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Key Points:
- Natural variability can significantly influence model-based projections of climate change impacts on air quality
- Multidecadal simulations or initial condition ensembles are needed to identify an anthropogenic-forced climate signal in O_3 concentrations
- It is difficult to attribute the impacts of climate change on O_3 to human influence before midcentury or under stabilization scenarios

Supporting Information:
- Supporting Information S1

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The role of natural variability in projections of climate change impacts on U.S. ozone pollution

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Abstract Climate change can impact air quality by altering the atmospheric conditions that determine pollutant concentrations. Over large regions of the U.S., projected changes in climate are expected to favor formation of ground-level ozone and aggravate associated health effects. However, modeling studies exploring air quality-climate interactions have often overlooked the role of natural variability, a major source of uncertainty in projections. Here we use the largest ensemble simulation of climate-induced changes in air quality generated to date to assess its influence on estimates of climate change impacts on U.S. ozone. We find that natural variability can significantly alter the robustness of projections of the future climate's effect on ozone pollution. In this study, a 15 year simulation length minimum is required to identify a distinct anthropogenic-forced signal. Therefore, we suggest that studies assessing air quality impacts use multidecadal simulations or initial condition ensembles. With natural variability, impacts attributable to climate may be difficult to discern before midcentury or under stabilization scenarios.

1. Introduction

Changes in climate may lead to changes in ozone (O_3) pollution, and associated health and environmental impacts, by altering atmospheric chemistry and transport [*Fiore et al.*, 2012; *Jacob and Winner*, 2009; *Kirtman et al.*, 2013]. Meteorological conditions under a warmer climate may exacerbate the public health burden related to ground-level O_3 and make regulatory standards harder to meet. In the U.S., changes in climate are expected to worsen O_3 pollution, aggravating premature mortality, acute respiratory symptoms, and other detrimental health effects [*Bell et al.*, 2007; *Fann et al.*, 2016; *Fiore et al.*, 2015; *Post et al.*, 2012; *United States Environmental Protection Agency (U.S. EPA)*, 2014]. Exposure to O_3 can also have damaging impacts on terrestrial ecosystems, including crops, pastures, and forests, leading to negative consequences for global and regional economies [*Felzer et al.*, 2007; *Reilly et al.*, 2007; *Tai et al.*, 2014]. Furthermore, tropospheric O_3 is a significant contributor to the global radiative forcing of climate [*Kirtman et al.*, 2013]. The impacts of climate-induced changes to global and regional O_3 pollution have been explored by numerous modeling studies using different climate change projections. However, large underlying uncertainties in climate simulations propagate to the estimates of future air quality generated by these efforts. It is important that impact projections derived from climate-air quality modeling be placed in the context of this uncertainty.

Climate projections are influenced by three key sources of uncertainty: emissions scenario, model response, and natural variability [*Hawkins and Sutton*, 2009]. In contrast to emissions scenario and model response, uncertainty associated with natural climate variability (i.e., unforced internal variability, which stems from the chaotic nature of the simulated climate system) is not expected to diminish as models and emissions projections improve [*Deser et al.*, 2012a]. Recently, large ensemble simulations have allowed a closer examination of natural variability [*Deser et al.*, 2012b; *Kay et al.*, 2015; *Monier et al.*, 2015; *Sriver et al.*, 2015]. Its role has been investigated in projections of different climate change impacts, including sea-level rise [*Bordbar et al.*, 2015], sea ice loss [*Swart et al.*, 2015], agriculture [*Cohn et al.*, 2016], and extreme weather [*Fischer et al.*, 2013]. However, for air quality applications, high computational costs have limited many studies based on chemical transport models or coupled global chemistry-climate models to a small sample of years, seasons, or months. Figure 1 shows simulation lengths used by 41 studies modeling climate change impacts on U.S. O_3 pollution. Of these, 29 relied on 5 years or less of simulated climate-air quality; only 6 used more than 10 years. Distinct from the studies included in Figure 1, *Barnes et al.* [2016] simulate

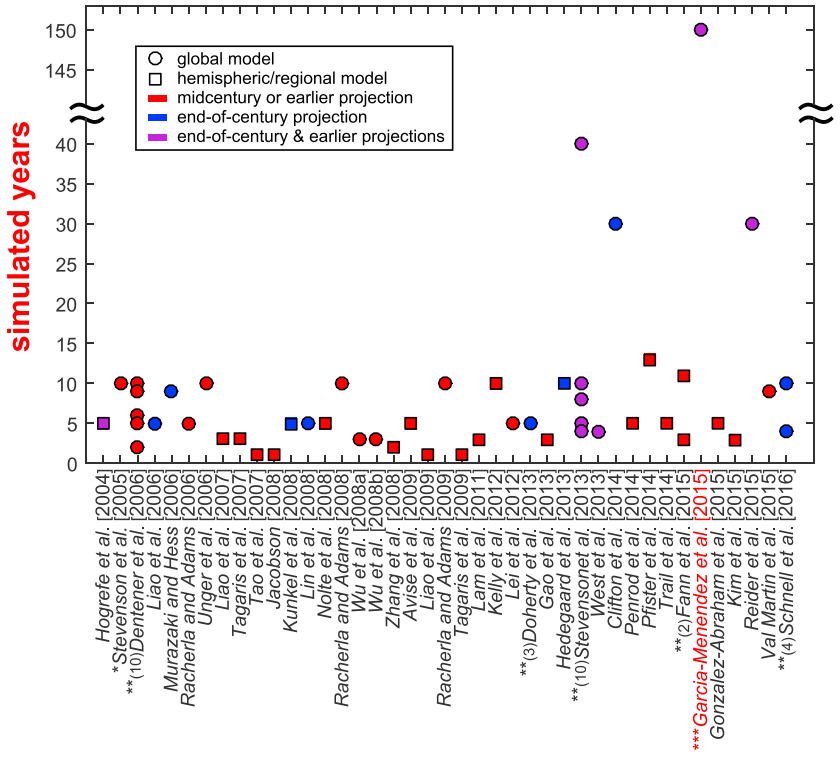

Figure 1. The number of years used in 41 modeling studies to characterize present/future air quality and estimate the impact of climate change on U.S. O_3 pollution. Circles and squares specify use of global or regional/hemispheric models. Colors indicate studies projecting end-of-century impacts, midcentury or earlier impacts, or both. **Stevenson et al.* [2005] simulate climate/air quality from 1990 to 2030 but use decadal averages to estimate climate impacts. **These studies include multiple models which may use different numbers of simulated years; the number of models is included in brackets. ***Ensemble used for the analysis in this article.

climate/air quality from 2006 to 2055 using three ensemble members to remove internal variability and compare the ensemble-mean trend in surface O_3 to that obtained from a control simulation (200 years) in which all emissions and greenhouse gas concentrations are fixed at 1990 levels. A concern arising from some of these analyses is that simulations used to contrast present and future climates may be too short to effectively differentiate an anthropogenic-forced impact from internal variability [*Fiore et al.*, 2015; *Nolte et al.*, 2008]. The extent to which climate-induced changes may be confounded depends on the air quality metric, area, and period evaluated [*Barnes et al.*, 2016; *Fiore et al.*, 2015]. Drawing conclusions from a small sample size may be premature, particularly at a regional scale, few decades into the future, or under a stabilization scenario. Natural variability, for instance, has been shown to influence U.S. temperature and precipitation projections on timescales as long as 50 years [*Deser et al.*, 2014; *Monier et al.*, 2015].

To examine the role of natural variability in estimates of climate impacts on O_3 pollution, we simulate air quality based on an ensemble of integrated economic and climate projections of the 21st century generated using the Massachusetts Institute of Technology Integrated Global System Model. The ensemble serves as the basis for the U.S. Environmental Protection Agency's (EPA) Climate Change Impacts and Risks Analysis (CIRA) project, which quantifies impacts across a wide range of sectors [*U.S. EPA*, 2015]. Multidecadal global atmospheric chemistry simulations and subsets of model initializations are used to investigate natural variability including both internally generated climate variability and natural variability forced by year-to-year variations in solar forcing, as well as interactions between externally forced and internally generated components, such as changes in natural emissions of greenhouse gases (i.e., CO_2 from terrestrial ecosystems, CH_4 from wetlands, or N_2O from unfertilized soils). In total, 1050 years of atmospheric chemistry are simulated, making this the largest modeling effort to date specifically investigating climate-induced changes to air quality.

2. Methods

A modeling framework that links the Massachusetts Institute of Technology Integrated Global System Model to the Community Atmosphere Model (MIT IGSM-CAM version 1.0) [*Monier et al.*, 2013] was used to generate an ensemble of integrated economic and climate projections. The Economic Projection and Policy Analysis model is a multisector, multiregion computable general equilibrium model used within the MIT IGSM to project economic activity and associated climate-relevant gas and aerosol emissions under policy constraints [*Paltsev et al.*, 2005]. Climate-relevant emissions related to fuel combustion, agricultural activity, industrial processes, and waste handling are used to simulate climate in the IGSM's Earth system component. Climate policy is integrated by implementing a uniform global tax on greenhouse emissions required to achieve a total radiative forcing target. The MIT IGSM's Earth system component is a model of intermediate complexity that comprises the atmosphere, ocean, sea ice, carbon and nitrogen cycles, and terrestrial water, energy, and ecosystem processes. In the IGSM-CAM framework, the Community Atmosphere Model (version 3) is used to produce three-dimensional climate fields at $2^\circ \times 2.5^\circ$ resolution and a height of approximately 40 km using 26 vertical layers. The IGSM-CAM also allows climate model response to be modified by altering climate sensitivity through a cloud radiative adjustment method [*Sokolov and Monier*, 2012]. A climate sensitivity of 3°C was used for the ensemble simulation described in this study. Climate simulations were carried out under five different representations of natural variability. Each representation was generated by perturbing the model's initial atmospheric and land conditions and the ocean surface forcing [*Monier et al.*, 2013].

Three integrated climate policy and greenhouse gas emissions scenarios are included, a no-policy reference scenario (REF) and two climate stabilization scenarios with 2100 total radiative forcing targets of 4.5 and 3.7 $W\,m^{-2}$ (P45 and P37). Global CO_2 concentration reaches $>$800 ppm in 2100 under the reference scenario but is constrained to $<$500 ppm in the stabilization scenarios. Global mean surface temperature is projected to increase by 6°C at the end of the century in the absence of climate policy and limited to $<$1.5°C under greenhouse gas mitigation. The REF scenario is comparable to RCP8.5 from the Representative Concentration Pathway (RCP) scenarios, while the P45 scenario is similar to the RCP4.5 scenario, and the P37 scenario falls between the RCP2.6 and RCP4.5 scenarios. A more detailed comparison to the RCP and Special Report on Emissions Scenarios is included in *Paltsev et al.* [2015]. In addition, a comparison between the IGSM-CAM climate projections and a multimodel ensemble based on the Coupled Model Intercomparison Project phase 5 under the RCP8.5 and RCP4.5 scenarios shows similar changes projected for several climate variables [*Monier et al.*, 2016]. The CIRA emissions scenarios and their associated climate projections have been used to quantify risks and benefits of climate policy across several U.S. sectors, including water resources, electricity, infrastructure, health, agriculture and forestry, and ecosystems [*U.S. EPA*, 2015]. Additional information about the socioeconomic and climate change scenarios is available in *Paltsev et al.* [2015], and details further describing climate projections for the U.S. can be found in *Monier et al.* [2015].

The influence of natural variability on projections of climate change impacts on O_3 pollution in the U.S. was assessed by simulating global atmospheric chemistry under the scenarios developed for EPA's CIRA project. Meteorological fields derived from the MIT IGSM-CAM for each climate scenario and realization were archived at 6 h intervals and regridded using a bilinear interpolation method to drive offline simulations of the Community Atmosphere Model with atmospheric chemistry (CAM-Chem version 4) [*Lamarque et al.*, 2012]. CAM-Chem simulates over 100 gas and aerosol species, including ground-level O_3, and was run at $1.9^\circ \times 2.5^\circ$ resolution with 26 vertical layers extending into the lower stratosphere (approximately 40 km). Model performance for surface O_3 has previously been evaluated against ground-based observations across the U.S. [*Brown-Steiner et al.*, 2015; *Lamarque et al.*, 2012; *Tilmes et al.*, 2015]. Anthropogenic emissions, mostly drawn from the Precursors of Ozone and their Effects in the Troposphere database, are described in *Lamarque et al.* [2012]. Anthropogenic emissions are held constant at start-of-century (year 2000) levels in the CAM-Chem simulations, including emissions of greenhouse gases with dual roles as short-lived climate forcers and O_3 precursors (e.g., methane), to isolate the effect of climate change on O_3 concentrations. This baseline level of emissions is comparable to that used by other studies quantifying climate-induced impacts on O_3, including the Atmospheric Chemistry and Climate Model Intercomparison Project [*Stevenson et al.*, 2013]. Within CAM-Chem, biogenic emissions of isoprene and monoterpenes, as well as

their response to changing temperature, are simulated using the Model of Emissions of Gases and Aerosols (MEGAN2) and the Community Land Model (CLM3) [*Lamarque et al.*, 2012]. The optimized dry deposition scheme developed by *Val Martin et al.* [2014] was included in all simulations. Climate-induced changes to several natural emissions sources, including lightning, soils, wetlands, and wildfires, are not modeled within CAM-Chem.

The impact of climate change on U.S. O_3 concentrations was estimated as the difference between atmospheric chemistry simulations under present and future climates. Thirty year simulations were used to characterize air quality at the start of the century (1981–2010) and, for each climate policy scenario, the middle (2036–2065), and end of the century (2086–2115). Additionally, CAM-Chem simulations were run for each of the IGSM-CAM's five representations of natural variability. Ultimately, 150 years of modeled air quality (five sets of 30 year simulations) were generated for each timeframe under each policy scenario. Two dimensions of natural variability are considered: (1) interannual variability by analyzing continuous 30 year simulations and (2) multidecadal variability by contrasting simulations with different initial conditions. Here we weigh the impacts of climate change on 50 and 100 year timescales against interannual variability on timescales under 30 years. Therefore, to account for externally forced climate change during each 30 year period and center on air quality impacts in 2050 and 2100, concentrations simulated in each model run were detrended by computing the least squares fit of a straight line for the region of interest and subtracting the function from the data. The statistical significance of ensemble-mean concentration changes is evaluated through a Student's *t* test for a 95% confidence level. The confidence interval at 95% for the difference in means is used to represent the range of reported concentration changes. Margin of error estimates for reported climate impacts assume sample standard deviations ($n \geq 30$) represent the population standard deviation. Population-weighted concentrations based on present-day U.S. population distribution [*Consortium for International Earth Science Information et al.*, 2005] are used to represent O_3 pollution. U.S. regions used for regional estimates correspond to those defined in the 2014 U.S. National Climate Assessment [*United States Global Change Research Program*, 2014].

3. Results and Discussion

Projected climate impacts on U.S. ground-level O_3 are significant and differ among regions. Ensemble-mean estimates of climate-induced changes are shown in Figure S1 in the supporting information. At national scale, simulated ensemble-mean climatic influences on annual-average population-weighted daily maximum 8 h O_3 (8 h max O_3) are $+0.8 \pm 0.3$ and $+3.2 \pm 0.3$ ppb_v in 2050 and 2100 under the no-policy REF scenario. The influence of climate change on O_3 increase is largest over the Northeast and Southeast, while a climate-induced reduction is projected over large areas in the Midwest and West. Under greenhouse gas mitigation scenarios, the effect of climate change is diminished. Health and economic consequences have been previously estimated for these ensemble-mean impacts [*Garcia-Menendez et al.*, 2015].

Multiyear model runs can be used to account for natural variability and capture the anthropogenic-forced signal in simulated air quality. A comparison of climate impact projections estimated from single-year simulations reveals a considerable amount of interannual variability in the ensemble. Figure 2 shows end-of-century climate influence on O_3 season (May–September) concentrations for individual years over a 30 year period. While only 30 consecutive pairwise combinations for a single climate model initialization are depicted, the influence of natural variability is evident. Estimating the 2100 REF scenario influence on annual population-weighted 8 h max O_3 from years that atypically favor or hamper O_3 formation can lead projections higher than $+4$ ppb_v or less than -2 ppb_v. In locations throughout the eastern and southern U.S., single-year projections for annual-average concentrations can show different directions of change (supporting information Figure S2). During O_3 season the national-average impact is consistently an increase in O_3 pollution, although variability is larger than for the annual-average concentration. Large seasonal variability could be especially relevant to health impacts and attainment of regulatory standards.

Ensembles of simulations in which all forcings remain unchanged but initial conditions are perturbed can reproduce the internal variability inherent to a model [*Xie et al.*, 2015]. Figure 3 shows the effect initial conditions can have on different metrics weighing the impact on O_3 pollution. Within the set of five model initializations, projections estimated from short simulations can differ substantially, including the sign of change. However, as simulation length is extended, mean projections display a tendency to converge to

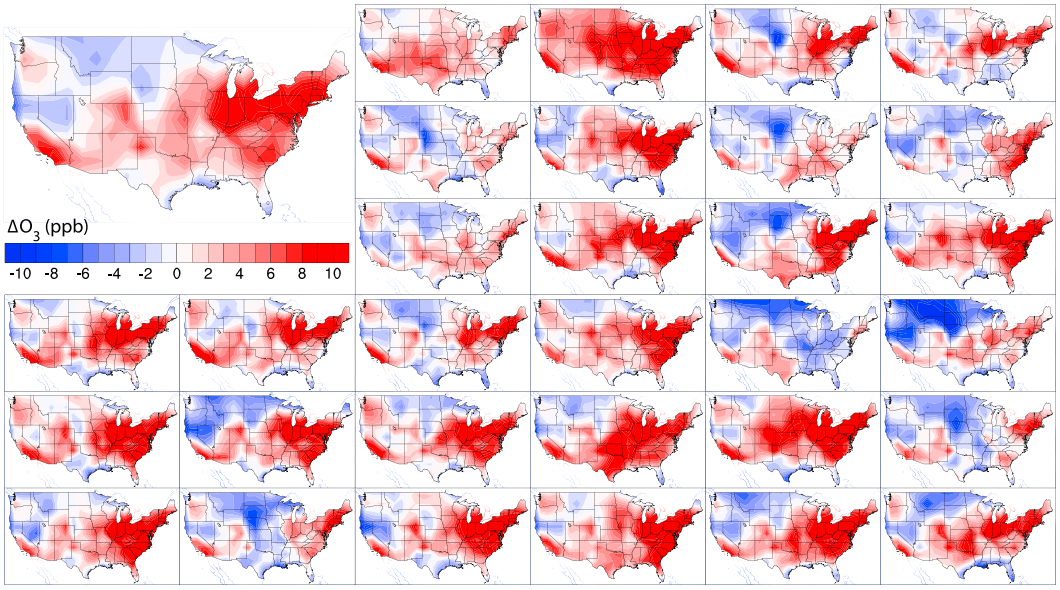

Figure 2. Interannual variability in projected climate impact. Mean 2100 reference scenario climate impact (2085–2115 mean relative to 1981–2010 mean) on O_3 season (May–September) ground-level 8 h max O_3 estimated from a single climate model initialization and 30 year present and future simulations (top left panel). Projections estimated from each 1 year present/future simulation pair are shown in 30 smaller panels to exemplify the variability contained within the mean projection.

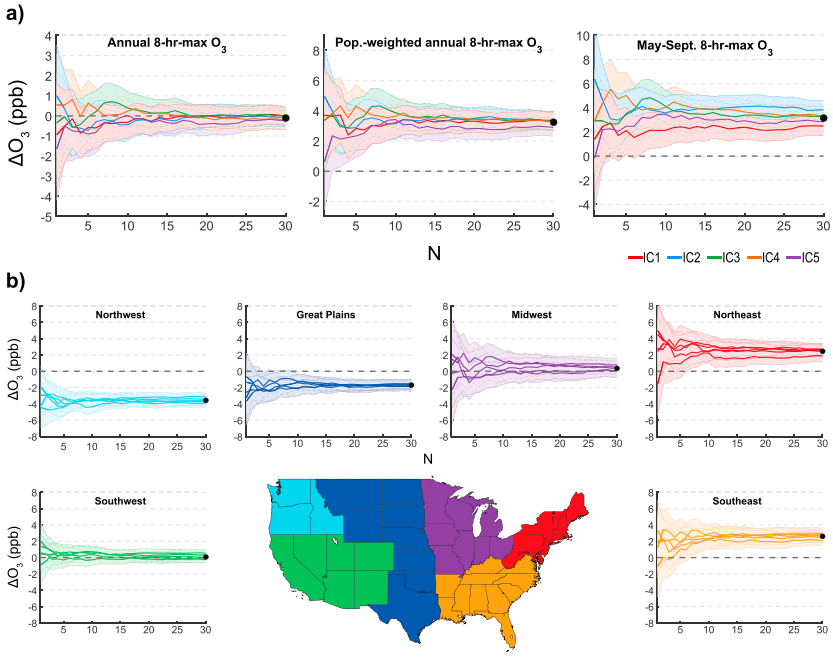

Figure 3. Anthropogenic-forced impacts on O_3 pollution. (a) 2100 reference scenario U.S.-average climate impact on annual-average, population-weighted, and ozone season ground-level 8 h max O_3 estimated using 5 climate model initializations (denoted by different colors) and simulation lengths increasing from 1 to 30 years. (b) The 2100 reference scenario regional-average climate change impact on annual-average ground-level 8 h max O_3 estimated using five climate model initializations (denoted by different lines) and simulation lengths increasing from 1 to 30 years. Shaded regions indicate the margin of error for each estimate at a 95% confidence level based on the 30 year sample standard deviation. Black dots indicate ensemble-mean projections.

the same anthropogenic-forced signal. Estimating the 2100 REF scenario climate influence on annual U.S.-average 8 h max O_3 from 5 year simulations can lead to projections of increases or decreases greater that ±0.5 ppb_v, depending on the initialization. Despite substantive regional differences, the 150 year ensemble-mean value is in fact close to zero (-0.1 ± 0.2 ppb_v). Simulations under all initializations project an increase in population-weighted concentrations, although with greater variability than unweighted estimates. The variability in O_3 season impacts is larger still.

At finer scale, the ensemble reveals clear differences among U.S. regions (Figure 3b). The northeast and southeast show the highest climate-induced O_3 increases, along with large interannual variability. Natural variability is largest in the Midwest, while the direction of climate-induced change remains uncertain. Variations are significantly smaller over the western U.S. As for national estimates, the robustness of regional impacts projected by a single model run, assessed by comparing across initial conditions, strengthens as number of years simulated is increased. It is important to note that while extended simulation lengths or ensembles increase the analysis's range of possible futures, in reality a single climate realization will occur. However, better accounting for natural variability provides a clearer image of true anthropogenic climate change impacts and useful information for future attribution.

For model-based assessments of O_3 impacts, adequate simulation size depends on the strength of the climate change signal and precision required. In this ensemble, achieving a ±1.0 ppb_v margin of error for the projected 2100 REF scenario change in U.S.-average annual 8 h max O_3 at a 95% confidence level entails using seven simulation years. Estimating the population-weighted O_3 change with the same margin of error requires an 11 year sample. At regional scale, this level of confidence comes at higher computational cost: 14 years or more are needed for Midwest, northeast, and southeast estimates. A ±1.0 ppb_v margin of error may be adequate for metrics or regions exhibiting strong signals but may be insufficient to confidently project smaller externally forced impacts. Lowering the margin of error to ±0.5 ppb_v requires considerably larger sample sizes (Table S1 in the supporting information includes additional values, as well as the ensemble-mean surface O_3 and temperature changes). In comparison, the magnitude of U.S. climate change impacts estimated by previous modeling studies can be a few parts per billion or smaller [*Fiore et al*., 2015].

While climate-induced change in U.S. O_3 pollution is apparent in end-of-century projections, a midcentury anthropogenic-forced signal is not nearly as evident. *Barnes et al.* [2016] suggest that O_3 changes driven by the meteorological response to climate change may not emerge before 2050. Figure 4 shows population-weighted 8 h max O_3 from each of the 150 annual REF scenario simulations representing the start, middle, and end of the century. Distributions for each period are also compared. Detecting a mean difference between start-of-century and end-of-century values equal to the ensemble-mean projected change using a paired t test ($\alpha = .05$, $\beta = .2$) requires a sample size of five present/future year pairings. However, when comparing values representative of 2000 and 2050 climates, the required sample size is greater than 35. The impact of greenhouse gas mitigation is also shown in Figure 4. Under a stabilization scenario targeting 4.5 W m^{-2} total radiative forcing by 2100, the distribution of 2086–2115 population-weighted concentrations remains close to that of 1981–2010 values. The direction of climate-induced change is much more uncertain under policy; only 57% of present/future annual simulation pairings project an O_3 pollution increase by the end of the century. Further increasing policy stringency to stabilize radiative forcing at 3.7 W m^{-2} leads to only small changes in concentration mean and distribution.

Under a warmer climate, O_3 concentrations are affected through multiple coupled pathways. The mechanisms through which climate change impacts ground-level O_3 in our simulations are further discussed in *Garcia-Menendez et al.* [2015]. In the ensemble, we observe correlations between meteorological variables and ground-level O_3 similar to those reported by prior studies: positive correlations with near-surface air temperature and biogenic isoprene emissions and negative correlations with near-surface humidity and wind speed [*Dawson et al*., 2007; *Doherty et al*., 2013; *Weaver et al*., 2009]. Under unchanging anthropogenic emissions of O_3 precursors, these relationships between O_3 and meteorology drive the variability and climate-induced change in concentrations. Competing and coupled interactions also imply that robust projections for individual meteorological variables do not denote a clear anthropogenic-forced signal in simulated O_3 pollution. Surface temperature, for instance, shows a significant correlation with annual U.S.-average 8 h max O_3 for population-weighted concentrations ($r = .75$, $p < .001$), as well as deviations

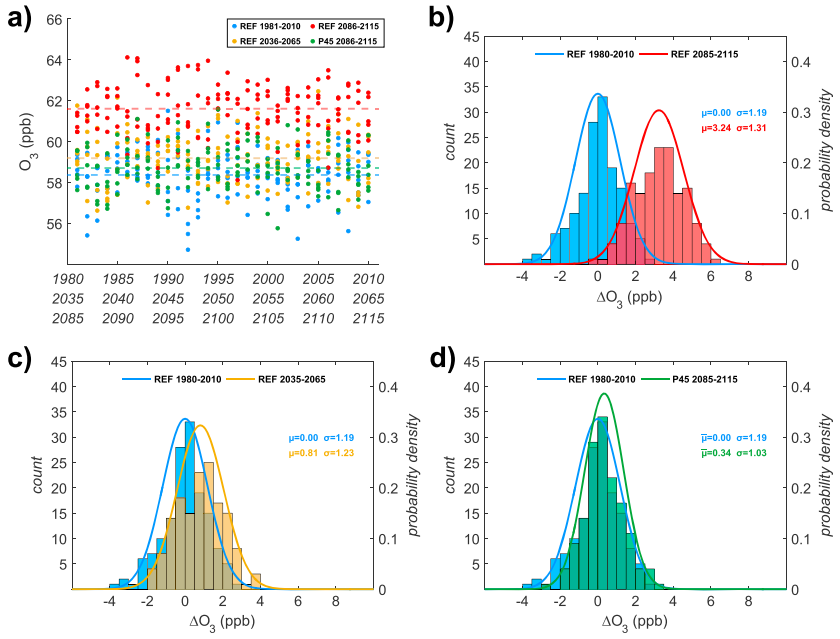

Figure 4. Range of simulated O_3 pollution. (a) U.S.-average annual population-weighted 8 h max O_3 estimated for each 1 year simulation and five climate model initializations under the reference (REF) scenario during the 1981–2010, 2036–205, and 2086–2115 periods, and a policy scenario targeting 4.5 W m^{-2} total radiative forcing by 2100 (P45) during the 2086–2115 period. Dashed lines indicate ensemble-mean estimates. (b–d) Distribution of anomalies from 1981 to 2010 mean for each future period included compared to the start-of-the-century distribution. Fitted normal distributions, including their mean (μ) and standard deviation (σ), are shown for each period.

from ensemble means ($r = .59$, $p < .001$). The year-to-year variability in U.S. temperature has been attributed to internal atmospheric circulation variability [*Deser et al.*, 2016], largely associated with teleconnections and modes of variability including the Pacific/North American teleconnection pattern [*Leathers et al.*, 1991; *Ning and Bradley*, 2016] and El Niño–Southern Oscillation [*Ropelewski and Halpert*, 1986]. However, the projection of an externally forced increase is considerably more robust for surface air temperatures than O_3; comparing the distributions of simulated REF scenario temperatures (shown in supporting information Figure S3) reveals a higher likelihood of midcentury and end-of-century anthropogenic-induced change.

4. Conclusions

Our ensemble-mean projections agree with prior modeling analyses anticipating climate-induced O_3 increases over polluted U.S. regions, including the northeast, Midwest, southeast, and California [*Fiore et al.*, 2015]. However, there are also inconsistencies among the regional impacts reported by different studies [*Jacob and Winner*, 2009; *Weaver et al.*, 2009]. These discrepancies may be in part a function of the limited number of years simulated in most studies to date. In addition, similar to most modeling efforts attempting to quantify the impacts of climate change on air quality, we rely on a single climate model. The response to climate forcing and internally generated variability is model dependent. Atmospheric chemistry multimodel ensembles can capture additional natural variability if it differs among the models included, potentially reducing required simulation lengths. However, multimodel ensembles also encompass model response uncertainty arising from structural differences among models, making it more difficult to discern between the two sources of uncertainty and quantify the influence of natural variability specifically, in the context of O_3 modeling. Multimodel ensemble projections may also make it difficult to explore the mechanisms through which climate impacts O_3 pollution; analyses investigating climate-air quality feedbacks in-depth typically examine the simulations from a single model. Although prior studies have used fully coupled chemistry-climate models as well as modeling frameworks without two-way interactions, natural variability influences estimates of climate-induced air quality generated with either approach.

Based on our ensemble, we recommend using a 15 year minimum for simulated climate and air quality to evaluate anthropogenic-induced impacts. The recommendation is derived from the simulations carried out in this study, using a single modeling framework and specific scenarios. It is intended to provide basic guidance to air quality modelers and encourage others to undertake similar analysis and evaluate the robustness of our results. Typically, however, the degree of variability in projections will not be known a priori and cannot be determined from a small sample. Should the resulting signal be weak, caution should be exercised and extended modeling may be necessary before attributing changes to human influence. Furthermore, relying on simulation length to discern a forced signal in sequential simulations may overlook longer-term multidecadal variability. Initial condition ensembles may be better suited toward this end.

Recent policy analyses have included climate change impacts on air pollution and associated health and economic consequences [*Fann et al*., 2015; *Garcia-Menendez et al*., 2015; *West et al*., 2013]. It is important that natural variability is appropriately considered in these studies' central modeling. While the significance of natural variability has recently come to light for heat-related mortality [*Shi et al*., 2015], most air quality studies fail to assess its role and carefully account for its influence on estimates of climate-induced air quality impacts. Large year-to-year variations in the climate system resulting from natural variability can be associated with severe air pollution episodes and related health effects. For the climate ensemble used in this study, an important effect of natural variability in the projections of climate change impacts on meteorological extremes has been demonstrated [*Monier and Gao*, 2015]. Given the interactions between meteorological variables and ground-level O_3 noted above, natural variability can be expected to have a significant influence on projections of climate-induced impacts on extreme air quality events. As a consequence of the high value placed on human life, these impacts have major implications for climate change mitigation analyses; avoided air quality health damages can be the largest component of climate policy benefits assessments [*U.S. EPA*, 2015]. Although broader modeling efforts may better capture the range of potential air quality impacts, only one future outcome will ensue in the real climate system. Labeling causes of change requires information on their underlying distributions. Recognizing the limitations imposed on climate-air quality projections by natural variability will allow more realistic attribution of climate change impacts and better inform decision making.

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