolet (UV) radiation at wavelengths below 345 nm (Moch et al., 2023). This enhanced UV flux accelerates ozone photolysis ($O_3 + h\nu \rightarrow O_2 + O(^1D)$) and triggers chemical feedbacks that can influence surface ozone and oxidant levels, including increased production of hydroxyl radicals (OH), the primary sink of methane (CH_4). These chemical feedbacks linking photolysis, ozone, OH, and CH_4 are omitted in simulations without interactive photolysis, limiting our ability to capture the full extent of surface ozone responses and oxidant perturbations under geoengineering. Nevertheless, studies including interactive tropospheric photolysis like Moch et al. (2023) do not show a particularly large impact from its inclusion on surface ozone concentrations.~~ in the troposphere.

~~There are other factors that might lead us to different estimates of both $PM_{2.5}$ and ozone-related mortality rates.~~

All mortality estimates in our future scenarios are calculated using the fixed 2020 population distribution. This approach isolates the effects of air quality changes by removing confounding influences from projected population growth or redistribution. However, ~~it is important to note that~~ mortality rates could be significantly affected by demographic and population changes, such as aging, urbanization, or overall population growth, which are not considered in this study. As a result, our estimates may not fully reflect future health impacts under evolving demographic conditions. ~~Also, while CESM2-WACCM generally performs comparably to other CMIP6 models in simulating surface air pollution (Turnock et al., 2020), it exhibits a high bias in surface ozone relative to TOAR observations and a low bias in surface $PM_{2.5}$ compared to observational datasets.~~

Furthermore, our analysis is based on a single climate model and two closely related SAI scenarios, and thus the results may be both scenario and model-dependent. However, comparisons between ARISE-SAI-1.0 and ARISE-SAI.1.5 indicate that global $PM_{2.5}$-related mortality does not increase ~~due to SAI with~~ significantly under higher SO_2 injection amounts, whereas ozone-related mortality decreases sightly with higher injection rates due to lower temperatures. This suggests that variability in PM2.5-related mortality may be more strongly influenced by changes in dust or biomass-burning-derived PM2.5 driven by circulation responses to SAI, rather than directly by the total amount of SO_2 injected. However, because SO_2 is primarily injected in the SH for these scenarios, it may also be relevant to examine whether similar observations emerge under a broader set of scenarios.

Future assessments of SAI impacts on air quality and related mortality could be improved by multi-model intercomparisons to better constrain the contributions of non-sulfate aerosol species, such as dust, ~~black carbon, and secondary organic aerosols~~BC, and SOA, as well as to capture the range of model uncertainty in aerosol-chemistry climate interactions. Additionally, improved representation and observational verification of large-scale circulation responses, particularly changes in the Brewer-Dobson Circulation and STE, are essential for understanding the transport and distribution of injected aerosols, as well as their downstream effects on regional air quality. Furthermore, the incorporation of more detailed aerosol microphysics, including size-resolved coagulation, nucleation, and heterogeneous chemistry, would allow for a more accurate simulation of aerosol

growth, lifetime, and radiative properties. Together, these efforts would enable more comprehensive and policy-relevant evaluations of SAI's atmospheric and health impacts.

While this study focused on the air quality-related health impacts of SAI, it is important to acknowledge that other health-relevant outcomes, such as changes in surface UV radiation and regional ~~temperature~~temperatures, were not evaluated here but may also carry significant implications. Preliminary analysis of surface UV radiation differences between ARISE-SAI-1.5 and SSP2-4.5~~indicate very modest changes and were not included in the manuscript. Like air quality , these impacts are embedded within~~, calculated with the Tropospheric Ultraviolet and Visible (TUV) model developed at NCAR (Madronich and Flocke, 1999; Visioni e, show that changes in surface UV are small (between -5.3 to -6.1% globally). This finding is broadly consistent with previous studies that examined UV responses to SAI, including recent work highlighting that while atratospheric aerosol perturbations can modify photolysis rates, the net surface UV changes tend to be modest (Bardeen et al., 2021). Although small, such changes could still influence surface ozone through altered photochemistry and may affect secondary particulate matter, such as POM and SOA, by modifying photolysis-driven oxidation pathways. These potential impacts remain an important avenue for future investigation.

In addition, other processes known to affect air quality under climate change, such as changes in planetary boundary layer height (Deng et al., 2023; Li et al., 2017, 2019) and lighting activity (Murray, 2016; Grewe, 2009) could play a significant role to the simulated air quality response to SAI. However, the scope of this paper is to assess the net outcome of these combined processes for surface-level $PM_{2.5}$ and ozone concentrations, and their associated health effects, across large ensembles. As with air quality, they are part of a broader suite of SAI-induced environmental changes that ~~merit further explorations. Furthermore, internal~~ warrant further exploration.

Internal climate variability plays a critical role in modulating aerosol transport, chemical processes, regional temperature responses, and stratospheric ozone dynamics. By resolving dynamic feedbacks between aerosols, transport, and atmospheric chemistry, our modeling approach overcomes key limitations of earlier CTM-based studies, enabling more realistic estimates of SAI-induced air quality and health outcomes. This highlights the importance of using fully coupled Earth system models when evaluating the policy-relevant consequences of geoengineering strategies and reinforces the need to account for natural variability when assessing human health impacts. Our results, which emphasize the importance of ensemble approaches for air pollution mortality estimates, highlight a general need for robust ensemble-based evaluations across all dimensions of SAI's potential risks and trade-offs.

When viewed in the context of climate change impacts on air quality, our findings suggest that the additional effects of SAI are small relative to both internal variability and policy-driven improvements. Prior studies have identified a "climate penalty" on air quality, in which rising temperatures and shifts in meteorology under climate change can increase surface ozone and fine particulate concentrations, resulting in increases in air pollution-related mortality (Fiore et al., 2015; Doherty et al., 2013; Fu and Tian, 201. SSP2-4.5 represents a moderate mitigation and policy pathway, in which partial greenhouse gas reductions are achieved, leading to some reductions in CO_2, CH_4, and co-emitted air pollutants, and consequently modest improvements in air quality relative to higher-emission futures (Hussain, 2025; Nazarenko et al., 2022; Shim et al., 2021). In our simulations, SSP2-4.5 leads to an 18% (ensemble range: -19 to -17%) reduction in air pollution-related mortality relative to present day (2030-2039),

driven primarily by emissions policies. Under ARISE-SAI-1.5, mortality is reduced by a similar amount (19%; -20 to -18%), with the net impact of SAI largely falling within the range of internal variability. This finding highlights that while SAI can shift the spatial distribution of ozone and particulate matter, particularly through hemispheric asymmetries in stratospheric aerosol loading and associated dynamical responses, the dominant driver of future health outcomes remains the strength of air
495 quality policies (Vandyck et al., 2018). Our results therefore align with the broader literature emphasizing that while internal variability can obscure the precise effects of climate change (Pienkosz et al., 2019; Garcia-Menendez et al., 2017) and even climate interventions on air quality, sustained emissions reductions are important in determining future air quality and health outcomes.

. Code used in computing the $PM_{2.5}$ and ozone-related mortality can be found at https://doi.org/10.5281/zenodo.15696232 (Wang, 2025). All
500 the data presented in this paper are available at https://doi.org/10.5281/zenodo.6473954 (Richter and Visioni, 2022b) from the CESM2(WACCM6) SSP2–4.5 simulations and at https://doi.org/10.5281/zenodo.6473775 (Richter and Visioni, 2022a) from the ARISE-SAI simulations.

5   Appendix

A comparison of the AF used in this study to calculate $PM_{2.5}$ and ozone associated mortality is presented in Fig. ??. The AF for $PM_{2.5}$ (NCD+LRI) is lower per μg/m^3 of $PM_{2.5}$ because its harm is spread across many diseases, while ozone's AF is
505 higher per ppb for cardiovascular and respiratory diseases because it acts more acutely and specifically on those systems. In Fig. ??a, $PM_{2.5}$ exhibits a near-linear dose-response relationship with mortality , indicating that risks accumulate gradually. In contrast, Fig. ??b shows that ozone has a steeper curve at lower doses, meaning even small increases in surface O_3 can disproportionately elevate the risk of cardiovascular and respiratory diseases (Burnett and Cohen, 2020).

(a) Attributable fraction (AF) of mortality due to PM2.5 exposure as a function of PM2.5 concentration, based on the
510 mean relative risk (RR) across age groups for non-communicable diseases (NCD) and lower respiratory infections (LRI). (b) Attributable fraction of mortality due to ozone (OSMDA8) exposure, with separate curves for cardiovascular and respiratory causes. Risk functions are based on excess mortality relative to baseline thresholds (2.5 ppm for NCD+LRI, 40 ppb for cardiovascular and 32.4 ppb for respiratory outcomes).

Mean fractional contribution of PM2.5 species under ARISE-SAI-1.5 (2060–2069). Color shading shows the ensemble-mean
515 fraction of each species relative to total PM2.5. Stippling marks regions where the ensemble standard deviation exceeds the 90th percentile, indicating high internal variability.

Changes in the fractional contribution of $PM_{2.5}$ species between ARISE-SAI-1.5 and SSP2-4.5 (2060–2069). Positive (red) values indicate an increased contribution under ARISE-SAI-1.5, while negative (blue) values indicate a decreased contribution. Sparse stippling marks areas where changes are not statistically significant (p>0.05) based on a two-sided t-test across
520 ensemble members.

Seasonal changes in ozone between ARISE-SAI-1.5 and SSP2-4.5.

Zonal-mean differences in methane (a-b), NO$_X$ (d-f), and OH (g-i) concentrations (mol mol^{-1}) are presented for ARISE-SAI-1.5 (2060–2069) minus SSP2-4.5 (2030–2039), SSP2-4.5 (2060–2069) minus SSP2-4.5 (2030–2039), and ARISE-SAI-1.5 (2060–2069) minus SSP2-4.5 (2060–2069). Shading indicates the concentration differences, with black contours in the first two columns showing the absolute zonal-mean concentrations.

. CW performed the analysis of the model simulations and wrote the manuscript. DV helped outline and write the manuscript and provided insight on the analyses. GC provided guidance on how to conduct the mortality calculations and helped write the manuscript. EB provided insight on the analysis and contributed to the writing of the manuscript.

. At least one of the (co-)authors is a member of the editorial board of Atmospheric Chemistry and Physics.

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