Global Air Quality and Climate

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March 2012, submitted to Chemical Society Reviews
Abstract

Emissions of air pollutants and their precursors determine regional air quality and can alter climate. Climate change can perturb the long-range transport, chemical processing, and local meteorology that influence air pollution buildup. We review the implications of projected changes in methane (CH$_4$), ozone (O$_3$), and aerosols for climate and hemispheric-to-continental scale air quality. Reducing the O$_3$ precursor CH$_4$ would slow near-term warming by decreasing both CH$_4$ and tropospheric O$_3$. Uncertainty remains as to the net climate forcing from anthropogenic nitrogen oxide (NO$_x$) emissions, which increase O$_3$ (warming) but decrease CH$_4$ (cooling). Anthropogenic emissions of carbon monoxide (CO) and non-CH$_4$ volatile organic compounds (NMVOC) warm by increasing both O$_3$ and CH$_4$; radiative impacts from secondary organic aerosol (SOA) are poorly understood. Strong absorption of sunlight by black carbon (BC) in the atmosphere or on snow and ice implies that BC emission reductions would slow near-term warming, but uncertainties in coincident emissions of reflective (cooling) aerosols and poorly constrained cloud indirect effects confound robust estimates of net climate impacts. Reducing sulfate and nitrate aerosols would improve air quality and lessen interference with the hydrologic cycle, but lead to warming. A holistic and balanced view is thus needed to assess how air pollution controls influence climate. On 50-100 year timescales, the anthropogenic power and industry sectors contribute most to warming; of the transportation sub-sectors, road transportation contributes most to warming, while shipping cools. Modeling and observational analyses suggest a warming climate degrades air quality (surface O$_3$ and particulate matter) in many populated regions, including during pollution episodes. The Representative Concentration Pathway emission scenarios (RCPs), developed for the next Intergovernmental Panel on Climate Change (IPCC) report, assume a uniform reduction in pollutant emission factors with increasing GDP and thus imply improved air quality relative to older IPCC scenarios (SRES), which projected unconstrained growth in air pollutant emissions. We find that uncertainty in emission-driven changes is generally wider than climate-driven changes. Confidence in air quality projections is limited by the reliability of anthropogenic emission trajectories and the uncertainties in regional climate responses, feedbacks with the terrestrial biosphere, and oxidation pathways affecting O$_3$ and SOA.

1. Introduction

Air pollutants and their precursors can force the climate system by altering solar and terrestrial radiation budgets, and their distributions are in turn highly dependent upon regional climate. We focus here on two major global air pollutants, ozone (O$_3$) and aerosol (referred to interchangeably with particulate matter below) in surface air. Increases in surface O$_3$ and particulate matter (with diameter $<$ 2.5$\mu$m, hereafter PM$_{2.5}$) driven by human activities since the pre-industrial have been blamed for
0.7 ± 0.3 million annual respiratory mortalities and 3.5 ± 0.9 million annual cardiopulmonary mortalities, respectively. When considering strategies to abate air pollution and mitigate climate change, policymakers face tradeoffs and synergies. For example, sulfate is a major component of PM$_{2.5}$ pollution in many regions, but reducing sulfate for health reasons could lead to a rapid rise in surface temperatures (e.g.,), possibly at rates threatening the survival of some ecosystems. In the absence of emission changes, a warming climate may degrade air quality in many polluted regions, an impact that has been referred to as “a climate change penalty” on air quality. Our review of the current understanding of the interactions between air pollutants and climate aims to identify robust conclusions that may guide decision-making and to highlight critical knowledge gaps.

Any successful climate mitigation effort must address carbon dioxide (CO$_2$). The relatively short lifetimes of other radiatively active air pollutants and their precursors (days to weeks for O$_3$ and aerosols, and approximately a decade for methane) implies that changes in their atmospheric abundances could induce rapid climate responses in the next few decades. These species have been referred to as “Short-Lived Climate Forcers” (SLCFs) and the studies reviewed here compare climate impacts of different SLCFs by estimating radiative forcing (RF). RF is a metric defined by the Intergovernmental Panel on Climate Change (IPCC) as the change in net radiative flux at the tropopause, after allowing stratospheric temperatures to adjust (note that other RF estimates allow other fast time scale adjustments), induced by a change in atmospheric abundance or distribution of a radiatively active species, typically expressed for present day relative to the pre-industrial (e.g., 1750). The equilibrium global mean surface temperature response corresponds to the annual globally averaged RF after accounting for the climate sensitivity, which encompasses the feedbacks in the climate system. Determining relative climate impacts from different species by comparing RFs assumes that climate responses (e.g., temperature, precipitation, circulation changes) scale accordingly. This assumption is not well-suited to the short-lived, heterogeneous distributions of air pollutants and their precursors (except for the well-mixed greenhouse gas CH$_4$) which vary in their forcing efficacy. A major advance in recent years is the incorporation of interactive chemistry into general circulation models (GCMs), which allows for the direct examination of climate responses (e.g., temperature, precipitation, circulation changes) to air pollutants and their precursors, and some chemistry-climate feedbacks. Below, we review this newer work as well as studies considering only RF from air pollutants.

The interactions between air pollutants and climate are often studied in the context of future emission scenarios. For projecting the future evolution of air quality under climate change, the most widely used scenarios to date are those described in the Special Report on Emission Scenarios (SRES) emission trajectories. A newly developed set of
scenarios, the Representative Concentration Pathways (RCPs), is widely used for ongoing international, multi-model activities in support of the upcoming IPCC Fifth Assessment Report (AR5). The RCP scenarios are described in detail and contrasted with SRES scenarios in several recent publications\textsuperscript{15-18}. All RCPs assume aggressive air pollution abatement measures (Figure 1) and correspondingly large decreases in PM and O\(_3\) precursors globally\textsuperscript{17,19}. Exceptions include ammonia (NH\(_3\)), which increases in nearly all scenarios\textsuperscript{19} and CH\(_4\), which ranges from a 30% decrease to a more than doubling by 2100 (Figure 1). We emphasize that the assumption of aggressive air pollution measures implemented globally is a major caveat in the RCP-based projections, as the small range of possible air pollutant emission trajectories across the RCPs may not represent the true uncertainty in emission pathways.

In Section 2, we review the literature on climate responses to emissions of air pollutants and their relevant precursors from anthropogenic sources and the terrestrial biosphere. Section 3 focuses on the processes by which changes in climate may influence air quality. Section 4 reviews projections for air quality over the next century, including changes driven by climate and emissions under a suite of scenarios. We end in Section 5 by discussing critical uncertainties and promising new directions targeting these knowledge gaps.

2. Sensitivity of climate to changes in air pollutant emissions

We review the literature describing climate responses to changes in emissions of O\(_3\) precursors (Section 2.1), and aerosols and their precursors (Section 2.2). Understanding the climate responses to the particular mix of pollutant sources from individual anthropogenic emission sectors (Section 2.3) is particularly relevant for policy decisions targeting controls on specific human activities. We briefly highlight the potential for emissions from the terrestrial biosphere to respond to changing pollution levels and climate, and thereby further change air quality (Section 2.4). Biospheric feedbacks to climate have been addressed recently\textsuperscript{20}.

2.1 Ozone (O\(_3\)) precursors

The increase in tropospheric O\(_3\) from pre-industrial to present-day driven by human activities\textsuperscript{21} has led to a positive climate forcing ($+0.35^{+0.3}_{-0.1}$ Wm\(^{-2}\))\textsuperscript{10} and substantial increases in O\(_3\) RF are estimated for the next century with the SRES scenarios\textsuperscript{22}. We discuss below the role of specific O\(_3\) precursor emissions, with a strong emphasis on the role of CH\(_4\), which has received growing attention in recent years as a means to address jointly air pollution and climate goals\textsuperscript{3,23-25}. The dominant CH\(_4\) sink occurs through oxidation by the hydroxyl radical (OH) in the troposphere, which leads to a unique
sensitivity of this greenhouse gas to air pollutants and their precursors. We also note the potential for oxidant changes induced by changes in O₃ precursor emissions to alter aerosol burdens (and aerosol burdens to alter O₃ burdens) with additional radiative implications.

Nitrogen Oxides (NOₓ). Increases in NOₓ (NO + NO₂) raise tropospheric O₃ levels (e.g., 29), thereby exerting a positive climate forcing. This positive forcing may be offset by an accompanying negative forcing due to lower CH₄ abundances because increasing NOₓ enhances tropospheric OH, and reaction with OH is the primary CH₄ loss pathway in the atmosphere. The net RF of these opposing influences on the climate system depends upon the emission location and season, with surface anthropogenic NOₓ emissions generally leading to net cooling but aircraft NOₓ leading to warming. Consideration of gas-aerosol interactions suggests that global increases in NOₓ emissions may exert an even larger negative forcing (by enhancing sulfate burdens via increased oxidant levels) than estimated by earlier studies neglecting these interactions. The aerosol response, however, is not robust across models and carbon cycle impacts from O₃ exposure may be large enough to compensate for negative aerosol forcings.

Carbon Monoxide (CO) and Non-Methane Volatile Organic Compounds (NMVOC). Increases in CO and in global anthropogenic NMVOC lead unambiguously to additional climate forcing by raising both tropospheric O₃ and CH₄ abundances (by lowering OH). Gas-aerosol interactions may augment this positive forcing by decreasing sulfate burdens. Projected decreases in these species under the RCPs would thus help to mitigate climate warming. In contrast, the impact of biogenic VOC (BVOC) emissions on climate is highly uncertain. BVOC include a broad suite of carbon compounds emitted naturally from vegetation, including isoprene (C₅H₈), monoterpenes (C₁₀ compounds), sesquiterpenes (C₁₅ compounds), and a large suite of oxygenated VOC (e.g., methanol, ethanol, acetaldehyde, and methylbutenol). These emissions comprise approximately two-thirds of the non-methane VOC budget at the regional and global scale and the terpenoid compounds (isoprene and monoterpenes) are typically more reactive than many anthropogenic VOC. BVOC oxidation can contribute to O₃ formation when NOₓ is present. BVOC are also expected to alter global oxidant levels and thereby impact CH₄ abundances. The magnitude and sign of the net climate influence, however, is uncertain given incomplete knowledge of BVOC oxidation chemistry and corresponding OH changes, particularly in low-NOₓ regions of the atmosphere as discussed further in Section 5.

Methane (CH₄). Although not a direct air pollutant, CH₄ oxidation in the presence of NOₓ enhances global tropospheric O₃. From pre-industrial to present, the RF from the more-than-doubling of the atmospheric CH₄ abundance is estimated to be
+0.48 ± 0.05 W m$^{-2}$, second after CO$_2$ in terms of anthropogenic RF from greenhouse gases$^{10}$. Considering the impact of CH$_4$ on tropospheric O$_3$, stratospheric water vapor, and aerosols permits an estimate of the net RF associated with the preindustrial-to-present growth in CH$_4$ emissions. From this “emission-based view”, RF from CH$_4$ almost doubles from the abundance-based estimate, to 0.8-1.0 W m$^{-2}$; the high end of the range includes the indirect influence of changes in oxidant chemistry on sulfate aerosol$^{36,38}$ (more CH$_4$ leads to less OH and correspondingly less cooling via sulfate aerosol). A multi-model study, however, finds a smaller sensitivity of aerosol to oxidant changes$^{28}$.

Furthermore, the net RF due to increases in CH$_4$ emissions since pre-industrial times has been partially offset by increases in NO$_x$ emissions (which decrease CH$_4$ by increasing global OH). An additional indirect RF occurs through surface O$_3$ increases from CH$_4$ oxidation in the presence of NO$_x$, which damage vegetation and thereby interfere with the carbon cycle$^{37,49}$. Scenarios for the 21$^{st}$ century indicate a wide range of possible CH$_4$ abundances (Figure 1; Nakicenovic et al.$^{14}$) and numerous abatement opportunities could lessen future CH$_4$ RF$^{24,50-52}$. The recent WMO/UNEP report$^{25}$ estimates a 0.2-0.4K decrease by 2050 relative to a reference emission scenario if a defined set of CH$_4$ control technologies were implemented worldwide by 2030, corresponding to a 24% decline in anthropogenic CH$_4$ emissions relative to 2010 levels (Figure 2; see also Shindell et al.$^3$).

Much uncertainty remains in our understanding of the contributions from specific source sectors to CH$_4$ emissions$^{53}$, the underlying factors contributing to recent observed trends(e.g.,$^{54,55,56}$), and in feedbacks from the biosphere$^{13}$. Although these uncertainties limit confidence in accurately projecting the future evolution of CH$_4$, it is clear that decreasing atmospheric CH$_4$ would slow near-term warming and decrease tropospheric O$_3$, including the baseline O$_3$ levels in surface air, thereby lessening the adverse impacts on vegetation and human health$^{23,24,52,57-61}$.

### 2.2 Aerosols

Atmospheric aerosols impact climate in numerous ways (e.g., Isaksen et al.$^{13}$). Briefly, aerosols scatter and absorb shortwave and longwave radiation, known as the aerosol direct effect. Aerosols can also alter the properties of clouds through altering cloud microphysics, known as the aerosol indirect effect, and through changing the temperature structure of the atmosphere, known as semi-direct effects. When absorbing aerosols (i.e. BC and dust) are deposited onto snow and ice surfaces, they reduce surface albedo, which amplifies the radiative forcing. In addition, deposition of aerosols and their dissolved nutrients to the biosphere (land and ocean) may modify biogeochemical processes causing further changes to climate. It is unlikely that these effects add linearly.
Anthropogenic aerosol. Anthropogenic aerosol consists mainly of sulfate, nitrate, ammonium, black carbon (BC) and organic carbon (OC). These aerosol components interact with radiation in different ways and contribute either a cooling or a warming forcing on climate. The AR4 IPCC report\(^\text{10}\) estimated that the net aerosol direct forcing from all anthropogenic components is likely to be a cooling with a best estimate of -0.5±0.4 Wm\(^{-2}\). Myhre et al.\(^\text{62}\) used a combination of models and satellite observations to estimate the direct radiative forcing at -0.35 Wm\(^{-2}\) whereas Bellouin et al.\(^\text{63}\) used satellite observations and estimated -0.65 Wm\(^{-2}\). Sulfate, nitrate and OC aerosol scatter radiation with direct aerosol forcings estimated as -0.4 Wm\(^{-2}\), -0.1 Wm\(^{-2}\) and -0.05 Wm\(^{-2}\) respectively\(^\text{10}\). BC absorbs solar and infrared radiation resulting in a positive aerosol direct effect that is estimated to be between 0.25 and 0.44 Wm\(^{-2}\) in model studies\(^\text{64}\) but potentially as large as 0.9 Wm\(^{-2}\) when constrained by observations\(^\text{65, 66}\). A multi-model assessment by Koch et al.\(^\text{67}\) shows that global models underestimate observed aerosol absorption optical depth (AAOD); scaling model AAOD to match observations resulted in BC multi-model average direct effect of 0.55 Wm\(^{-2}\). The absorption effect of BC in liquid cloud droplets may cause an additional BC radiative forcing of 0.1±0.4 Wm\(^{-2}\)\(^\text{68}\).

The impact of anthropogenic aerosol on clouds is more uncertain. The first aerosol indirect effect, also known as the cloud albedo effect, was estimated by the IPCC AR4 to be -0.7 Wm\(^{-2}\) (range -0.3 to -1.8 Wm\(^{-2}\)\(^\text{10}\). Quaas et al.\(^\text{69}\) used satellite data to estimate a smaller cloud albedo effect of -0.2 Wm\(^{-2}\), potentially suggesting that models may be overestimating the effect. However, a more recent multi-model estimate linked to satellite observations quantified the aerosol indirect effect as -0.7±0.45 Wm\(^{-2}\)\(^\text{70}\). Very little is known on the contribution of different aerosol components to the aerosol indirect effect.

A number of previous studies have quantified the cloud albedo radiative effect of BC-containing particles\(^\text{68, 71-75}\). A multi-model study finds that removal of biomass BC and OC leads to warming, with an average cloud-sky radiative forcing (direct, indirect and semi-direct effects) of 0.11 Wm\(^{-2}\) (range -0.08 to 0.2 Wm\(^{-2}\)) whereas removing only fossil fuel BC yields cooling, with a radiative forcing of -0.08 Wm\(^{-2}\) (range -0.28 to 0.03 Wm\(^{-2}\)\(^\text{73}\). The sign of the cloud forcing is model-dependent and varies with the BC to OC mass ratio, the size of emitted particles, and the magnitude of emission change.

Absorbing aerosols also impact climate after deposition to bright surfaces. In particular, BC causes additional warming of climate after deposition onto snow and ice surfaces through reducing the albedo of snow\(^\text{76}\). Flanner et al.\(^\text{77}\) estimate that the snow-albedo forcing from anthropogenic BC emissions is 0.043 Wm\(^{-2}\) and Jacobson\(^\text{78}\) find that the snow-albedo effect results in a warming of 0.06°C.

The overall effect of anthropogenic aerosol is likely to be a cooling impact on climate. The IPCC AR4 estimated a total forcing of -1.2 Wm\(^{-2}\) from anthropogenic aerosol, partly
offsetting the warming of 2.6 Wm\(^{-2}\) from long-lived greenhouse gases\(^{10}\). These estimates do not include aerosol-induced changes to biogeochemical cycles, which were recently estimated to be -0.5\(\pm\)0.4 Wm\(^{-2}\)\(^{79}\). Uncertainty in the magnitude of the aerosol cooling leads to large uncertainties in projections of future climate change (e.g., \(^{80,81}\)). Removal of this cooling influence due to air pollution abatement policies is expected to enhance future warming\(^{82-85}\).

The strong warming due to the direct effect and snow-albedo effect of BC imply that BC emission reductions could yield a short-term climate benefit\(^{86-92}\). Models indicate that the Arctic is particularly sensitive to BC forcing\(^{77,89,93}\). In particular, Flanner et al.\(^{94}\) showed that BC/OC deposition and snow/ice albedo feedback induces 95% as much springtime snow cover loss over Eurasia as anthropogenic carbon dioxide. The combustion sources that emit BC, also emit other aerosol components that tend to cool climate, principally OC, so the net climate impact of emission controls is uncertain. Ramanathan and Carmichael\(^{66}\) suggest that elimination of BC sources would reduce global surface temperatures by 0.5\(^\circ\) to 1\(^\circ\)C. Shindell et al.\(^{3}\) simulated the effect of implementing a range of air quality measures that control the emissions of BC-containing particles and O\(_3\) precursor species and estimate 0.19\(^\circ\)C avoided warming by 2050. They found that reducing BC emissions resulted in greater reductions in warming from the semi-direct effect than reductions in cooling from the indirect effect, although they note that the magnitude of these different effects is model dependent. In contrast to many earlier studies, Leibensperger et al.\(^{95,96}\) found little warming due to BC over the US and suggested that BC emission controls in the US have little climate mitigation potential. The large uncertainty of the net impact of BC mitigation on surface temperatures primarily reflects uncertainty in cloud feedbacks\(^{72-74,97}\).

Regional temperature responses to aerosol forcings have been examined in a few studies. One study suggests that reductions in sulfate emissions alongside increases in BC emissions have contributed to some of the recent warming observed in the Arctic\(^{98}\). Leibensperger et al.\(^{96}\) found that direct radiative forcing from anthropogenic aerosol peaked in 1970-1990 at -2Wm\(^{-2}\) over the eastern US where it cooled annual mean temperatures over the central and eastern United States by 0.5-1\(^\circ\)C. By 2010, the forcing declined to -1.2Wm\(^{-2}\) due mainly to decreasing SO\(_2\) emissions. They further suggested that U.S. anthropogenic aerosols are currently sufficiently low that future air quality improvements projected to occur over the period 2010-2050 will result in minimal warming (0.1\(^\circ\)C) over the United States\(^{95}\). Mickley et al.\(^{99}\) calculated that complete removal of US anthropogenic aerosol sources would increase annual mean temperatures in the eastern US by 0.4-0.6\(^\circ\)C, with larger increases of 1-2\(^\circ\)C during summertime heat waves due to feedbacks with soil moisture and low cloud cover. Similar feedbacks, including through fog reduction, have also been identified over Europe\(^{100-102}\).
Aerosols may alter regional atmospheric circulation patterns, ranging from shifts in the width of the tropics, Arctic-Oscillation phasing, monsoons, jet locations, and associated precipitation\textsuperscript{66, 95, 103-108}. Absorbing aerosols are possibly more potent at altering circulation patterns than CO\textsubscript{2} and scattering aerosols\textsuperscript{109-112}. Observed drying trends over Africa, South Asia and northern China over the past decades have been attributed at least partially to anthropogenic aerosol forcing (e.g., \textsuperscript{66, 104, 113, 114}), suggesting that aerosol decreases in these regions over the next century will reverse these trends. Precipitation responses to changes in aerosol optical depth, anthropogenic aerosols, and specific fuel emissions sectors have been documented in several modeling studies (e.g., \textsuperscript{5, 26, 115}). Some studies have noted that the sign of the precipitation response to BC depends on the BC vertical distribution\textsuperscript{109, 116} although this is poorly constrained in current models\textsuperscript{67, 117}. Shindell et al.\textsuperscript{118} point out that controlling anthropogenic aerosols should restore disrupted regional precipitation patterns and consideration of this additional climate response may offset some of the adverse effects of the temperature rise induced by removing anthropogenic aerosol.

**Secondary organic aerosol.** SOA arises from both biogenic and anthropogenic sources, and continuously evolves in the atmosphere as a function of multi-generational oxidation and dynamic gas-particle partitioning. Biogenic VOC oxidation can lead to the formation of SOA through a suite of multi-phase reactions, as reviewed recently\textsuperscript{119, 120}. SOA likely contributes a substantial fraction of total organic aerosol\textsuperscript{121} but the impact of SOA on the radiative balance of the atmosphere is poorly understood (e.g., \textsuperscript{122}). Some global models neglect SOA formation from anthropogenic precursor emissions\textsuperscript{64}, which would lead to an underestimate of RF from SOA. Furthermore, formation of biogenic SOA can be enhanced in the presence of anthropogenic organics and particulate matter, where the anthropogenic species provides greater surface area for condensation and enables additional chemical interactions\textsuperscript{123-126}. As recently reviewed\textsuperscript{127}, the formation mechanisms of SOA can be affected by interactions with anthropogenic pollution in multiple ways, including (1) the partitioning processes to transfer gas-phase biogenic VOC to the particulate phase, (2) the role of NO\textsubscript{x} through nitrate-initiated reactions or changing yields depending on NO\textsubscript{x} conditions (with changes depending on the biogenic VOC in question), and (3) the contribution of biogenic VOC nucleation to the formation of new particles in the atmosphere. Recent studies estimate that approximately 20-50\% of SOA could result from anthropogenic activity\textsuperscript{127-132}. Anthropogenic enhancement of SOA implies an additional aerosol forcing beyond that reported in the IPCC AR4. Both Hoyle et al.\textsuperscript{124} and Myhre et al.\textsuperscript{62} report that increases in the SOA burden from pre-industrial to present day have resulted in a direct radiative forcing of nearly -0.1 Wm\textsuperscript{-2}. Spracklen et al.\textsuperscript{74} estimate that if a substantial
fraction of global SOA burden is from anthropogenic activity, the aerosol direct forcing is larger (-0.26±0.15 Wm$^{-2}$); they further estimate a cloud albedo effect of -0.6 Wm$^{-2}$.

O'Donnell et al. use simulations with and without SOA for year 2000 meteorological conditions to estimate an overall climate impact of -0.09 Wm$^{-2}$, which includes opposing influences from the direct effect (-0.31 Wm$^{-2}$) and indirect effect (+0.23 W m$^{-2}$); in their model, particle growth from SOA condensation combined with a larger coagulation sink for small particles leads to a warming indirect effect from SOA. Further work is needed to reconcile the sign of the SOA influence on the cloud albedo feedback.

2.3 Anthropogenic emission sectors

In contrast to long-lived greenhouse gases, the climate impact of SLCFs can depend on emission location and chemical interactions with co-emitted species, which can vary strongly by emission sector. Determining the net climate impact of the suite of emissions from an anthropogenic activity requires explicit three-dimensional model calculations that consider interactions and nonlinearities between the different chemical species emitted from the same sector, which may have offsetting or additive climate impacts. State of the science global chemistry-climate models are often employed to determine the sectoral RF impacts of the SLCFs but only a few studies extend to estimate the more computationally expensive climate response (e.g., 5, 85). The sector-based RFs reviewed below have been calculated for the historical period, referring to the change between the preindustrial and present, and for the future based on current emissions.

The transportation sub-sectors have received the greatest attention. The net climate impacts of biomass burning, electric power production, household fuel burning, shipping, and regional sector impacts have also been assessed. Newer studies consider climate impacts across both gas and aerosol components including aerosol-cloud interactions. Sectoral RFs from the only published study that accounts for all sectors and a broad range of effects have been implemented into a two-box analytical model representation of the carbon cycle and climate that includes heat transfer to the deep ocean to calculate global mean surface temperature change for sustained year 2000 emissions (Figure 3). Based on this work, the power generation and industrial sectors contribute most to warming over the next 50-100 years. On-road transportation and household biofuel usage are also large contributors to warming especially on shorter timescales. Across different modeling systems and emission inventories, road transportation is consistently the largest contributor to warming on short to long timescales, aviation has a small net warming impact, while shipping emissions yield net cooling even on century scale timescales.

In one study, full implementation of state-of-the-art technologies for aerosol control
alongside greenhouse gas increases increased global mean temperature by 2.2K in 2030 relative to today, nearly doubling the effect of greenhouse gases alone over this period.\textsuperscript{85} Maximum abatement in the industry and power sectors (dominated by sulfate aerosol) yielded a somewhat lower response of 1.9 K; maximum abatement in the domestic and transport sectors (dominated by black carbon) still caused an increase of 1.4 K.\textsuperscript{85}

A limitation of the sector-based approach to date is that it refers to a fixed mix of emissions. The most desirable approach is to quantify the climate impact of specific policy-relevant energy shifts or emission control policies. Progress has been made in this direction, for example investigating the trade off of emissions in electrification of transport,\textsuperscript{146} the impacts of tighter vehicle emission standards,\textsuperscript{156} ethanol versus gasoline fuel use in U.S. fleet,\textsuperscript{157} and hydrogen vehicle fuel.\textsuperscript{158, 159} Reducing particulate emissions from residential cooking across the developing world would have strong health benefits, however the co-benefits\textsuperscript{160} or tradeoffs with climate need further investigation.

2.4 Terrestrial biogeochemical feedbacks

Climate impact assessments have largely neglected feedbacks between air pollutants and biogeochemical cycles.\textsuperscript{79} O$_3$ deposition on vegetation suppresses CO$_2$ uptake by the land sink.\textsuperscript{161} One study suggests the magnitudes of the O$_3$ direct RF and the indirect RF through CO$_2$ are comparable.\textsuperscript{49} Aerosols reduce the total amount of radiation reaching the surface but can enhance the diffuse component, which may penetrate deeper into a vegetation canopy depending on the ecosystem type and magnitude of aerosol loading.\textsuperscript{162-164} It is feasible that anthropogenic aerosol may have further substantial effects on the CO$_2$ land sink, by altering the surface energy budget and by altering the amount of precipitation and thereby influencing water stress in vegetation.\textsuperscript{165-167} Atmospheric deposition of sulfate, nitrate and carbonaceous aerosol to surface ecosystems may impact the carbon cycle but these feedbacks are highly uncertain. Overall, pollution-ecosystem feedbacks generally imply enhanced warming from O$_3$-rich anthropogenic emission sectors like road transportation and enhanced cooling from aerosol-rich sectors like power generation.

Because biogenic emissions are climatically driven, multiple feedbacks can occur between climate and terrestrial biogenic emissions and thereby air quality. For example, biogenic VOC emissions are dependent on surface climate including radiation, temperature and soil moisture.\textsuperscript{40} Atmosphere-biosphere feedbacks through biogenic aerosols have been proposed in the literature,\textsuperscript{169-171} but these are largely estimated by models as it is difficult to quantify these feedbacks with observational studies. A qualitative summary of several of these feedbacks is included in Table 1 where we provide an assessment of the level of confidence in the sign of the response to rising
temperatures, along with the implications of these climate-driven feedbacks to O$_3$ and PM levels in surface air. Finally, land-use changes have also been shown to influence regional air quality by altering emissions from the biosphere$^{172-175}$.

3. Air quality response to changing climate

There are several pathways by which climate change may influence O$_3$ and PM air quality. Observational evidence and model studies of these processes have been reviewed recently$^7,13$ and are summarized in Table 1. Briefly, the formation and accumulation of air pollutants is known to correlate strongly with local meteorological variables (e.g., temperature, precipitation, relative humidity, and wind). In many cases, the underlying driver of these strong relationships is variability in synoptic conditions (e.g.,$^{176,177}$). For example, over the eastern United States and Europe, observations demonstrate that extreme air pollution (O$_3$ and PM$_{2.5}$) is typically associated with air stagnation events$^{176,178-184}$. As described below, both modeling and observational analyses suggest an exacerbation of air pollution in a warmer climate, including extreme episodes, at least in some populated regions. We note that some regions may be particularly sensitive to large feedbacks from “natural” O$_3$ and aerosol sources, such as wildfires, dust, and biogenic precursors and from changes in chemical and depositional sinks$^{185-191}$. For example, Southern Europe may be particularly sensitive to climate change due to vegetative feedbacks including increased biogenic VOC emissions and decreased dry deposition when heat leads to closure of stomata$^{192}$.

To understand the processes by which O$_3$ and PM in surface air are influenced by changes in climate, several approaches are used (as reviewed by Jacob and Winner$^7$): (1) sensitivity studies in which individual meteorological parameters are perturbed (e.g.,$^{193,194}$); (2) statistical downscaling of future changes in meteorological fields, using correlations between observed changes in air quality indices and meteorological variables from climate models (e.g.,$^{195,196}$); and (3) the direct calculation of air quality by various global and regional modeling approaches that include: (i) fully coupled global models of chemistry and climate (e.g.,$^{17,197-199}$), often referred to as chemistry-climate models (CCMs); (ii) off-line global chemical transport models (CTMs) forced using projections of meteorological fields from separate AOGCMs (e.g.,$^8$) (iii) global-to-urban models (e.g.,$^{200}$) (iv) dynamical downscaling by linking a suite of climate and atmospheric chemistry models from global to regional scales (e.g.,$^{201,202}$).
Approaches (1) and (2) provide valuable process-level relationships that can be used to evaluate the present-climate simulations in the modeling systems under Approach (3). Caution is needed when applying air pollution relationships with meteorological variables on small spatial scales to project air quality responses to future climate change since the sensitivities of air pollution to individual meteorological variables vary spatially and are non-linear. Statistical downscaling based on synoptic conditions may provide more accurate projections, though models must credibly simulate changes in these synoptic conditions.

3.1 Ozone

In the case of surface O$_3$, it is well established that in many polluted regions, high-O$_3$ events correlate strongly with temperature due to associations of temperature with stagnation episodes, with enhanced photochemistry, and with biogenic and wildfire emissions. Table 1 summarizes current understanding of potential feedbacks to a warming climate and the impacts of increases in these processes on surface O$_3$. Globally, baseline surface O$_3$ levels (defined as those not influenced directly by local emissions) in surface air are very likely to decrease in a warmer climate because higher water vapor abundances will enhance O$_3$ destruction in low-NO$_x$ regions of the atmosphere, leading to a shorter O$_3$ lifetime. Climate-driven increases in lightning NO$_x$ and exchange of stratospheric O$_3$ into the troposphere oppose the negative feedback from rising humidity, but are unlikely to offset the humidity-driven decrease in global surface O$_3$. In some regions and seasons, higher biogenic emissions from vegetation and soils and shifts in transport pathways for intercontinental pollutant transport, could offset the decrease in baseline O$_3$ associated with higher atmospheric water vapor abundances. On large spatial scales, Doherty et al. find little response of intercontinental transport pathways themselves to a warming climate, but note enhanced sensitivity of O$_3$ to emissions within the source region (reflecting isoprene increases and thermally driven decomposition of peroxy acetyl nitrate which could otherwise export NO$_x$ downwind) and decreased sensitivity to intercontinental sources (mainly due to higher water vapor).

We synthesize the surface O$_3$ response to climate change estimated by various Approach (3) modeling systems (blue bars and symbols in Figure 4). A major caveat is that many studies to date use present-day and climate simulations spanning only a few years each, due to computational limitations. While these studies highlight the responses of air quality to meteorological changes, the simulations are too short to distinguish a true anthropogenic-forced climate signal from internally generated climate variability (see also Nolte et al.). The ranges in Figure 4 reflect spatial variability as well as differences across models, including in simulation length, scenarios, reported O$_3$ statistics and thus are not a good measure of the true uncertainty. For example, the multi-model
annual, spatial averages with standard deviations that bracket zero\textsuperscript{221} are the net sum of opposing influences of a warming climate to decrease baseline \( O_3 \) levels but increase \( O_3 \) in polluted regions and seasons\textsuperscript{7,215}. These results are plotted alongside several studies that report spatial ranges for daytime statistics during the high-\( O_3 \) season. Figure 4 shows that climate change-induced increases in surface \( O_3 \) of up to 10 ppb have been estimated for populated regions over the United States by 2050 and up to 6 ppb over Europe by 2030 during the high-\( O_3 \) season. At the sub-continental scale, models often disagree in terms of the sign and magnitude of changes, such as for summer over the Midwest, Southeast and Western United States in 2050, though in some regions, such as the Northeastern United States in summer, they consistently show \( O_3 \) increases\textsuperscript{6,7}.

### 3.2 Particulate Matter

Particulate matter is also influenced by many of the same processes as \( O_3 \) (Table 1), but additional complications include opposing influences on the various PM components and the dependence on precipitation as a major loss pathway\textsuperscript{7,180,222}. Climate-driven changes in PM can be large, but they are highly uncertain and model-dependent (Table 1)\textsuperscript{7}.

Additionally, aerosol burdens are particularly sensitive to precipitation changes and are expected to decrease in regions with increased precipitation\textsuperscript{172,222-226}. Regional and seasonal changes in precipitation must be considered since global soluble aerosol burdens do not typically scale with global precipitation changes\textsuperscript{85,227}. Changes in mixing depths and ventilation of the continental boundary layer also contribute to the sign of the aerosol changes but are highly uncertain\textsuperscript{7,201,204,228}.

Large feedbacks are possible from “natural” aerosol sources (Table 1), particularly carbonaceous aerosols from wildfires, mineral dust, and biogenic precursors to secondary organic aerosol\textsuperscript{188-190,210,229-233}. In regions experiencing warming and drying, wildfires are expected to increase under climate change\textsuperscript{234}. Dust events are known to affect urban areas seasonally, increasing hazardous levels of PM2.5 and reducing visibility in regions downwind of major desert source regions, and in some cases, leading to long-range transport across oceans\textsuperscript{235-238}. As with other types of particulate matter, dust can provide a surface for heterogeneous reactions, leading to enhancement\textsuperscript{239} and uptake of pollutants which can alter the optical properties and solubility of the primary emission\textsuperscript{240,241}.

Although dust is mostly considered as “natural”, the fraction of “anthropogenic” dust (such as from disturbed soils or construction) is subject to debate with possible values ranging from 5-7\%\textsuperscript{230} to 60\%\textsuperscript{229}, with the latest estimate around 25\%\textsuperscript{242}. Primary biogenic aerosols are emitted to the atmosphere in the form of pollen, bacteria, spores, or plant fragments\textsuperscript{243-245}, though knowledge is limited as to the contribution to PM\textsubscript{2.5} concentrations or how they will change with climate\textsuperscript{5}. 
Individual aerosol species respond differently to meteorological changes. Rising temperature and water vapor enhance SO\textsubscript{2} oxidation relative to surface loss, increasing sulfate aerosol but decreasing nitrate aerosol\textsuperscript{222, 223, 228, 246-249}. The biogenic contribution to PM may rise with warming temperatures\textsuperscript{173, 224, 225}, although warmer temperatures should reduce the partitioning of gas phase secondary organics into the aerosol phase\textsuperscript{188}. SOA formation pathways are strongly influenced by climate as shown in multiple regional and global modeling studies\textsuperscript{124, 188, 250}. Future changes in oxidant levels can influence PM distributions\textsuperscript{27}; Leibensperger et al.\textsuperscript{251} point out that intercontinental influences of NO\textsubscript{x} and CO emissions on PM can exceed those from SO\textsubscript{2} emissions\textsuperscript{252}, particularly in regions with high PM pollution.

3.3 Air Pollution Episodes

Air pollution events are generally associated with stagnation events, sometimes coincident with heat waves\textsuperscript{182, 183, 212, 253}. Even under limited global mean warming scenarios, the frequency of heat waves may increase\textsuperscript{254-256}. A warmer climate is projected to decrease rainfall over southern Europe, creating wintertime deficits that lowers soil water content, contributing to extreme heat wave conditions\textsuperscript{257}. A few studies have emphasized positive feedbacks from vegetation (higher emissions and lower stomatal deposition) during heat waves\textsuperscript{183, 257, 258}.

Growing record lengths from surface O\textsubscript{3} and PM observing networks allow for new analyses on connections between extreme pollution events and meteorological conditions, though additional work is needed to determine their relevance to the response to changing climate. For example, over the Northeastern United States, Leibensperger et al.\textsuperscript{181} report a strong inverse relationship between cyclone frequency in summer, and the number of air stagnation events conducive to high-O\textsubscript{3} episodes and Tai et al.\textsuperscript{180} find that stagnant days over the United States are associated with a 2.6 µg m\textsuperscript{-3} increase in PM\textsubscript{2.5} in the United States. Appelhans et al.\textsuperscript{177} find similar correlations between winter PM events in Christchurch, New Zealand and daily to interannual variations in synoptic conditions.

Several modeling studies for the United States and Europe indicate that high-O\textsubscript{3} events are likely to increase in frequency and duration with climate change\textsuperscript{8, 182, 188, 195, 202, 203, 205, 206, 211, 225, 258-264} but the large variability in the incidence of these events makes projections of their changes highly uncertain\textsuperscript{181, 263} and models often disagree at the regional level\textsuperscript{6, 7, 209, 228, 265}. New approaches with centuries-long simulations from CCMs should permit cleaner separation of impacts from climate change versus variability on pollution episodes.

4. Projections of regional air quality changes over the next century
Spatial variations in emissions will influence the local response of O\textsubscript{3} and PM to climate-driven changes (e.g., \textsuperscript{127, 128, 180, 191, 193, 264}). The air quality response to changes in climate is thus intimately tied to the future emissions trajectory, and we consider here projections for regional air quality driven by changes in climate and anthropogenic emissions, both separately and combined. For the combined impacts of climate and anthropogenic emission changes over the next century, we focus on the first multi-model surface O\textsubscript{3} and PM estimates based on the RCP scenarios. Specifically, we include projections from transient climate simulations with interactive chemistry conducted by several CCMs in support of the Coupled Model Intercomparison (CMIP5; lines and shading in Figures 5 & 7) with those from decadal “time slice” simulations in CCMs and CTMs conducted in support of the Atmospheric Chemistry and Climate Model Intercomparison (ACCMIP; circles and black vertical bars in Figures 5 & 7). Ongoing analyses of CMIP5 and ACCMIP simulations will inform the upcoming Intergovernmental Panel on Climate Change Fifth Assessment (IPCC AR5) report. All models use the same anthropogenic emissions\textsuperscript{17, 266} and include changes in both climate and air pollutant emissions along the RCP trajectories. Emissions from the biosphere and lightning NO\textsubscript{x} generally differ across the models, with some models additionally including climate feedbacks to air pollutant emissions from the biosphere. We present these results as a first overview; deeper analysis is beyond the scope of our review.

4.1 Ozone

Projected changes in surface O\textsubscript{3} due solely to changes in climate or solely to changes in emissions of O\textsubscript{3} precursors as published in the literature for 2030 and 2050 show some general patterns across the globe (Figure 4). The largest surface O\textsubscript{3} changes under the recently developed RCP emission pathways are much smaller than those projected under the older SRES scenarios\textsuperscript{17, 58} (Figure 4). These future RCP pathways, however, rely on the accuracy of the underlying O\textsubscript{3} precursor emission trajectories which do not include a high emission trajectory as they assume implementation of NO\textsubscript{x} (and other O\textsubscript{3} precursor) emission controls globally\textsuperscript{19}. Even in the case of the narrower RCP scenario range, we conclude that uncertainty in the emission-driven changes generally spans a wider range than climate-driven changes\textsuperscript{221}; Figure 4).

The large spatial regions considered here and the coarse resolution of the models may mask oppositely signed changes within smaller regions (e.g., within megacities, whose emissions may increase even under emission control scenarios with nationally declining total emissions\textsuperscript{267}). Most models do not consider the impact of land-use changes on biogenic emissions and thereby air pollution, but some initial work suggests these impacts could be substantial. For example, Ganzeveld et al.\textsuperscript{268} find that global changes in land-use and land-cover (LULC) under the SRES A2 scenario could lead to changes of
~20% by 2050 in the tropics, with increases of 6 ppb over the Amazon forest and of 9 ppb over the central African rain forest, and smaller changes (less than 5-10%) over populated regions at northern mid-latitudes. Over the United States, Chen et al. suggest LULC can lead to ±5 ppb changes in surface O$_3$ by 2050 in some locations.

Changes in baseline O$_3$ levels must be considered alongside the impacts of regional emission changes in any future projection as these components together determine the overall air quality response. Over regions with declining regional emissions of O$_3$ precursors, rising global emissions (particularly NO$_x$ and CH$_4$) can offset the O$_3$ decreases obtained with regional emission controls. For example, under the RCP8.5 scenario, the O$_3$ decreases over North America are at least partially offset by rising external sources until 2040, and rising global CH$_4$ raises baseline O$_3$ levels over the entire northern hemisphere (Figure 6). The contribution from CH$_4$ to baseline surface O$_3$ is fairly uniform globally, although the annual mean O$_3$ sensitivity to CH$_4$ ranges by a factor of two across individual models. This uncertainty leads to discrepancies in the magnitude and, in the case of Europe under the RCP8.5 scenario, the sign of the overall surface O$_3$ change (Figure 6). Future increases in shipping emissions, which are subjected to different regulations than land-based emission sectors, may also contribute to raising baseline pollution levels in some regions. Both rising baseline O$_3$ levels and a warming climate could contribute to lengthening the O$_3$ pollution season.

Figure 5 shows projected annual mean surface O$_3$ estimates obtained with a new generation of chemistry-climate models for selected world regions. The models span a fairly wide range of O$_3$ responses, consistent with prior studies, so we focus here on the robust features. Over many regions, the surface O$_3$ trajectories over the next century roughly follow the regional NO$_x$ emissions (compare Figures 5 and 7) and are approximately equivalent to estimates that consider emission changes in the absence of climate change. Under all of the RCP scenarios, global NO$_x$ emissions decline over the next century (Figure 1), decreasing surface O$_3$ in all regions by 2100 with the notable exception of RCP8.5. Under this scenario, the rise in CH$_4$, as well as enhanced stratosphere-to-troposphere O$_3$ transport may be raising O$_3$ levels in many regions. Large inter-annual variations are evident, reflecting model-generated climate variability, and even larger variability is expected over smaller regions, within specific seasons, and in the frequency of high-O$_3$ events.

### 4.2 Aerosols

As compared to O$_3$, we find fewer projections of PM distributions over the next century in the published literature. Comparison across the existing literature is complicated as individual studies focus on specific PM components (e.g., sulfate, nitrate, carbonaceous,
organic). As for O\textsubscript{3}, the relative contributions from changes in emissions versus climate to PM are expected to vary regionally.

Changes driven by anthropogenic emissions are complex and depend on oxidant levels (e.g., \textsuperscript{27,228}). Sulfate generally follows SO\textsubscript{2} emissions, while nitrates follow NH\textsubscript{3} but are also inversely dependent on sulfate (and less sensitive to NO\textsubscript{x}) such that allowing NH\textsubscript{3} to increase while reducing sulfate will offset some of benefit from SO\textsubscript{2} controls\textsuperscript{222}. Continued reductions in SO\textsubscript{2} emissions alongside rising NH\textsubscript{3} emission could lead to nitrate aerosol levels equivalent to sulfate aerosol levels in some regions by 2030\textsuperscript{273}. Projected changes in U.S. aerosol for 2050 are dominated by anthropogenic emission changes except in regions where large precipitation changes are projected\textsuperscript{172,222,224,226} or those that are heavily impacted by wildfire\textsuperscript{190}. Future growth in anthropogenic aromatic emissions could lead to a larger anthropogenic contribution to secondary organic aerosol\textsuperscript{173}. Carmichael et al.\textsuperscript{274} project that changes in BC and SO\textsubscript{2} emissions from Asia over the period 2000 to 2030 will result in increased PM2.5 concentrations.

The regional PM\textsubscript{2.5} projections obtained with the CMIP5 and ACCMIP chemistry-climate models are shown in Figure 7. In several regions such as North America, Europe, and Australia, there is little change projected over the next century, and little difference across the four scenarios. The particularly noisy projections over Africa, the Middle East, and to some extent Australia, reflect the dominance of dust sources, and their strong dependence on inter-annual meteorological variability, in these regions. The Asian regions show clear differences across the scenarios, at least partially reflecting divergent regional emission trajectories.

5. Bridging Knowledge Gaps

Multi-decadal and multi-century simulations with fully coupled chemistry-climate models (e.g., those summarized in Figures 5 and 7) offer a new approach to study interactions between climate and air quality. In particular, these models are well suited to isolate an anthropogenic climate signal on air quality relative to that from climate variability, which has not been possible in most published studies to date. Inclusion of prognostic aerosol-cloud interactions in these models further allows for new insights into the processes contributing to regional climate responses to aerosol forcings. Ongoing community efforts to evaluate rigorously the current generation of chemistry-climate and chemical transport models with process-level observational and laboratory constraints should help bridge knowledge gaps. We focus below on a few areas where current research efforts are expected to advance knowledge rapidly.
Informing air pollution and climate change mitigation. One of the more robust conclusions from work over the past decade is that CH$_4$ controls are a viable strategy for joint mitigation of climate warming and global O$_3$ pollution (e.g., $^{25}$; Section 2.1). Abating CH$_4$ should help to offset some of warming expected from continued removal of atmospheric aerosol (sulfate) motivated by improving public health. The quantitative impact of CH$_4$ oxidation on baseline surface O$_3$ is uncertain by a factor of two$^{58}$. Narrowing this range likely requires better constraints on global NO$_x$ distributions$^{51}$; new space-based approaches may be particularly useful$^{275-281}$. Black carbon mitigation is another approach under consideration to improve air quality and lessen global warming, with several emission control strategies identified$^{25}$ but substantial uncertainty remains regarding the varied roles by which black carbon affects climate (Section 2.2). Recent advances have been made in characterizing and propagating uncertainties through the multitude of processes governing the emission of radiatively active species and their atmospheric distributions and climate impacts$^{282-284}$. Additional work is needed to extend these uncertainty analyses to other chemical species and anthropogenic sectors. By identifying which processes contribute most to uncertainties, these approaches inform the science community as to where future investments may be most rewarding.

Evaluating chemistry-climate models. A wide range of model estimates exists for regional air quality both at present and in the future, even along a given emission trajectory (Figures 4,5,7). For example, the average surface O$_3$ during the 1980-2005 reference period is 5-10 ppb higher in the transient simulations as compared to the ACCMIP CTM and CCM simulations over several regions (Figure 5). Growing record lengths of historically observed relationships between relevant meteorological variables and air quality provide useful information for evaluating models (e.g.$^{180,191,208,285-288}$) and may also help to improve our understanding of the links between air quality and climate though further study is required. Space-based constraints on instantaneous radiative forcing from SLCFs could aid in characterizing model bias in RF estimates from models$^{63,69,289,290}$. These observational approaches, combined with multi-model analysis that seeks to link differences across models to specific processes$^{291,292}$ would help to reduce uncertainties in both present-day and projected air pollution estimates.

Constraining chemical mechanisms. Fundamental questions remain in our mechanistic understanding that severely limit confidence in projecting future air quality in a changing climate. For example, meta-analysis indicates that the sensitivity of O$_3$ to climate change is strongly dependent on the treatment of organic nitrates (RONO$_2$), specifically those from isoprene$^{6,7,293}$. The rate at which RONO$_2$ reacts to recycle NO$_x$ is poorly understood$^{294,295}$ and ranges from 0-100% are used in CCMs and CTMs; this range affects the sign of the O$_3$ response to changes in BVOC emissions$^{296-299}$. Chemical mechanisms that include RONO$_2$ formation without recycling NO$_x$ via subsequent
reaction display little biogenic VOC-ozone-climate sensitivity⁸, and mechanisms that ignore RONO₂ altogether show large biogenic VOC-ozone-climate sensitivity²²³. There is additional uncertainty in the RONO₂ yield from isoprene oxidation with laboratory yields ranging from 4-12% (e.g., see summary in Horowitz et al.²⁹⁷, and newer findings³⁰⁰,³⁰¹). Knowledge of BVOC oxidation and subsequent SOA formation is advancing rapidly but uncertainties remain⁴⁶,¹¹⁹,¹²⁷,³⁰². Aerosol-oxidant interactions also require further study as they may determine PM air quality in some regions²⁷,²⁵¹. Oceanic reactive halogen species released into the atmosphere by the photodecomposition of organohalogens (including iodine-containing species) and via autocatalytic recycling on sea-salt aerosols contribute to O₃ destruction and may play an important role in the O₃ budget and tropospheric oxidizing capacity³⁰³. One study finds inclusion of halogen chemistry to be an important component of the natural background O₃ budget and thus preindustrial O₃ baseline concentrations³⁰⁴ but further research is needed to fully examine the climate and air quality impacts.

**Reducing uncertainty in aerosol forcing.** A new generation of global models allows deeper study at the process level as they include aerosol microphysics and prognostic aerosol cloud interactions. However, large variability between models⁷³,¹¹⁷ highlights the need for detailed evaluation of aerosol microphysical schemes before uncertainty in aerosol forcing can be reduced. Synthesis of field observations to develop datasets with global coverage to confront models will be important. New approaches are also required to understand the microphysical processes that are most responsible for model diversity helping to prioritize future research directions³⁰⁵.

**Identifying robust regional air pollution and climate responses.** The northern mid-latitudes have garnered the most attention in the literature in terms of air quality and climate interactions; more work is needed to understand the future evolution of air quality in the tropics and southern hemisphere. More reliable projections of air quality require confidence in the regional climate responses, including precipitation, the positioning of mid-latitude storm tracks and subtropical high pressure systems as well as convection (e.g.,⁶,⁷,³⁰⁶). Improved understanding of the spatially resolved climate response to specific emission control strategies (including effects on hydrological cycle, temperature and circulation) are needed, particularly for the SLCFs in order to bypass current limitations in the RF metric. However, identifying a statistically significant signal detectable over internal climate model variability for relatively small net climate impacts from regional or sectoral emissions represents a non-trivial challenge and may even be impossible²²⁶. The published literature debates whether the spatial pattern of the future surface temperature response to aerosol forcing mirrors that from greenhouse-gas forcing or rather follows the local aerosol forcing patterns⁹⁵,⁹⁸,⁹⁹,³⁰⁷-³⁰⁹. A multi-model analysis begins to reconcile these previous findings, indicating a strong sensitivity of the surface
temperature response to the latitudinal forcing distribution but limited sensitivity to longitude\textsuperscript{310}. Regional precipitation may be particularly sensitive (both sign and magnitude) to aerosol forcing location\textsuperscript{118}.

Advancing knowledge of anthroposphere-biosphere interactions. Human interactions with the terrestrial biosphere are a major uncertainty but crucial to understand because vegetation acts as both a source and a sink for many air pollutants (e.g., \textsuperscript{13, 185}). The attribution of O\textsubscript{3} and PM air pollution to “anthropogenic” versus “biogenic” sources is complicated by chemistry that involves both anthropogenic and biogenic precursors, for both O\textsubscript{3} and aerosol\textsuperscript{128, 311}. A major uncertainty regarding BVOC feedbacks is the opposing influences of rising CO\textsubscript{2} versus rising temperature\textsuperscript{312}. Sources from agriculture and livestock sectors are generally poorly constrained but non-negligible, particularly for CH\textsubscript{4}\textsuperscript{313} and NH\textsubscript{3}\textsuperscript{314}. Human-driven changes in land-use and land cover, such as urbanization or shifts between forests and agriculture, could dramatically alter future O\textsubscript{3} and aerosol precursor emissions from the biosphere\textsuperscript{173, 175, 258} as well as dry deposition\textsuperscript{174}, and should be considered in future assessments.

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### Table 1. Impacts on surface O$_3$ and PM over land from increases in specific temperature-driven pathways, adapted from Table 1 of Jacob and Winner; see also Figure 2 of Isaksen et al.\textsuperscript{13}.

<table>
<thead>
<tr>
<th>Process</th>
<th>Level of confidence that warmer climate leads to increase\textsuperscript{a}</th>
<th>Impact of increase in process on PM\textsuperscript{b,c}</th>
<th>Impact of increase in process on O$_3$\textsuperscript{b,c}</th>
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<tr>
<td>CH$_4$ from wetlands</td>
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<td>* (-)</td>
<td>++ B</td>
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<tr>
<td>Dust</td>
<td>Low</td>
<td>++</td>
<td>- , LR</td>
<td>229, 230, 320-322</td>
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<tr>
<td>Soil NO$_x$</td>
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<td>+ B, LR</td>
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<tr>
<td>STE of O$_3$</td>
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<td>++ B</td>
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<td>++ B</td>
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<tr>
<td>Dry Deposition</td>
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<td>=</td>
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<tr>
<td>precipitation</td>
<td>Low</td>
<td>--</td>
<td>= LR</td>
<td>/</td>
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</table>

a. Low indicates conflicting evidence on sign of response to a warmer climate; Medium indicates that at least in some regions (e.g., for fires, warm and dry; for wetlands warm and wet) increases are expected; High indicates sign of response to a warming climate is well understood.

b. Symbols follow those in Jacob and Winner: ++ consistently positive, + generally positive, = weak, - generally negative -- consistently negative in response to an increase in the pathway. In addition we use ? for uncertainty in sign of response and * indicates the response depends on changing oxidant levels; the sign, if known, is shown in parentheses.

c. B denotes impact on baseline O$_3$ levels; LR indicates local-to-regional responses.
**Table 2.** Simulations used in Figures 5 and 7 (all models provided O₃; those providing PM2.5 are italicized).

<table>
<thead>
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Figure 1. Future evolution of CH₄ abundance (upper left) and selected air pollutant precursor emissions (SO₂ (upper right), NO (bottom left), and BC (bottom right)) from anthropogenic, biomass burning and shipping sources combined, under the RCP scenarios.¹⁷,¹⁸
Figure 2. Observed deviation of temperature through 2009 and projections under various scenarios, all relative to the 1890-1910 mean. Results for future scenarios are the central values from analytic equations estimating the response to forcings calculated from composition-climate modeling and literature assessments (see online material of Shindell et al.). The rightmost bars give 2070 ranges, including uncertainty in radiative forcing and climate sensitivity. A portion of the uncertainty is systematic, so that overlapping ranges do not mean there is no significant difference (for example, if climate sensitivity is large, it is large regardless of the scenario, so all temperatures would be toward the high end of their ranges; see www.giss.nasa.gov/staff/dshindell/Sci2012). Reproduced from Figure 2 of Shindell et al. [will need formal permission from AAAS].
Figure 3. Global mean surface temperature change for sustained constant emissions from global sectors based on present day emission rates and mixes. Signal includes long-lived greenhouse gases, short-lived climate forcing agents, and aerosol-cloud interactions. Values are determined by implementing sectoral RF results\textsuperscript{143,154} into a 2-box analytical climate-carbon cycle model\textsuperscript{135}.
Figure 4. Changes in surface $O_3$ solely due to climate (blue) or emissions (colored by emission scenario) reported in the literature in 2030 (top) and 2050 (bottom) for selected world regions. Results from individual studies are labeled by letters underneath the corresponding plot symbols. Vertical bars represent a combination of ranges as reported in the literature: (1) multi-model mean and standard deviations in annual mean, spatial averages from the ACCENT/Photocomp study for 2030 (A$^{221}$); (2) application of a parameterization developed from the multi-model ensemble of the Task Force on Hemispheric Transport of Air Pollution regional source-receptor relationships to estimate surface $O_3$ response for several emission scenarios in 2030 and 2050 globally and within the TF HTAP continental regions (C$^{58}$); (3) spatial averages across a region, denoted by filled squares (D$^{262}$, E$^{172}$, I$^{225}$, K$^{223}$, M$^{202}$, Q$^{247}$, T$^{51}$, U$^{60}$, W$^{23}$, X$^{265}$); (4) spatial ranges across a region as estimated with one model or combined across several individual modeling studies, denoted by dashed lines (B$^{257}$, D$^{262}$, F$^{206}$, G$^{8}$, H$^{220}$, J$^{205}$, K$^{223}$, L$^{226}$, P$^{27}$, R$^{328}$, S$^{229}$, V$^{24}$, Y$^{330}$, Z$^{331}$, I$^{266}$). Regional definitions, methods, and reported metrics (e.g., 24-h versus daily maximum values over a 1-hour or 8-hour averaging period, annual or seasonal averages) vary across studies. Climate change scenarios vary across studies, but are combined into ranges denoted by blue bars for two reasons: (1) there is...
little detectable cross-scenario difference in the climate response, particularly in 2030, and (2) many of these estimates are based on simulations that are too short to cleanly attribute a climate change signal and thus it is not appropriate to attribute differences to particular climate forcing scenarios.

Figure 5. Changes in annual mean surface ozone (volume mixing ratio) following the RCP scenarios over the next century, spatially averaged over selected world regions (shaded land regions). For each RCP, the colored lines denote 3-model mean estimates from transient simulations with CMIP5 chemistry-climate models; shading covers the full range across models for each RCP (Table 2). Filled circles with vertical lines indicate the multi-model average and full cross-model range from the 2010, 2030, 2050, and 2100 ACCMIP decadal time slice simulations, colored by RCP scenario (Table 2). The circles are offset by a year to clearly distinguish the vertical lines. Changes are relative to the 1986-2005 reference period for the transient simulations, and relative to the average of the 1980 and 2000 decadal time slices for the ACCMIP ensemble. The average ozone value during the reference period, spatially averaged over each region, is shown in each panel, with the standard deviation reflecting the cross-model range (transient CMIP5 models on the upper left; ACCMIP models on the upper right). In cases where multiple ensemble members are available from a single model, they are averaged prior to inclusion in the multi-model mean.
Figure 6. Regional mean O\textsubscript{3} changes along the RCP 8.5 scenario showing the ensemble mean (coloured line) and individual model responses (grey lines). A source attribution is presented for each region in the lower panels. Reproduced from Figure 10 of Wild et al.\textsuperscript{58} [Need formal permission].
Figure 7. As in Figure 5 but for PM2.5 (mass mixing ratios, ppb). PM2.5 estimates are from the italicized models in Table 2, calculated as the sum of individual aerosol components (black carbon + organic carbon, + sulfate + secondary organic aerosol + 0.1*dust + 0.25*sea salt).