Characterizing and quantifying uncertainty in projections of climate change impacts on air quality

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Abstract
Climate change can aggravate air pollution, with important public health and environmental consequences. While major sources of uncertainty in climate change projections—greenhouse gas (GHG) emissions scenario, model response, and internal variability—have been investigated extensively, their propagation to estimates of air quality impacts has not been systematically assessed. Here, we compare these uncertainties using a coupled modeling framework that includes a human activity model, an Earth system model of intermediate complexity, and a global atmospheric chemistry model. Uncertainties in projections of U.S. air quality under 21st century climate change are quantified based on a climate-chemistry ensemble that includes multiple initializations, representations of climate sensitivity, and climate policy scenarios, under constant air pollution emissions. We find that climate-related uncertainties are comparable at mid-century, making it difficult to distinguish the impact of variations in GHG emissions on ozone and particulate matter pollution. While GHG emissions scenario eventually becomes the dominant uncertainty based on the scenarios considered, all sources of uncertainty are significant through the end of the century. The results provide insights into intrinsically different uncertainties in projections of air pollution impacts and the potential for large ensembles to better capture them.

1. Introduction
Climate change is expected to exacerbate air pollution during the 21st century [1, 2]. Under a warmer climate, projected changes in meteorology, atmospheric chemistry, and natural emissions will deteriorate air quality over many populated areas [3]. This climate-induced impact on air quality, commonly termed the climate penalty, is distinct from changes brought about by changing pollutant emissions and can have significant public health and economic consequences [4–8]. The Fourth National Climate Assessment projects that in the absence of climate policy, by 2090 the climate penalty on ozone (O\textsubscript{3}) pollution will annually cause 500 additional deaths and cost $8 billion in the U.S., compared to air quality under a climate-mitigation pathway [6, 9]. Although the interactions between particulate matter pollution and climate change are less clear [3, 10], a climate penalty is also expected for PM\textsubscript{2.5} (particulate matter with aerodynamic diameter \( \leq \)2.5 \( \mu \)m) [11–13], the environmental pollutant responsible for the largest public health burden [14]. Climate change mitigation policies can reduce health damages by modulating the climate penalty and its impacts [15], but estimates of these co-benefits are subject to significant uncertainties.

Characterizing uncertainty is integral to projections of climate change and its impacts [16, 17]. Uncertainty in climate projections stems from three major sources: greenhouse gas (GHG) emissions scenario, climate model response, and internal variability [18]. Internal variability arises from natural variations in the climate system and is most effectively captured by initial condition ensembles [19–23]. Model response uncertainty is the differing response...
of climate models to the same radiative forcing and has been assessed by quantifying variability in projections across multi-model ensembles [21]. In the absence of multi-model ensembles, altering the climate sensitivity in a single-model framework has been shown to approximate model response for uncertainty analyses [24]. Emissions scenario uncertainty can be addressed by increasing the accuracy of emissions estimates or projecting multiple climate policy futures. Understanding how these drivers of uncertainty propagate to projections of climate impacts is critical for policy analyses considering air quality impacts and co-benefits of mitigation strategies. While climate uncertainties in air quality projections have been explored [23, 25–28], these uncertainties have not been systematically weighed or compared.

Here, we quantify and compare major uncertainties in projections of the climate penalty on U.S. air quality. Uncertainties are derived from an extensive simulation ensemble designed to simulate climate-induced impacts on air pollution and generated with a modeling framework that includes a human activity model, an Earth system model of intermediate complexity, and a global atmospheric chemistry model. Uncertainties in PM$_{2.5}$ and O$_3$ impacts due to internal variability, GHG emissions scenario, and model response are characterized in the ensemble by multiple model initializations, climate policy scenarios, and representations of climate sensitivity. We find that these uncertainties are comparable at mid-century and can make it difficult to discern the impact of changes in GHG emissions on U.S. air quality. By the end of the century, however, GHG emissions scenario is the dominant source of uncertainty followed by climate sensitivity, while internal variability remains close to its mid-century levels. We further examine the influence of internal variability on air quality projections ranging from annual to decadal scale. The analysis also examines the influence of climate sensitivity in simulations of air quality under climate change for the first time, providing insights into the weight of model response uncertainty in projections of air quality impacts. The results of this analysis highlight the potential impacts that future large and multi-model climate-chemistry ensembles can have on improving our understanding of climate change effects on air pollution in the face of uncertainty.

2. Methods

2.1. Modeling framework

We use a modeling framework that couples the Massachusetts Institute of Technology Integrated Global System Model and the National Center for Atmospheric Research Community Atmosphere Model (MIT IGSM-CAM) [29] to generate the simulation ensemble used in this study. The MIT IGSM is an integrated assessment model that links the MIT Earth System model (MESM) [30], an earth system model of intermediate complexity, and Economic Projection and Policy Analysis (EPPA) model [31, 32] to produce internally consistent projections of global climate, policy, and economic activity. The MESM includes atmosphere, ocean, and land components to represent Earth system processes and interactions [30]. The EPPA model is a computable general equilibrium model of the world economy that projects economic activity and emissions of climate forcers under policy constraints. A feature of the MIT IGSM-CAM framework is the ability to alter climate model response by modifying climate sensitivity with a cloud radiative adjustment method [33] and to conduct simulations under different representations of internal variability generated by perturbing initial atmospheric, land, and ocean conditions in the MESM. MIT IGSM-CAM simulations have been shown to realistically reproduce observations of the historical and present-day climate system [29, 30]. The framework has been used to project climate change impacts on multiple sectors, including public health, energy use, and infrastructure [34]. Here, three-dimensional meteorological fields projected by the MIT IGSM-CAM framework are used to simulate global atmospheric chemistry with the CAM with Chemistry (CAM-Chem) at 1.9° × 2.5° horizontal resolution. Interactive chemistry and two-way feedbacks between the MIT IGSM-CAM and CAM-Chem fields are not modeled. By using one-way coupling and relying on an Earth system model of intermediate complexity with high computational efficiency, the framework allows the simulations required by the study’s uncertainty analysis, thousands of years of modeled global air quality, to be completed. Detailed descriptions of the MIT IGSM-CAM modeling framework and approach are included in [29, 35].

Global atmospheric composition is modeled with CAM-Chem (version 1.1.2) [36], driven by MIT IGSM-CAM meteorological fields. In the configuration used, CAM-Chem simulates atmospheric concentrations of over 100 gas phase and aerosol species from surface level to the lower stratosphere. For this study’s projections of climate impacts on PM$_{2.5}$, we consider changes in ground-level concentrations of sulfate (SO$_4$), ammonium nitrate (NH$_4$NO$_3$), organic aerosol (OA), and black carbon, and estimate PM$_{2.5}$ mass following [37]. Secondary OA production is simulated with a two-product scheme linking its formation to oxidation of non-methane hydrocarbons [36]. The simulations use an optimized dry deposition scheme [37] and model the effect of temperature on biogenic isoprene and monoterpene emissions based on the Model of Emissions and Aerosols [36]. The response of other internal emissions sources to climate change, including wildfires and dust emissions, is not included in the projections. CAM-Chem predictions of O$_3$ and PM$_{2.5}$ concentrations have been
evaluated against observations, showing the model is able to generally reproduce the distribution of ground-level \( \text{O}_3 \) and aerosol species over the U.S. [36, 38, 39]. When CAM-Chem is coupled to the MIT IGSM-CAM, the frameworks’ performance for historical \( \text{O}_3 \) compared against monitoring site measurements is similar to that reported in CAM-Chem model evaluations [36], with the simulations reproducing observed spatiotemporal patterns but exhibiting a positive bias against observations, a typical characteristic of chemistry-climate models [40]. The framework’s predictions of \( \text{PM}_{2.5} \) reproduce average annual \( \text{PM}_{2.5} \) concentrations observed at over 400 U.S. EPA monitoring sites during the historical period, although the comparisons indicate a positive bias for \( \text{SO}_2 \) and a negative bias for \( \text{EC} \) and \( \text{OA} \) in the modeled results [26]. In the CAM-Chem simulations, emissions of air pollutants and precursors are based on the Precursors of Ozone and their Effects in the Troposphere inventory [36]. To isolate the effect of climate on air quality, anthropogenic emissions are fixed at start-of-century levels (2000) in all atmospheric chemistry simulations. Additional description and discussion of the CAM-Chem framework and simulations are included in [15, 25, 26].

2.2. Simulation ensemble of climate change impacts on U.S. air quality

The study’s modeling framework was used to generate a simulation ensemble of 21st century climate change impacts on air quality with multiple realizations of internal variability, climate sensitivity, and climate policy and is illustrated in figure S1. Three scenarios of economic activity, climate forcer emissions, and climate change are used to investigate emissions scenario uncertainty. The scenarios, developed for U.S. Environmental Protection Agency’s (EPA) Climate Change Impacts and Risk Analysis (CIRA) Program [9], include a reference scenario (REF) with no global climate policy, unconstrained GHG emissions, and a total radiative forcing of 10 W m\(^{-2}\) at the end of the century, and two stabilization scenarios that constrain total radiative forcing in 2100 to 4.5 W m\(^{-2}\) (P4.5) and 3.7 W m\(^{-2}\) (P3.7). The REF scenario is comparable to the Representative Concentration Pathway (RCP) 8.5, with atmospheric \( \text{CO}_2 \) reaching approximately 830 ppm by 2100, compared with over 900 ppm under RCP 8.5 [41]. Higher radiative forcing in REF is primarily due to larger anthropogenic and biogenic \( \text{CH}_4 \) emissions [42]. Climate stabilization scenarios P4.5 and P3.7 lead to end-of-century \( \text{CO}_2 \) concentrations between those of RCP4.5 and RCP2.6 [41], with the implementation of a global tax on carbon emissions constraining them to lower than 500 ppm. Scenario likelihood is not considered in uncertainty estimates. The scenarios cover a broad range of emissions and radiative forcing levels, from very high GHG emissions to ambitious emissions reductions. While recent analyses suggest that high \( \text{CO}_2 \) emissions scenarios (e.g. RCP 8.5 and Socio-economic Pathway SSP5-8.5) have a low likelihood [43], we include the REF scenario as an informative upper limit scenario without climate change mitigation, in line with the Intergovernmental Panel on Climate Change’s (IPCC’s) Sixth Assessment Report (AR6) which concludes that such future concentration levels cannot be disregarded [44].

To investigate the influence of climate model response in projections of climate change impacts on air quality, simulations are conducted with three climate sensitivities corresponding to the IPCC AR6’s best estimate, 3.0 °C (CS30), and values at the lower and upper end of the assessment’s very likely range, 2.0 °C (CS20) and 4.5 °C (CS45) [44]. Under the REF scenario and a 3.0 °C climate sensitivity, U.S. average temperature is projected to increase by 5.4 °C relative to start-of-century, within the National Climate Assessment’s projected range for the RCP8.5 scenario [45]. Temperature changes projected in our ensemble under each climate sensitivity and policy scenario considered are included in table S2. While different climate sensitivities among ensemble members are used to represent varying climate model response, they do not fully capture model response uncertainty. Meteorological variables influencing air pollution may not scale directly with climate sensitivity, and multi-model ensembles are better suited to capturing differences responses to forcings. This analysis examining climate sensitivity, however, is the first to specifically explore a major component of structural uncertainty in projections of climate change impacts on air quality. To represent a range of internal variability, five realizations of internal variability are included in the ensemble, each generated by applying initial conditions in the atmosphere and land components in the MIT IGSM-CAM simulations taken from a random year of a pre-industrial control simulation [29]. Detailed descriptions of the scenarios’ economic, emissions, and climate projections are presented in [24, 42].

Atmospheric chemistry is modeled with CAM-Chem for 30 year periods at the start (1981–2010), middle (2036–2065), and end (2086–2115) of the 21st century, under each policy scenario, climate sensitivity, and realization of internal variability. The resulting ensemble includes 150 annual simulations of U.S. air quality for each of the three policy scenarios, climate sensitivities, and time-periods considered. In total, over 2800 annual simulations of atmospheric chemistry were conducted for this analysis. Climate change impacts on \( \text{O}_3 \) and \( \text{PM}_{2.5} \) pollution are estimated as the difference between concentrations simulated with REF at the start of the century and the future periods simulation with the corresponding climate sensitivity. Population-weighted estimates are based on U.S. population distribution at the start of the century using the Gridded Population of the World, Version 3 dataset [46]. Regional averages are
2.3. Uncertainty estimates
We weigh climate-related uncertainties in projections of climate change impacts on U.S. air quality by applying the methodology of [24] to the CAM-Chem simulation ensemble. Here, we define scenario, climate sensitivity, and internal variability uncertainty in air quality projections as the range in simulated pollutant concentration impacts resulting from different GHG emissions scenarios, climate sensitivities, and climate model initializations, respectively. Uncertainties are determined at the middle and end of the 21st century. We begin by computing the climate-induced change (the climate penalty) for each annual projection in the ensemble as the change in annual average ground-level daily maximum 8 h average O$_3$ or daily average PM$_{2.5}$, relative to start-of-century concentrations of the corresponding annual simulation in the matching initial condition and climate sensitivity ensemble member.

To determine scenario uncertainty, we first control for internal variability by averaging across 30 year periods and initial condition ensemble members under each emissions scenario and climate sensitivity. This results in projected mean changes in air quality at the middle and end of the 21st century for each emissions scenario and climate sensitivity. Next, we find the range between the emissions scenarios resulting in the largest and smallest air quality impact at each climate sensitivity. Finally, we calculate the mean of these ranges to control for climate sensitivity. The resulting range is defined as the uncertainty in the climate impact on air quality due to emissions scenario. For climate sensitivity uncertainty we repeat the process, instead calculating the range between climate sensitivities resulting in the largest and smallest impacts under each emissions scenario, and then averaging these ranges.

To compute uncertainty due to internal variability, we first average across each 30 year period in the ensemble, each of which is associated with a different climate model initialization. We then calculate the difference between the initial condition ensemble member means with the largest and smallest air quality impacts under each policy scenario and climate sensitivity. The average of these ranges represents a range of internal variability uncertainty in 30 year mean projections. In addition to estimating internal variability uncertainty for 30 year simulations, we explore internal variability for 15, 10, 5, and 1 year time slices. We measure these uncertainties by calculating rolling 15, 10, 5, or 1 year averages within the 30 year period of each initial condition ensemble member, resulting in a series of average air quality impact values for each time slice length, under each climate sensitivity and emissions scenario. We group rolling averages by climate sensitivity and policy scenario, and then calculate the internal variability uncertainty as the average of the ranges within each group.

3. Results
3.1. Uncertainty in climate change impacts on U.S. air quality
Climate change is projected to negatively impact ground-level O$_3$ and PM$_{2.5}$ pollution in the U.S. Climate stabilization policies targeting total radiative forcings of 4.5 and 3.7 W m$^{-2}$ (P4.5 and P3.7) by 2100 under a climate sensitivity of 3.0 $^\circ$C (mean surface temperature change per doubling of atmospheric CO$_2$) limit projected population-weighted annual impacts at the end of the century to 0.4 and 0.3 $\mu g$ m$^{-3}$ for PM$_{2.5}$, and 0.3 and 0.6 ppb daily maximum 8 h O$_3$ (all O$_3$ concentrations are reported here as daily maximum 8 h average). However, under a no-policy reference GHG emissions scenario (REF; see section 2), a less likely but informative scenario, mean climate penalties on annual U.S. population-weighted PM$_{2.5}$ and O$_3$ are projected to be 0.6 $\mu g$ m$^{-3}$ and 0.8 ppb at mid-century, and 1.5 $\mu g$ m$^{-3}$ and 3.2 ppb by the end of the century. For O$_3$-season (May–September) concentrations, the projected mean climate penalty under the REF scenario at century end is 8.1 ppb. Climate change impacts on O$_3$ levels projected under a 3 $^\circ$C climate sensitivity agree with previously reported climate penalty estimates over the Northeast, Midwest, Southeast, and California, but show differences in other areas, in particular over the Northwest [3, 48, 49]. There is less consistency among reported simulations of climate change impacts on PM$_{2.5}$ pollution. The projections used here agree with those from specific studies over particular areas (e.g. the Northeast and Southeast), but can differ with others depending on PM$_{2.5}$ component or region [13, 50, 51]. Comparisons between this ensemble’s projections and others are further discussed in [25, 26]. Despite clear mean penalties by the end of the century, a closer look at the ensemble of over 2800 years of simulated air quality (figures 1(a) and (b)) reveals substantial uncertainty in these estimates of climate change impacts on pollutant concentrations. Internal variability can lead to projections of climate-induced changes on annual U.S. population-weighted PM$_{2.5}$ and O$_3$ that range from –0.2 to 3.2 $\mu g$ m$^{-3}$ and –1.5 to 8.0 ppb, respectively, and simulations under the different climate sensitivities considered (2.0 $^\circ$C, 3.0 $^\circ$C, and 4.5 $^\circ$C) differ by up to 1.5 $\mu g$ m$^{-3}$ and 4.1 ppb in the mean climate impacts projected for a given emissions scenario.

Uncertainties in projected climate change impacts on U.S. air quality arising from GHG emissions scenario, climate sensitivity, and internal variability are compared in figures 1(c)–(f). Here, the uncertainty associated with each source is defined.
as the average range across 30 year mean climate change impacts projected under the different combinations of emissions scenario, climate sensitivity, and internal variability included in the ensemble (see figure S1 and section 2 for a detailed description). At mid-century, the uncertainties in projected climate impacts associated with each source are similar for annual population-weighted concentrations of both \(O_3\) (0.5–0.8 ppb) and \(PM_{2.5}\) (0.3–0.4 \(\mu\)g m\(^{-3}\)). These uncertainties are significant relative to mid-century ensemble-mean projections of climate penalties on U.S. air pollution. By the end of the century, the uncertainty associated with GHG emissions scenario is clearly largest, 30% higher than climate sensitivity uncertainty in \(PM_{2.5}\) impacts and over twice as high as climate sensitivity uncertainty in \(O_3\) impacts, while uncertainty due to internal variability is the smallest. However, even at the end of the century all sources of climate-related uncertainty remain significant. Internal variability remains consistent throughout the simulations. It is comparable with the uncertainties associated with GHG emissions scenario and climate sensitivity at mid-century and is approximately 20%–40% of that imposed by emissions scenario at century end. Climate sensitivity uncertainty grows by the end of the century, becoming distinctly larger than internal variability. The relative weights of these climate-related uncertainties reflect those reported for projections of surface temperature and precipitation \[18, 21, 24\], with internal variability as the major source of uncertainty early in the 21st century and GHG emissions scenario becoming the largest towards the end of the century. These estimates also highlight the challenges imposed by internal variability and uncertain climate sensitivity on projecting a climate response in air quality to variations in GHG emissions for time horizons extending over few decades.

### 3.2. Uncertainty in projections of regional air quality impacts

Uncertainties in climate change impacts on air pollution across U.S. National Climate Assessment regions \[47\] are compared in figure 2. The largest uncertainties in projections of regional climate-induced changes to \(O_3\)-season and annual \(PM_{2.5}\) concentrations are in the Northeast, Midwest, and Southeast, the regions anticipated to experience the largest climate penalties on air quality. Uncertainties associated with GHG emissions scenario and climate sensitivity are 2–8 times larger in the Northeast than the Western U.S. Internal variability is largest in the Midwest and Northeast, and lowest in the Southwest and Northwest. Although the relative weights of each uncertainty vary regionally, and internal variability has a larger influence on regional penalties, they are consistent
Figure 2. Uncertainties in U.S. regional projections of climate-induced impacts on regional May–September (O₃-season) O₃ and annual PM₂.₅ ground-level concentrations associated with GHG emissions scenario, climate sensitivity, and internal variability at mid-century and end-century.

Figure 3. Uncertainties in projections of climate-induced impacts on annual O₃ and PM₂.₅ ground-level concentrations over the U.S. associated with GHG emissions scenario, climate sensitivity, and internal variability at mid-century and end-century.

with uncertainties for national population-weighted concentrations. At mid-century, differences between uncertainties associated with each source are small and, for most regions, internal variability is the largest uncertainty in projections of climate change impacts on O₃ and PM₂.₅. Larger uncertainties related to GHG emissions scenario and climate sensitivity emerge by the end of the century. In all regions, uncertainty arising from GHG emissions is the largest in 2100. Across the simulations, the influence of climate sensitivity grows in response to GHG emissions, while the level of internal variability remains relatively consistent, rendering it the smallest uncertainty source at century end.

Figure 3 shows the progression of climate-related uncertainty in projections of U.S. air quality from middle to end of the 21st century. While the uncertainties in climate change impacts on O₃ and PM₂.₅ associated with different sources are comparable at mid-century, by end-century the spatial distributions of emissions scenario and climate sensitivity uncertainty resemble the patterns of climate-induced changes in pollutant concentrations. Although the projected climate impacts on PM₂.₅ are largely driven by enhanced SO₂ oxidation and nitrate partitioning to the gas phase under higher temperatures, as discussed in [15, 26], the spatial patterns of PM₂.₅ impacts and their uncertainty differ from those of
climate impacts on surface temperature. Uncertainty in PM$_{2.5}$ impacts is lowest in the Western U.S., where uncertainty in the temperature projections, estimated in [24], is greatest. High uncertainty in the Eastern U.S. is more consistent with reported uncertainty in projections of precipitation under climate change [24]. The simulated climate impacts on O$_3$, examined in [15, 25], are driven by competing positive associations with temperature and biogenic emissions and negative associations with humidity and wind speed. The largest projected impacts and uncertainties in climate-induced changes in U.S. O$_3$ concentrations are in the East and Midwest, while temperature is projected to increase most and be most uncertain in the West and Great Plains regions [24]. While uncertainty in future temperature and other weather variables propagates to projections of air quality, the complex relationships between pollutant concentrations, meteorological drivers, and emissions of pollutants and precursors make it difficult to infer the impacts and uncertainty in climate change impacts on air pollution based on projections of meteorology alone.

3.3. Influence of internal variability on projected air quality impacts

Internal variability, an irreducible uncertainty, propagates from climate simulations to projections of air quality. Most studies investigating the impact of climate change on air quality have relied on simulating time slices of ten or fewer years to account for internal variability when comparing present-day and future air quality [25]. However, sub-decadal simulations have proven insufficient to filter out the noise of internal variability in projections of climate impacts on O$_3$ and PM$_{2.5}$ and the associated health impacts [25, 26, 52, 53]. Although large ensembles offer an effective approach to capture internal variability in climate projections [19], few studies have used initial condition ensembles to simulate climate change impacts on air quality [23, 50, 51, 53]. Here, we rely on multiple model initializations and 30 year periods to represent a range of internal variability and weigh its influence on projected climate change impacts on pollutant concentrations. Figure 4 shows that uncertainty in projected impacts on mean O$_3$ and PM$_{2.5}$ concentrations for shorter time slices is significantly higher. The uncertainty associated with internal variability is approximately 2.5 times larger when projecting the end-century 15 year mean impact of climate change on population-weighted O$_3$ and PM$_{2.5}$ concentrations, relative to 30 year mean impacts. Projecting 5 year mean or single-year impacts leads to internal variability exceeding the uncertainties associated with emissions scenario or climate sensitivity throughout the entire 21st century. In projections of climate impacts on air quality, sufficient timescales or initial condition ensembles are needed to avoid internal variability exceeding other climate-related uncertainties.

3.4. Influence of climate sensitivity on projected air quality impacts

In this simulation ensemble, the uncertainty in projected climate change impacts on U.S. air quality...
associated with climate sensitivity is comparable to that stemming from emissions scenario and internal variability at mid-century and remains substantial throughout the 21st century. Figure 5 shows projected impacts at century end under the REF scenario and based on a climate sensitivity of 3.0 °C, the IPCC’s best estimate [44], and relative differences at lower and higher sensitivities. Climate sensitivity near the high end of the IPCC’s ‘very likely’ range, 4.5 °C, intensifies projected climate change impacts on annual O$_3$ and PM$_{2.5}$ concentrations by as much as 6 ppb and 2 µg m$^{-3}$, respectively, while climate sensitivity of 2.0 °C, at the low end of the ‘very likely’ range, diminishes them by more than 4 ppb and 2 µg m$^{-3}$, relative to the best-estimate sensitivity. Relative to a 3.0 °C climate sensitivity, projected REF-scenario population-weighted climate penalties on annual O$_3$ and PM$_{2.5}$ concentrations by as much as 6 ppb and 2 µg m$^{-3}$, respectively, while climate sensitivity of 2.0 °C, at the low end of the ‘very likely’ range, diminishes them by more than 4 ppb and 2 µg m$^{-3}$, relative to the best-estimate sensitivity. Relative to a 3.0 °C climate sensitivity, projected REF-scenario population-weighted climate penalties on annual O$_3$ and PM$_{2.5}$ concentrations at the end of the century increase by 2.5 ppb (78%) and 0.7 µg m$^{-3}$ (46%) under a 4.5 °C climate sensitivity, and decrease by 1.6 ppb (49%) and 0.8 µg m$^{-3}$ (55%) under a 2.0 °C climate sensitivity. Under the climate stabilization scenarios considered, differences in projected impacts across the range of climate sensitivities are significantly lower. Ensemble-average climate change impacts under all GHG emissions scenarios are included in table S1.

Uncertainties in climate impacts on air quality associated with emissions scenario, internal variability, and climate sensitivity are linked. Climate driven changes to meteorology that impact O$_3$ and PM$_{2.5}$ pollution depend on GHG emission levels, while the intensity of these impacts is determined in part by the earth system’s climate sensitivity. Internal variability adds noise to these forced signals in air quality. In this simulation ensemble, for example, we find that stronger climate sensitivity leads not only to a higher mean O$_3$ penalty over the U.S. by the end of the century, but also larger interannual spread in projected concentrations (figure 6). The increased level of internal variability at higher climate sensitivity suggests greater risks of extreme air pollution at the high ends of the distribution. For example, while the mean REF-scenario climate penalty on annual population-weighted O$_3$ concentration projected at century end under the highest climate sensitivity (4.5 °C) is approximately 4 ppb larger than that projected under the lowest climate sensitivity (2.0 °C), the 95th percentile year simulated among all initial conditions and 30 year periods is 5 ppb larger between the highest and lowest climate sensitivities. In contrast, internal variability in climate-induced changes to PM$_{2.5}$ remains consistent across the climate sensitivities explored. While several studies have reported a climate penalty on the upper tails of projected future air pollution distributions [5, 13, 23, 48, 54], further work investigating high pollution events can shed additional light on the interconnections between different sources of climate uncertainty in simulations of air quality impacts.
Figure 6. Interannual variability in climate change impacts on U.S. O$_3$ and PM$_{2.5}$ pollution at higher climate sensitivity. Distribution of single-year projected climate impacts on annual population-weighted O$_3$ (a) and PM$_{2.5}$ (b) ground-level concentrations at end-century, relative to the historical simulation, under the REF scenario and climate sensitivities of 2.0 °C, 3.0 °C, and 4.5 °C. Fitted normal distributions, means ($\mu$), and standard deviations ($\sigma$), are shown for each climate sensitivity considered.

4. Discussion

4.1. Outlook for impacts assessment in the face of uncertainty

Extensive research has projected significant impacts of climate change on O$_3$ and particulate matter pollution [3]. Here, we show that important uncertainties in these projections arise from recognized sources of uncertainty in climate simulations. For example, in our simulation ensemble we find that internal variability has the potential to far exceed GHG emissions scenario and climate sensitivity uncertainties throughout the 21st century if adequate timescales are not considered. We also observe that when considering uncertainty in climate sensitivity, a previously unexplored risk of climate-induced extreme O$_3$ pollution is revealed. Further propagation of these uncertainties to projections of pollution-related health damages and costs has implications for climate policy assessments [52]. More complete characterizations of uncertainty in climate-induced impacts on air pollution can aid decision-making.

Due to the high computational cost of atmospheric chemistry simulations, only three emissions scenarios and five model initializations, constrained by limited resolution and representations of atmospheric processes, are included in this analysis. Recent work has suggested that more initial conditions may be needed to fully capture internal climate variability [19]. Our finding that emissions scenario uncertainty is the largest climate-related uncertainty at the end of the century depends on the scenarios considered. While the analysis is based on an ensemble covering a wide range of climate policy and GHG emissions futures, including additional scenarios or accounting for scenario likelihood could reduce uncertainty in estimates of climate impacts on air pollution. By including a high-emissions REF scenario, projections with a large climate penalty on air quality in the ensemble strongly contribute to this uncertainty. The ensemble explored here is based on a single modeling framework and a cloud radiative adjustment method is used to project impacts under different climate sensitivities (see section 2). However, structural uncertainty can make the framework’s response to GHG concentrations differ from that in other models.

Models with more detailed chemical mechanisms may better capture atmospheric composition, but their computational costs currently render them challenging for the large ensembles required to assess uncertainty in climate change impacts. When coupled climate-chemistry simulations are not feasible, climate model ensembles offer opportunities to characterize variability in meteorological conditions conducive to air pollution, such as stagnation [55–58]. Additionally, the influence of air pollutant emissions, and thus atmospheric composition, on climate-induced impacts on air quality and their uncertainties remains largely unexplored. We rely on emissions at the beginning of the century to isolate climate change impacts and the associated uncertainty estimates are based on these emissions levels. However, pollutant and precursor emissions will change continuously during the 21st century [59], and ensembles like the one used here do not model the impacts of variations in air pollutant emissions. Analyses that consider changing air pollutant emissions (e.g. [53, 54]) can capture the effects of both climate and emissions, although separating these effects requires simulations in which radiative forcing or air pollutant emissions...
are fixed, and that inconsistencies arising from the dual role of some species as both conventional air pollutants and climate forcers be addressed. Despite these limitations, the ensemble examined here, encompassing over 2800 annual simulations of atmospheric chemistry, constitutes one of the largest modeling efforts to date investigating the impacts of climate on air pollution and allows for an unprecedented view into how three primary sources of uncertainty in climate modeling propagate to projections of air quality under climate change.

Considering its adverse effects on public health, including disease and premature mortality, a climate-induced deterioration of air quality would have major economic consequences. Larger uncertainty will likely increase the costs of strategies to mitigate or adapt to these impacts [18]. The potential to reduce uncertainty in projections of air quality under climate change differs among uncertainty sources. Reducing uncertainty arising from emissions of climate forcers requires a more certain climate policy outlook. Uncertainty associated with climate sensitivity can be diminished as representations of the earth system in models are improved. Internal variability uncertainty is not expected to decrease, but its effects on projections can be captured by large initial-condition simulation ensembles. Additionally, future air quality depends on uncertain emissions of air pollutants and precursors. In light of this complexity, large ensembles offer an approach to elucidate the impacts of climate change and anthropogenic emissions on air quality. Multi-model and large initial-condition ensembles, such as the Coupled Model Intercomparison Project and single model initial-condition large ensemble initiatives, have provided important insights into climate projections [19]. Climate impacts on air quality have not yet been explored with ensembles at this scale. Although the analysis presented here is based on a smaller-scale simulation ensemble, it shows the opportunities large ensembles generated with fully coupled climate-chemistry models would offer to investigate uncertainty in the impacts of both climate change and anthropogenic precursor emissions on atmospheric composition. Such an advance would improve co-benefits assessments and inform climate policy design in light of the uncertainties presented here.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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