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CRITICAL REVIEW

Global air quality and climate†‡

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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. We review the implications of projected changes in methane (CH₄), ozone precursors (O₃), and aerosols for climate (expressed in terms of the radiative forcing metric or changes in global surface temperature) and hemispheric-to-continental scale air quality. Reducing the O₃ precursor CH₄ would slow near-term warming by decreasing both CH₄ and tropospheric O₃. Uncertainty remains as to the net climate forcing from anthropogenic nitrogen oxide (NO_x) emissions, which increase tropospheric O₃ (warming) but also increase aerosols and decrease CH₄ (both cooling). Anthropogenic emissions of carbon monoxide (CO) and non-CH₄ volatile organic compounds (NMVOC) warm by increasing both O₃ and CH₄. Radiative impacts from secondary organic aerosols (SOA) are poorly understood. Black carbon emission controls, by reducing the absorption of sunlight in the atmosphere and on snow and ice, have the potential to 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; a first step

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towards this goal involves estimating net climate impacts from individual emission sectors. Modeling and observational analyses suggest a warming climate degrades air quality (increasing surface O₃ and particulate matter) in many populated regions, including during pollution episodes. Prior Intergovernmental Panel on Climate Change (IPCC) scenarios (SRES) allowed unconstrained growth, whereas the Representative Concentration Pathway (RCP) scenarios assume uniformly an aggressive reduction, of air pollutant emissions. New estimates from the current generation of chemistry–climate models with RCP emissions thus project improved air quality over the next century relative to those using the IPCC SRES scenarios. These two sets of projections likely bracket possible futures. We find that uncertainty in emission-driven changes in air quality is generally greater than uncertainty in 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 O₃ 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 (O₃) and aerosols (referred to interchangeably with particulate matter below) in the near-surface atmospheric boundary layer. Increases in surface O₃ and particulate matter (with diameter <2.5 μm, 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.¹ When considering strategies to abate air pollution and mitigate anthropogenic climate warming, policymakers face tradeoffs² and synergies.³ 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.*, ref. 4 and 5), possibly at rates threatening the survival of some ecosystems.² In the absence of emission changes, a warming climate may degrade air quality in many polluted regions,^{6,7} an impact that has been referred to as “a climate change penalty” on air quality.⁸ We review the current understanding of the interactions between air pollutants and climate, aiming to identify robust conclusions that may guide decision-making and to highlight critical knowledge gaps.

Any successful climate mitigation effort must address carbon dioxide (CO₂). The relatively short lifetimes of other radiatively active air pollutants and their precursors (days to weeks for O₃ 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.⁹ We thus refer to these species as “Near-Term Climate Forcers” (NTCFs). Many studies reviewed here compare climate impacts of different NTCFs by estimating radiative forcing (RF). RF is 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.¹⁰ 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, heterogenous distributions of air pollutants and their precursors (except for the well-mixed greenhouse gas CH₄) which vary in their forcing efficacy.^{11,12} 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 to air pollutants and their precursors, and some of the chemistry–climate interactions¹³ in Table 1. Below, we review this recent 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 emission trajectories to date are those described in the Special Report on Emission Scenarios (SRES).¹⁴ A newly developed set of scenarios, the Representative Concentration Pathways (RCPs), is currently in use 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 the SRES scenarios in several recent publications.^{15–22} All RCPs assume aggressive air pollution abatement measures (Fig. 1) and correspondingly large decreases in PM and O₃ precursors globally.^{17,23} Exceptions include ammonia (NH₃), which increases in nearly all scenarios and CH₄, which ranges from a 30% decrease to a more than doubling by 2100 (Fig. 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.

Table 1 Impacts of temperature-driven pathways on surface O₃ and PM over land, adapted from Table 1 of Jacob and Winner⁷ and Fig. 2 of Isaksen *et al.*¹³

Process	Level of confidence that warmer climate leads to increase ^a	Impact of increase in process on PM ^b	Impact of increase in process on O ₃ ^{b,c}	Example references
Non-CH ₄ BVOC emissions	Low	+	? B, LR	6, 13, 54, 205, 325, 346, 349 132, 134, 350
Wildfires	Medium	++	+ B ?LR	200, 201, 204, 229, 254, 351, 352
CH ₄ from wetlands	Medium	* (-)	++ B	13, 56, 353
Dust	Low	++	- LR	249, 250, 354–356
Soil NO _x	Medium	*	+ B, LR	13
STE of O ₃	Medium	++	++ B	13, 238, 320, 357
Lightning NO _x	Low	* (+)	++ B	5, 13, 320, 357–359
Dry deposition	Low	?	-- LR, B	13, 188, 199, 357
Humidity	High	+	-- B = LR	7, 228, 234, 357
Regional stagnation	Medium	++	++ LR	7, 190, 195, 360
Wind speed	Low	-	-LR	7, 361
Mixing depth	Low	--	= LR	7, 361
Cloud cover	Low	-	-LR	7
Precipitation	Low	--	= LR	7, 361

^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. We add ? to indicate uncertainty in sign of response and * to indicate the response depends on changing oxidant levels; the sign, if known, is shown in parentheses. ^c B denotes impact on baseline O₃ levels; LR indicates local-to-regional responses.

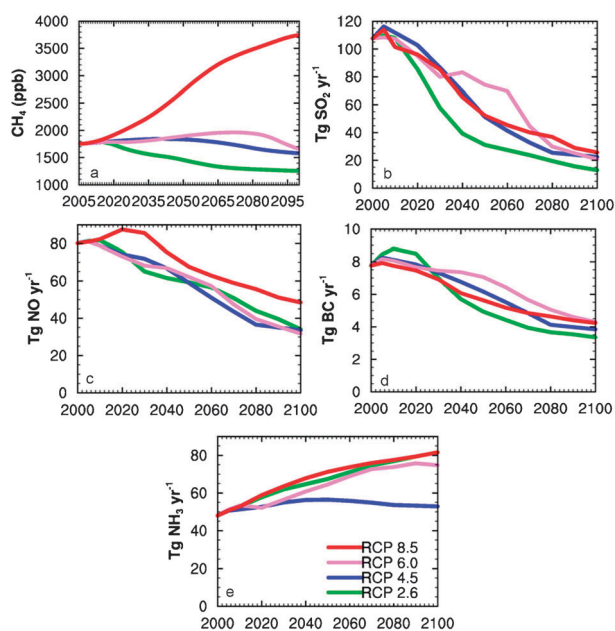


Fig. 1 Future evolution of (a) CH₄ abundance and selected global emissions of air pollutants and precursors, (b) SO₂, (c) NO, (d) BC, and (e) NH₃, from anthropogenic plus biomass burning sources combined, under the RCP scenarios.^{16–18}

2. Sensitivity of climate to changes in air pollutant emissions

We review the literature describing climate responses to changes in emissions of O₃ precursors (Section 2.1), and aerosols and their precursors (Section 2.2). Understanding the climate responses to the particular mix of pollutant plus greenhouse gas 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.²⁴ Throughout this section, we report RFs from pre-industrial to present-day as reported in the literature. Individual studies vary in their choice of base year for both pre-industrial and present-day but this is unlikely to be a major contribution to the range of reported RFs.

2.1 Ozone (O₃) precursors

The increase in tropospheric O₃ from pre-industrial to present-day driven by human activities²⁵ has led to a positive climate forcing ($+0.35^{+0.3}_{-0.1}$ W m⁻²)¹⁰ and substantial increases in O₃ RF are estimated for the next century with the SRES scenarios.²⁶ Emissions of O₃ precursors, particularly nitrogen oxides (NO_x) and volatile organic compounds (VOC) also produce secondary aerosols and thereby a negative climate forcing, though this aerosol RF is poorly quantified relative to the O₃ RF.²⁷ We discuss below the role of specific O₃ precursor emissions, with a strong emphasis on the role of CH₄, which has received growing attention in recent years as a means to address jointly air pollution and climate goals.^{3,28–30} The dominant CH₄ 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. Indirect RFs from O₃ precursors can occur *via* precursor-induced changes in oxidant concentrations, which can alter aerosol burdens; similarly, changes in aerosol burdens may alter O₃ concentrations.^{27,31,32} An additional, poorly quantified, indirect RF occurs when high surface O₃ concentrations damage vegetation and interfere with the carbon and hydrologic cycles.^{33,34}

Nitrogen oxides (NO_x). As NO_x levels are sufficiently low in much of the troposphere, increases in anthropogenic NO_x (NO + NO₂) raise global average tropospheric O₃

levels (e.g., ref. 35), thereby exerting a positive climate forcing. This positive forcing may be offset by an accompanying negative forcing due to lower CH₄ abundances, because increasing NO_x enhances tropospheric OH and reaction with OH is the primary CH₄ loss pathway in the atmosphere.³⁶ The net RF of these opposing influences on the climate system depends upon the emission location and season, with surface anthropogenic NO_x emissions generally leading to net cooling but aircraft NO_x aloft leading to small net warming.^{37–43} Consideration of gas–aerosol interactions suggests that global increases in NO_x emissions may exert a greater negative forcing by enhancing sulfate burdens *via* increased oxidant levels.⁴⁴ The aerosol response, however, is not robust across models²⁷ and carbon cycle impacts from O₃ exposure³⁴ 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 positive climate forcing by raising both tropospheric O₃ and CH₄ abundances through a reduction in OH.^{28,34,41,42,45,46} Individual anthropogenic NMVOC, however, can lead to secondary organic aerosol and an associated negative forcing.⁴⁷ Gas–aerosol interactions may augment this positive forcing by decreasing sulfate burdens.^{27,44} Projected decreases in these species under the RCPs¹⁷ 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 number of oxygenated VOC (e.g., methanol, ethanol, acetaldehyde, and methylbutanol). These emissions contribute approximately two-thirds of the non-methane VOC budget at the regional and global scale⁴⁸ and the terpenoid compounds (isoprene and monoterpenes) are typically more reactive than many anthropogenic VOC.⁴⁹ BVOC oxidation can contribute to O₃ formation when NO_x is present.^{50,51} BVOC also alter global oxidant levels and thereby impact CH₄ abundances.⁵² 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-NO_x regions of the atmosphere^{53,54} as discussed further in Section 5.

Methane (CH₄). Although not a direct air pollutant, CH₄ oxidation in the presence of NO_x enhances global tropospheric O₃.^{35,55,56} From pre-industrial to present, the RF from the more-than-doubling of the atmospheric CH₄ abundance is estimated to be $+0.48 \pm 0.05 \text{ W m}^{-2}$, second after CO₂ in terms of anthropogenic RF from greenhouse gases.¹⁰ An estimate of the net RF associated with the preindustrial-to-present growth in CH₄ emissions includes the impact of CH₄ on tropospheric O₃, stratospheric water vapor, and aerosols. From this “emission-based view”, RF from CH₄ almost doubles from the abundance-based estimate to $0.8\text{--}1.0 \text{ W m}^{-2}$.^{44,45} The high end of the range includes the indirect influence of changes in oxidant chemistry on sulfate aerosol, where more CH₄ leads to less OH and correspondingly less cooling *via* sulfate aerosol.

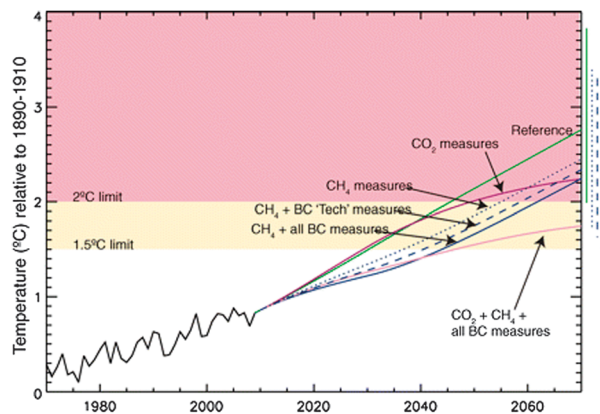


Fig. 2 Observed temperature anomaly through 2009³⁶² and projected temperature anomaly under various scenarios, all relative to the 1890–1910 mean. Results for future scenarios are the central values from analytic equations estimating the response to forcings calculated from composition–climate modeling and literature assessments (see online material of Shindell *et al.*³). The rightmost bars give 2070 ranges, including uncertainty in radiative forcing and climate sensitivity. A portion of the uncertainty is systematic, so statistically significant differences between scenarios may still occur even with overlapping ranges (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). From Shindell *et al.*³ (their Fig. 1). Reprinted with permission from AAAS.

A multi-model study, however, suggests a smaller sensitivity of aerosols to oxidant changes.²⁷ Furthermore, the net RF due to increases in CH₄ emissions since pre-industrial times has been partially offset by increases in NO_x emissions (which decrease CH₄ by increasing global OH). Scenarios for the 21st century indicate a wide range of possible CH₄ abundances (Fig. 1; Nakicenovic *et al.*¹⁴) and numerous abatement opportunities could lessen future CH₄ RF.^{29,57–59} The recent WMO/UNEP report³⁰ estimates a 0.2–0.4 K decrease by 2050 relative to a reference emission scenario if a defined set of CH₄ control technologies were implemented worldwide by 2030, corresponding to a 24% decline in anthropogenic CH₄ emissions relative to 2010 levels (Fig. 2; see also Shindell *et al.*³).

Much uncertainty remains in our understanding of the contributions from specific source sectors to CH₄ emissions,⁶⁰ the underlying factors contributing to recent observed trends (e.g., ref. 61–63), and in feedbacks from the biosphere.¹³ Although these uncertainties limit confidence in accurately projecting the future evolution of CH₄, it is clear that decreasing atmospheric CH₄ would slow near-term warming, due to its stronger climate impact on the 20 year (*versus* 100 year) time frame, which could help to slow Arctic sea ice loss over the next few decades.⁶⁴ Reducing CH₄ would also decrease tropospheric O₃, including the baseline O₃ levels in surface air, thereby lessening the adverse impacts on vegetation and human health.^{28,29,59,65–69}

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.⁷⁰ Aerosols can also alter the properties of clouds through indirect effects (by affecting cloud microphysics),⁷¹ the semi-direct effect (where aerosol absorption affects temperature structure),⁷² and cloud absorption effects (from absorbing aerosol inclusions within or interstitially between hydrometeors).⁷³ When absorbing aerosols (*i.e.* BC and potentially dust) are deposited onto snow and ice surfaces, they reduce surface albedo, which amplifies the positive RF. In addition, deposition of aerosols and their dissolved nutrients to the biosphere (land and ocean) may modify biogeochemical processes causing further changes to climate.⁷⁴ 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 depending on size and composition and contribute either a cooling or a warming forcing on climate. The AR4 IPCC report¹⁰ estimated that the net aerosol direct forcing from all anthropogenic components is likely to be a cooling with a best estimate of $-0.5 \pm 0.4 \text{ W m}^{-2}$. Myhre *et al.*⁷⁵ used a combination of models and satellite observations to estimate the direct RF at -0.35 W m^{-2} whereas Bellouin *et al.*⁷⁶ used satellite observations to estimate -0.65 W m^{-2} . Myhre⁷⁷ report a best estimate for aerosol direct RF of -0.3 W m^{-2} after reconciling divergent estimates from modeling and observational approaches by accounting for differences in cloud-masked forcing and changes over the last century in bulk aerosol optical properties. Sulfate, nitrate and OC aerosol scatter radiation with direct aerosol RFs estimated as -0.4 W m^{-2} , -0.1 W m^{-2} and -0.05 W m^{-2} respectively.¹⁰ BC absorbs solar and infrared radiation resulting in a positive aerosol direct effect that is estimated to be between $+0.25$ and $+0.44 \text{ W m}^{-2}$ in model studies⁷⁸ but potentially as large as $+0.9 \text{ W m}^{-2}$ when constrained by observations.^{79,80} A multi-model assessment by Koch *et al.*⁸¹ 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 $+0.55 \text{ W m}^{-2}$. The absorption effect of BC in liquid cloud droplets may cause an additional BC RF of $+0.1 \pm 0.4 \text{ W m}^{-2}$.⁸²

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 W m^{-2} (range -0.3 to -1.8 W m^{-2}).¹⁰ Quaas *et al.*⁸³ used satellite data to estimate a smaller cloud albedo effect of -0.2 W m^{-2} , potentially suggesting that models may be over-estimating the effect. However, a more recent multi-model estimate⁸⁴ linked to satellite observations quantified the aerosol indirect effect as $-0.7 \pm 0.45 \text{ W m}^{-2}$. Very little is known on the contribution of different aerosol components to the aerosol indirect effect. While several studies have quantified the cloud albedo radiative effect of BC-containing particles,^{82,85–89} 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 the 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 by

reducing the albedo of snow.⁹⁰ Flanner *et al.*⁹¹ estimate that the snow-albedo forcing and associated feedbacks from all anthropogenic BC emissions result in global warming (annual mean temperature) of between 0.1 and 0.15 K, while Jacobson⁹² finds that fossil and biofuel sources alone result in a warming of between 0.03 and 0.11 K through this effect.

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 W m^{-2} from anthropogenic aerosol, partly offsetting the warming of 2.6 W m^{-2} from long-lived greenhouse gases.¹⁰ These estimates do not include aerosol-induced changes to biogeochemical cycles, which were recently estimated to be $-0.5 \pm 0.4 \text{ W m}^{-2}$.⁷⁴ Uncertainty in the magnitude of the aerosol cooling leads to large uncertainties in projections of future climate change (*e.g.*, ref. 93 and 94). Removal of this cooling influence due to air pollution abatement policies is expected to enhance future warming.^{95–98}

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.^{99–105} Models indicate that the Arctic is particularly sensitive to BC forcing.^{91,101,106} In particular, Flanner *et al.*¹⁰⁷ find that deposition of BC and OC and the subsequent 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 Carmichael⁸⁰ suggest that elimination of BC sources would reduce global surface temperatures by 0.5 to 1 K. Shindell *et al.*³ simulated the effect of implementing a range of air quality measures that control the emissions of BC-containing particles and O₃ precursor species and estimate 0.19 K 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, Leibensperger *et al.*^{108,109} 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 reflects uncertainty in co-emitted species as well as cloud feedbacks.^{86–88,110}

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.¹¹¹ Leibensperger *et al.*¹⁰⁹ found that direct RF from anthropogenic aerosol peaked in 1970–1990 at -2 W m^{-2} over the eastern US where it cooled annual mean temperatures over the central and eastern United States by 0.5–1 °C. By 2010, the forcing declined to -1.2 W m^{-2} due mainly to decreasing SO₂ 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 °C) over the United States.¹⁰⁸ Mickley *et al.*¹¹² calculated that complete removal of US anthropogenic aerosol sources would increase annual mean temperatures in the eastern US by 0.4–0.6 °C,

with larger increases of 1–2 °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.^{113–115}

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.^{80,108,116–121} Absorbing aerosols are possibly more potent at altering circulation patterns than CO₂ and scattering aerosols.^{122–125} Observed drying trends over Africa, South Asia and northern China over the past decades have been partially attributed to anthropogenic aerosol forcing (*e.g.*, ref. 80, 117, 126 and 127), suggesting that aerosol decreases in these regions over the next century would 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.*, ref. 5, 31 and 128). Some studies have noted that the sign of the precipitation response to BC depends on the BC vertical distribution^{122,129} although this is poorly constrained in current models.^{81,130} Shindell *et al.*¹³¹ 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). 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.^{132,133} SOA likely contributes a substantial fraction of total organic aerosol¹³⁴ but the impact of SOA on the radiative balance of the atmosphere is poorly understood (*e.g.*, ref. 135). Some global models neglect SOA formation from anthropogenic precursor emissions,⁷⁸ 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.^{136–139} As recently reviewed,¹⁴⁰ 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_x through nitrate-initiated reactions or changing yields depending on NO_x conditions (with changes depending on the specific biogenic VOC), 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.^{140–145} Anthropogenic enhancement of SOA implies an additional aerosol forcing beyond that reported in the IPCC AR4. Both Hoyle *et al.*¹³⁷ and Myhre *et al.*⁷⁵ report that increases in the SOA burden from pre-industrial to present day have resulted in a direct RF of nearly -0.1 W m^{-2} . Spracklen *et al.*⁸⁸ estimate that if a

substantial fraction of global SOA burden is from anthropogenic activity, the aerosol direct forcing is larger ($-0.26 \pm 0.15 \text{ W m}^{-2}$), and they further estimate a cloud albedo effect of -0.6 W m^{-2} . O'Donnell *et al.*⁴⁷ use simulations with and without SOA for year 2000 meteorological conditions to estimate an overall climate impact of -0.09 W m^{-2} , which includes opposing influences from the direct effect (-0.31 W m^{-2}) and indirect effect ($+0.23 \text{ W m}^{-2}$). In their simulations, 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 NTCFs 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 co-emitted chemical species, which may have offsetting or additive climate impacts. Global chemistry–climate models are often employed to determine the sectoral RF impacts of the NTCFs but few studies extend to estimate the more computationally expensive climate response (*e.g.*, ref. 5 and 98).

The transportation sub-sectors have received the greatest attention.^{40,146–156} The net climate impacts of biomass burning,^{157,158} electric power production,^{159,160} household fuel burning,¹⁶¹ and regional sector impacts^{156,161–164} have also been assessed. Newer studies consider climate impacts across both long-lived greenhouse gases and NTCF gas and aerosol components including aerosol–cloud interactions.^{148,165} In terms of net RF, which has been estimated for several sectors across different modeling systems and emission inventories, road transportation is consistently the largest contributor to warming on short to long timescales, aviation generally produces a small net warming, while shipping emissions yield net cooling on century timescales.¹⁶⁶ Overall climate impacts may change in the future as various technology controls are applied to different sectors. Black carbon emissions from shipping have been comprehensively reviewed¹⁶⁷ and are of particular concern in the Arctic because of the local climate sensitivity to snow-ice albedo effects following deposition.¹⁵⁶

In one study, full implementation of state-of-the-art aerosol control technologies in conjunction with increasing greenhouse gases led to an increase in global mean temperature by 2.2 K in 2030 relative to today, nearly doubling the effect of greenhouse gases alone over this period.⁹⁸ 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.⁹⁸

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,¹⁵⁹ the impacts of tighter vehicle emission standards,¹⁶⁸ ethanol *versus* gasoline fuel use in U.S. fleet,¹⁶⁹ the implications of global and regional sulfur limits and the use of biofuels in shipping,^{170,171} and hydrogen vehicle fuel.^{172,173} Reducing particulate emissions from residential cooking across the developing world would have strong health benefits, however the co-benefits¹⁷⁴ or tradeoffs with climate need further investigation.

2.4 Terrestrial biogeochemical – air pollution feedbacks

Climate impact assessments have largely neglected feedbacks between air pollutants and biogeochemical cycles.⁷⁴ O₃ deposition on vegetation suppresses CO₂ uptake by the land or vegetation sink.¹⁷⁵ One study suggests the magnitudes of the O₃ direct RF and the O₃ indirect RF through CO₂ are comparable.³³ 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 the magnitude of aerosol loading.^{176–178} It is feasible that anthropogenic aerosol may have further substantial effects on the CO₂ land sink, by altering the surface energy budget^{179–181} and the amount of precipitation, thereby influencing water stress in vegetation.¹⁸² Atmospheric deposition of sulfate, nitrate and carbonaceous aerosol to surface ecosystems may impact the carbon cycle but these feedbacks are highly uncertain.

Because terrestrial biogenic emissions are climatically driven, multiple feedbacks can occur between climate, the emissions, and therefore air quality. For example, biogenic VOC emissions are dependent on radiation, temperature and soil moisture.⁴⁸ Atmosphere–biosphere feedbacks through biogenic aerosols have been proposed in the literature,^{183–185} 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 impacts of these climate-driven pathways on surface O₃ and PM. Finally, land-use changes have also been shown to influence regional air quality by altering emissions from the biosphere.^{186–189}

3. Air quality response to a warming climate

There are several pathways by which climate change may influence O₃ 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.*, ref. 190 and 191). For example, over the eastern United States and Europe, observations demonstrate that extreme air pollution (O₃ and PM_{2.5}) is typically associated with air stagnation events.^{190,192–198} 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 O₃ and aerosol sources, such as wildfires, dust, and biogenic precursors and from changes in chemical and depositional sinks.^{199–205} 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 plant stomata.²⁰⁶

To understand the processes by which O₃ and PM in surface air are influenced by changes in climate, several approaches are used (as reviewed by Jacob and Winner⁷):

(1) Sensitivity studies in which individual meteorological parameters are perturbed (*e.g.*, ref. 207 and 208);

(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.*, ref. 209–211); and

(3) The direct calculation of air quality by various global and regional modeling approaches that include:

(i) Fully coupled global chemistry–climate models (CCMs; Table S1, ESI†).

(ii) Fully coupled regional CCMs;^{215–217}

(iii) Global-to-urban CCMs (*e.g.*, ref. 218);

(iv) Off-line global or regional chemical transport models (CTMs) forced using projections of meteorological fields from separate atmosphere–ocean general circulation models (AOGCMs) (*e.g.*, ref. 8) or dynamically downscaled meteorology (*e.g.*, ref. 219 and 220).

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 in space and time and are non-linear.^{6,194,205,221–226} Statistical downscaling based on synoptic conditions may provide more accurate projections,^{190,191,210,227} though models must credibly simulate changes in these synoptic conditions and it is unclear if current statistical relationships are applicable to future climate conditions.

3.1 Ozone

In the case of surface O₃, it is well established that in many polluted regions, high-O₃ events correlate strongly with temperature due to associations of temperature with stagnation episodes, with enhanced photochemistry, and with biogenic and wildfire emissions.^{5–8,186,195,200,221,223,228–232} Table 1 summarizes current understanding of potential impacts of a warming climate on surface O₃ and distinguishes between baseline surface O₃ levels (defined as those not influenced directly by local emissions²³³) and local-to-regional (LR) responses. Baseline surface O₃ is very likely to decrease in a warmer climate because higher water vapor abundances will enhance O₃ destruction in low-NO_x regions of the atmosphere and lead to shorter O₃ lifetimes.²³⁴ The increase in temperature itself has little impact on O₃ in these regions.²²⁸ Climate-driven increases in lightning NO_x and exchange of stratospheric O₃

into the troposphere^{17,235,236} could oppose the negative feedback from rising humidity in some regions of the atmosphere, but are unlikely to offset the humidity-driven decrease in global surface O₃.^{236–238} 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 O₃ associated with higher atmospheric water vapor abundances.^{8,223,239} On large spatial scales, Doherty *et al.*²³² find little response of intercontinental transport pathways themselves to a warming climate, but note enhanced sensitivity of O₃ 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 O₃ response to climate change as estimated by various modeling systems in Fig. 3 (blue bars and symbols). A major caveat is that many studies 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.*²²³). The ranges in Fig. 3 reflect spatial variability as well as differences across models, including in simulation length, scenarios, and reported O₃ 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 zero²⁴⁰ are the net sum of opposing influences from a warming climate: lower baseline O₃ levels but higher O₃ in polluted regions and seasons.^{7,234} These results are plotted alongside several studies that report spatial ranges for daytime statistics during the high-O₃ season. Fig. 3 shows that climate change-induced increases in surface O₃ 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-O₃ 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

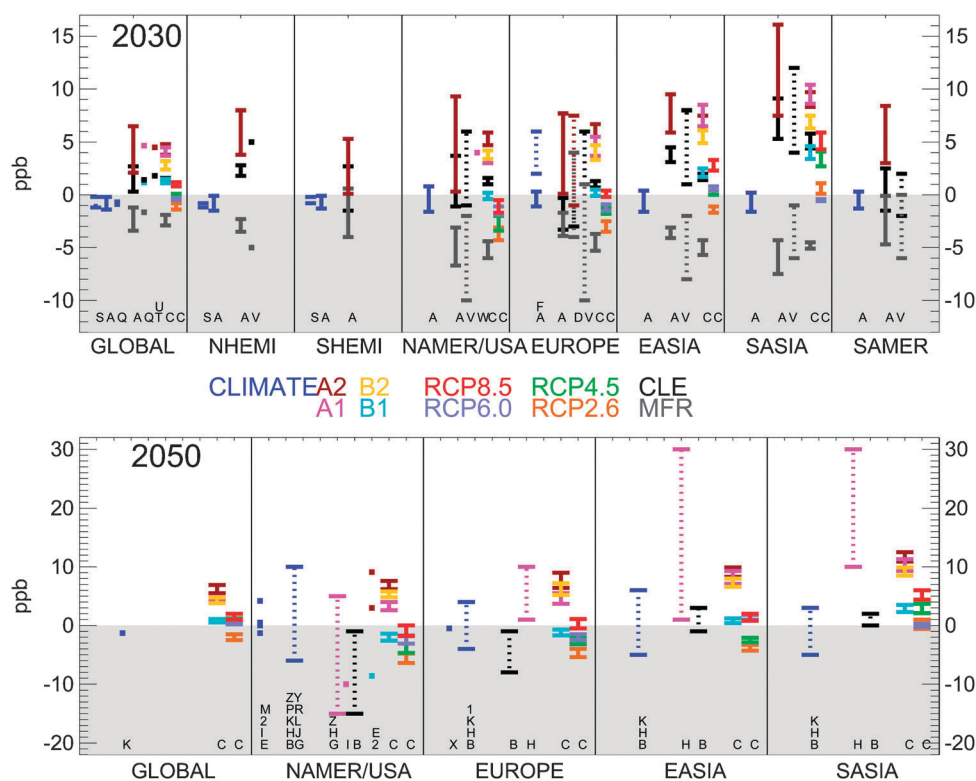


Fig. 3 Changes in surface O₃ (ppb) due to climate change alone (dark blue) or emissions changes alone (colored by emission scenario) reported in the literature in 2030 (top) and 2050 (bottom) for selected world regions. Results from individual studies are labeled by letters underneath the corresponding plot symbols. Solid 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 (A²⁴⁰); (2) application of a parameterization developed from the multi-model ensemble of the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) regional source-receptor relationships to estimate surface O₃ response over the TF HTAP continental regions to emission changes (C⁶⁶); (3) spatial averages across a region, denoted by filled squares (E,¹⁸⁶ I,²⁴⁴ K,²⁴² M,²²⁰ Q,²⁶⁷ T,⁵⁸ U,⁶⁸ W,²⁸ X,²⁸⁷ 2²⁸⁶); (4) spatial ranges across a region as estimated with one model or combined across several individual modeling studies, denoted by dotted lines (B,²⁷⁸ D,²⁸³ F,²²⁴ G,⁸ H,²³⁹ J,²²³ K,²⁴² L,²⁴⁵ P,²⁸⁶ R,³⁶³ S,²⁴⁶ V,²⁹ Y,²⁹⁸ Z,³⁰⁰ 1²⁸¹). 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, but the O₃ changes shown here are self-consistent. Climate change scenarios vary across studies, but are combined into ranges denoted by blue bars because there is little detectable cross-scenario difference in the climate response, particularly in 2030, and many of these estimates are based on simulations that are too short to differentiate cleanly the climate change signal. Tables S2–S5 in the ESI[†] contain details on the plotted values.

Western United States in 2050, though in some regions, such as the Northeastern United States in summer, they consistently show O₃ increases.^{6,7}

3.2 Particulate matter

Particulate matter is also influenced by many of the same processes as O₃ (Table 1), but additional complications include opposing influences on the various PM components.^{7,194,241} Climate-driven changes in PM can be large, but they are highly uncertain and model-dependent (Table 1).⁷ Aerosol burdens are particularly sensitive to precipitation changes and are expected to decrease in regions with increased precipitation.^{186,241–246} Regional and seasonal changes in precipitation must be considered since global soluble aerosol burdens do not typically scale with global precipitation changes.^{98,247} Changes in mixing depths and ventilation of the continental boundary layer also contribute to the sign of the aerosol changes but are highly uncertain.^{7,219,222,248}

Large feedbacks are possible from “natural” aerosol sources (Table 1), particularly carbonaceous aerosols from wildfires, mineral dust, and biogenic precursors to secondary organic aerosol.^{202–204,229,249–253} In regions experiencing warming and drying, wildfires are expected to increase under climate change.²⁵⁴ Dust events are known to affect urban areas seasonally, increasing hazardous levels of PM_{2.5} and reducing visibility in regions downwind of major desert source regions, and in some cases, leading to long-range transport across oceans.^{255–258} As with other types of particulate matter, dust can provide a surface for heterogeneous reactions, leading to enhancement²⁵⁹ and uptake of pollutants which can alter the optical properties and solubility of the primary emission.^{260,261} 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%²⁵⁰ to 60%,²⁴⁹ with the latest estimate around 25%.²⁶² Primary biogenic aerosols are emitted to the atmosphere in the form of pollen, bacteria, spores, or plant fragments,^{263–265} though knowledge is limited as to their contribution to PM_{2.5} concentrations or how they will change with climate.⁵

Individual aerosol species respond differently to meteorological changes. Rising temperature and water vapor enhance SO₂ oxidation relative to surface loss, increasing sulfate aerosol but decreasing nitrate aerosol.^{241,242,248,266–269} One study, however, shows that observed correlations of temperature and water vapor with sulfate, nitrate, and organic carbon largely reflect their co-variation with synoptic transport rather than these direct impacts.¹⁹⁰ While warmer temperatures should reduce the partitioning of gas phase secondary organics into the aerosol phase, *e.g.*, ref. 202, they may also increase the biogenic contribution to PM.^{187,243,244,270} SOA formation pathways are strongly influenced by climate as shown in multiple regional and global modeling studies.^{137,202,271} Future changes in oxidant levels can influence PM distributions,³² Leibensperger *et al.*²⁷² point out that intercontinental influences of NO_x and CO emissions on PM can exceed those from SO₂ emissions,²⁷³ 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.^{196,197,231,274} Even under limited global mean warming scenarios, the frequency of heat waves may increase.^{275–277} A warmer climate is projected to decrease rainfall over southern Europe, creating wintertime deficits that lowers soil water content for the following seasons, contributing to extreme summertime heat wave conditions.²⁷⁸ A few studies have emphasized positive feedbacks from vegetation (higher emissions and lower stomatal deposition) during heat waves.^{197,278,279} Furthermore, the probability of wildfires, a source of pollutant emissions, increases with temperature (Table 1).

Although the record lengths of surface O₃ and PM observations are increasing and allow for new analyses on connections between extreme pollution events and meteorological conditions, additional work is needed to determine their relevance to the response to changing climate. For example, over the Northeastern United States, Leibensperger *et al.*¹⁹⁵ report a strong inverse relationship between cyclone frequency in summer, and the number of air stagnation events conducive to high-O₃ episodes and Tai *et al.*¹⁹⁴ find that stagnant days over the United States are associated with a 2.6 μg m⁻³ increase in PM_{2.5} in the United States. Appelhans *et al.*¹⁹¹ find similar correlations between winter PM events in Christchurch, New Zealand and daily to inter-annual variations in synoptic conditions.

Several modeling studies for the United States and Europe indicate that high-O₃ events are likely to increase in frequency and duration with climate change^{8,196,202,209,220,221,223,224,230,244,279–286} but the large variability in the incidence of these events makes projections of their changes highly uncertain^{195,284} and models often disagree at the regional level.^{6,7,228,248,287} Analysis of centuries-long simulations with CCMs, such as those recently completed for CMIP5 (*e.g.*, the four models at the bottom of Table S1, ESI†), should permit an assessment of whether a climate change signal on pollution episodes can be cleanly isolated from climate variability.

4. Projections of regional air quality changes over the next century

The local response of surface O₃ and PM to climate-driven changes depends also on changing spatial patterns of pollutant emissions (*e.g.*, ref. 140, 141, 194, 205, 207, 285 and 288) and thus is tied to the future emissions trajectories for greenhouse gases and short-lived pollutants. We consider here projections for regional air quality driven by changes in climate and anthropogenic emissions, both separately (Fig. 3 and 4) and combined (Fig. 5 and 6). We focus on the multi-model surface O₃ and PM estimates based on the RCP scenarios:^{16–22} the transient climate simulations with interactive chemistry conducted by several CCMs in support of the Fifth phase of the Coupled Model Intercomparison Project (CMIP5;²⁸⁹ lines and shading in Fig. 5 and 6); and the decadal “time slice” simulations in CCMs and CTMs conducted in support of the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP; circles and black vertical bars in Fig. 5 and 6).

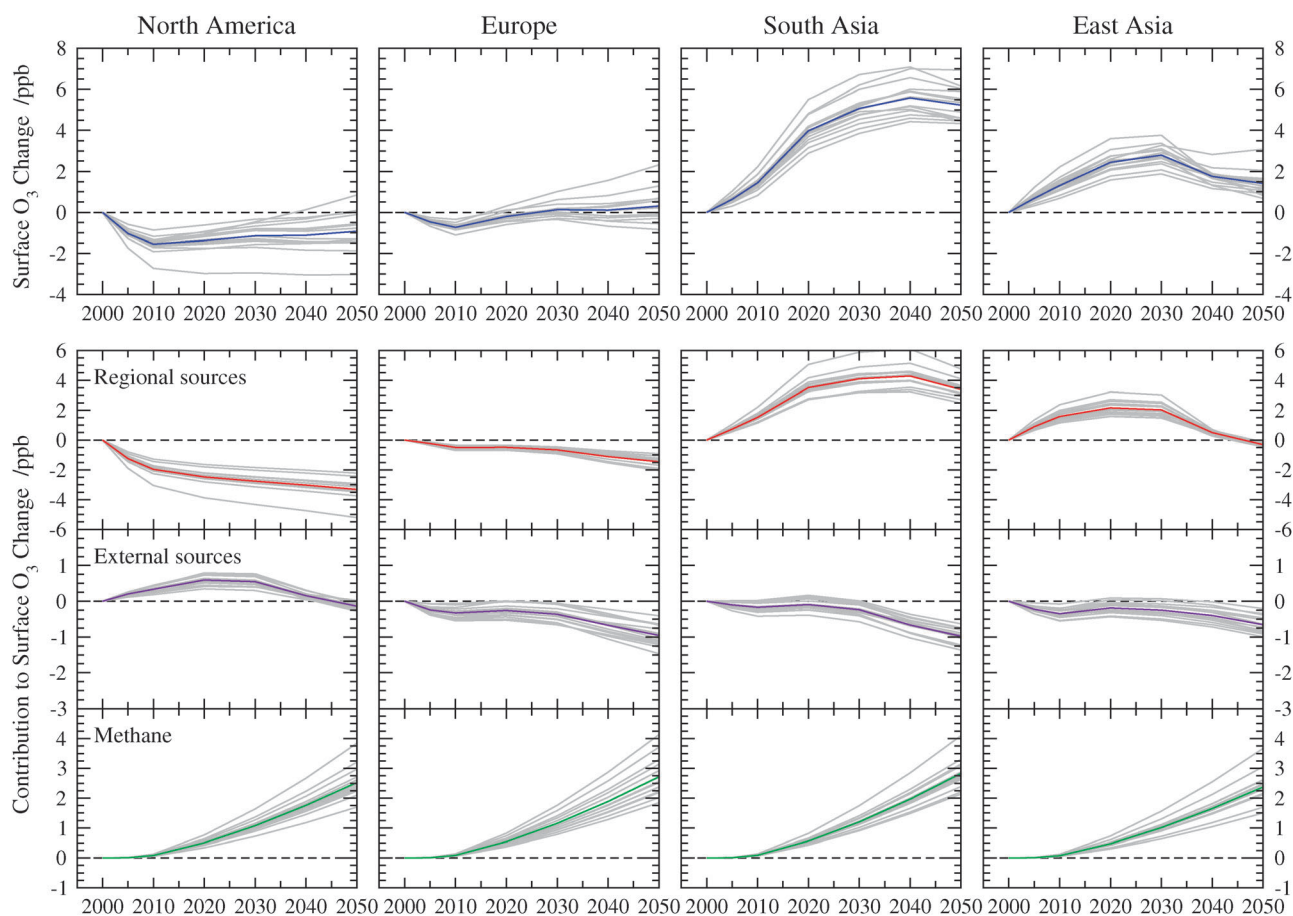


Fig. 4 Individual model (grey lines) and ensemble mean (colored line) spatially averaged O_3 projected from the RCP 8.5 emission scenarios for O_3 precursors from 2000 to 2050 for the four northern mid-latitude source-regions defined by the Task Force on Hemispheric Transport of Air Pollution (TF HTAP).^{66,364,365} The projections are estimated using model sensitivities diagnosed from emission perturbation simulations relative to a base year of 2001 as described by Wild *et al.*⁶⁶ using the CTMs that contributed to the TF HTAP source-receptor studies for surface O_3 .⁶⁵ A source attribution is presented for each region in the lower panels. Reproduced with permission from Fig. 10 of Wild *et al.*⁶⁶

Ongoing analyses of CMIP5 and ACCMIP simulations will inform the upcoming Intergovernmental Panel on Climate Change Fifth Assessment (IPCC AR5) report. The individual models are described in the references in Table S1 (ESI[†]). In addition, some results are shown for the SRES emissions pathways^{14,290} and for alternative, lower pollution pathways than under the SRES.²⁹ Emissions from the biosphere and lightning NO_x generally differ across the models, with some models additionally including climate feedbacks to air pollutant sources and sinks from the biosphere. These CMIP5/ACCMIP results are a first overview; further analysis is in progress.

4.1 Ozone

Projected changes in surface O_3 for the near-term (2030 and 2050) due to changes solely in climate or solely in emissions of O_3 precursors show some general patterns across the globe (Fig. 3). The largest surface O_3 changes under the recently developed RCP (2.6, 4.5, 6.0, and 8.5) emission pathways are much smaller than those projected under the older SRES (A1, A2, B1, B2) scenarios.^{17,66} Alternative, lower pollutant emission scenarios to the SRES scenarios (CLE, MFR)²⁹ tend to cover the RCP range. The RCPs assume aggressive control of NO_x and other O_3

precursor emission controls globally,²³ falling below the current legislation (CLE²⁹) 2030 projections and reaching levels by 2050 that are equivalent to maximum feasible reductions (MFR²⁹). Even within this narrow RCP range for global emissions, larger regional shifts in emissions occur. We conclude that the emission-driven changes across all available scenarios up to 2050 generally span a wider range than the climate-driven changes;²⁴⁰ Fig. 3).

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 emissions²⁹¹). 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.*²⁹² find that global changes in land-use and land-cover (LULC) under the SRES A2 scenario could lead to O_3 changes of $\sim 20\%$ 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.*²⁸⁰ suggest LULC can lead to ± 5 ppb changes in surface O_3 by 2050 in some locations.

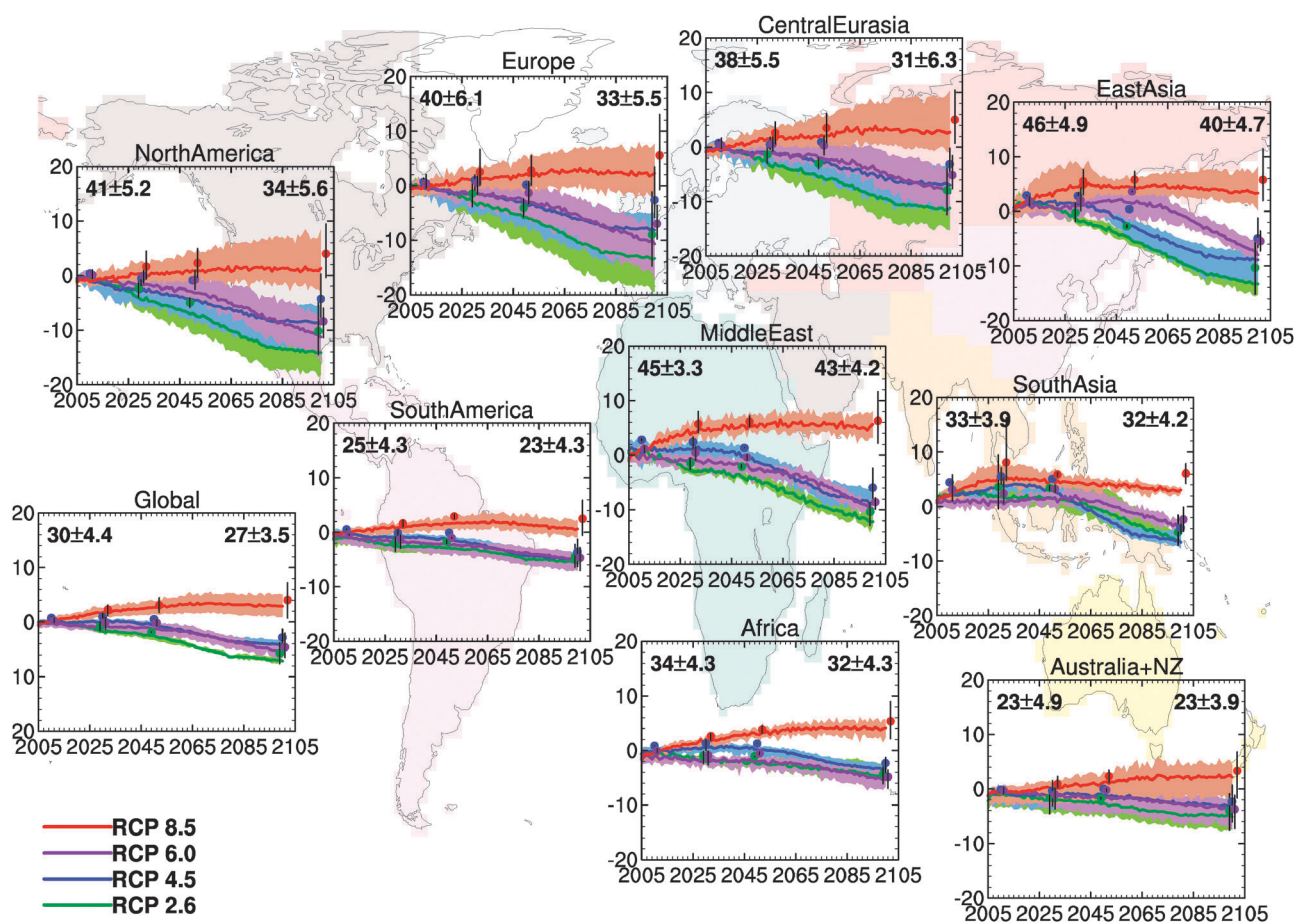


Fig. 5 Changes in annual mean surface ozone (ppb mole fraction) averaged over selected world regions (shaded land regions) following the RCP scenarios. Colored lines denote the 4-model average from transient simulations with CMIP5 chemistry–climate models; shading covers the full range across models (Table S1, ESI†). Filled circles with vertical lines represent decadal multi-model averages from the 2010, 2030, 2050, and 2100 ACCMIP time-slice simulations; the number of models varies with time slice and scenario (Table S1, ESI†). The circles are colored by RCP scenario and offset by a year to clearly distinguish the vertical black lines which denote the full range of ACCMIP model results. Changes are relative to the 1986–2005 reference period for the transient simulations, and relative to the average of the 1980 and 2000 decadal average time slices for the ACCMIP ensemble. The average value for this reference period, averaged across all models is shown in each panel, with the standard deviation reflecting the 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.

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 regional air quality. Rising global emissions (particularly NO_x and CH_4) can offset O_3 decreases obtained with regional emission controls, *e.g.*, ref. 28 and 66. 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 (Fig. 4).⁶⁶ 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.^{38,58,65,66} 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 (Fig. 4). Future increases in shipping emissions, which are subjected to less stringent regulations than land-based emission sectors, may also contribute to raising baseline pollution levels in some regions.^{293,294} Both rising baseline O_3 levels and

a warming climate could contribute to lengthening the O_3 pollution season.^{28,223,282}

Fig. 5 shows projected annual mean surface O_3 changes over the 21st century for selected world regions with the current generation of chemistry–climate models using the RCP trajectories. Large inter-annual variations are evident, reflecting model-generated climate variability, with larger variability over smaller regions, as well as within specific seasons, and in the frequency of high- O_3 events (not shown). The models span a fairly wide range of O_3 responses, consistent with prior studies,^{57,65,240} so we focus here on the robust features. Under all of the RCP scenarios, global NO_x emissions decline over the next century (Fig. 1), decreasing surface O_3 in all regions by 2100 with the notable exception of RCP8.5. Comparison of Fig. 5 (which includes changes driven by both emissions and climate) with previous published RCP emission-only projections to 2050 (see Fig. 8 of Wild *et al.*⁶⁶ and Fig. 4 for RCP8.5) suggests that over many regions, the range in surface O_3 projections is dominated by emission changes. This comparison

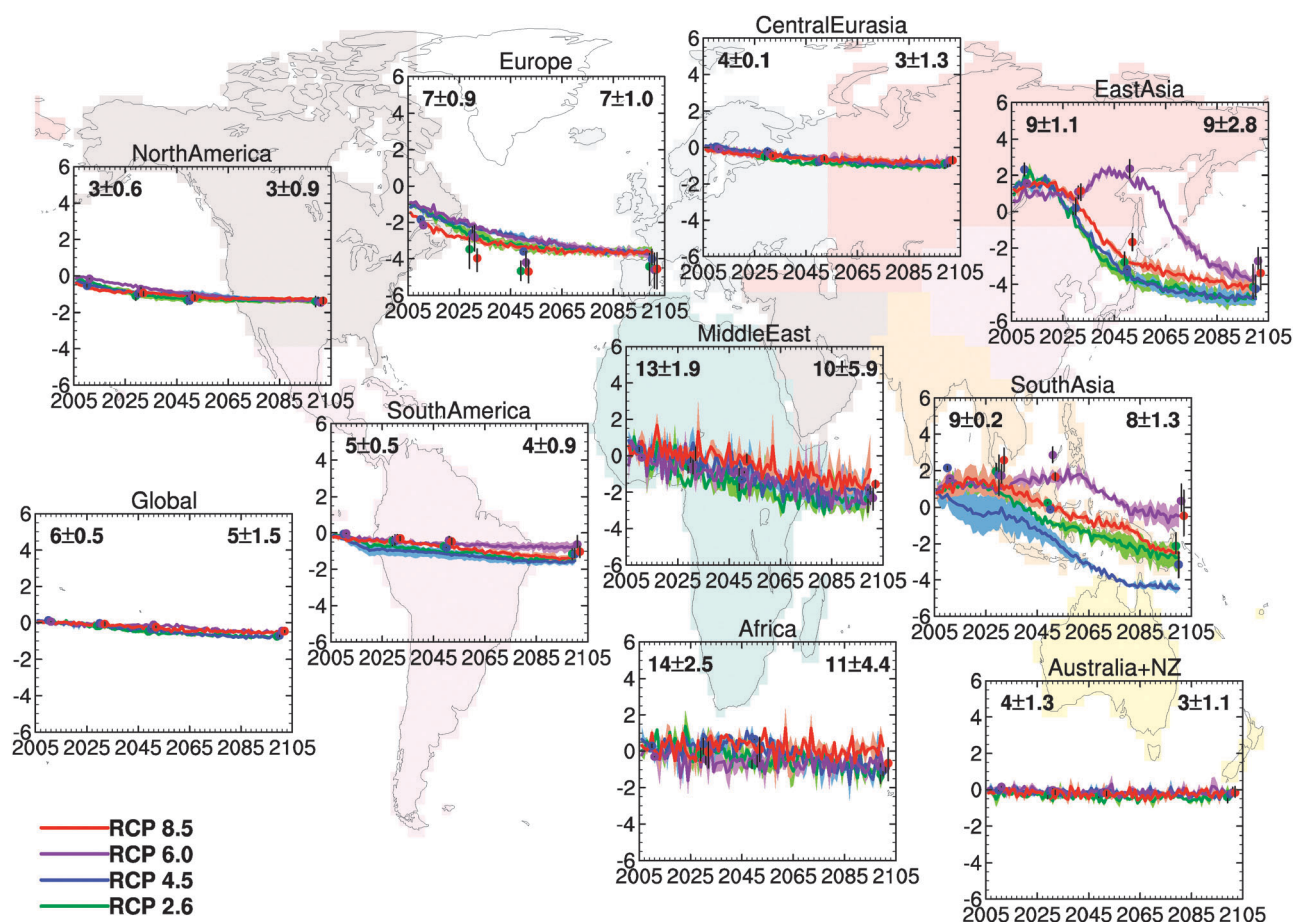


Fig. 6 Changes in annual mean surface $\text{PM}_{2.5}$ (ng per g-air) averaged over selected world regions (shaded land regions) following the RCP scenarios. See Fig. 5 caption for details. $\text{PM}_{2.5}$ estimates are from the italicized models in Table S1 (ESI[†]), calculated as the sum of individual aerosol components (black carbon + organic carbon, + sulfate + secondary organic aerosol + $0.1 \cdot \text{dust}$ + $0.25 \cdot \text{sea salt}$). Nitrate was not reported for most models and thus is not included here.

is limited, however, by different regional definitions (Fig. 4 includes oceanic boxes whereas Fig. 5 includes spatial averages over land only) and the use of different models (CTMs in Fig. 4 vs. mostly CCMs in Fig. 5). Under RCP8.5, the rise in CH_4 (Fig. 1) increases surface O_3 levels (Fig. 4). The strong warming in RCP8.5 may induce additional discrepancies between Fig. 4 and 5 by altering both regional O_3 production and baseline levels, e.g. ref. 8, 221, 232 and 239.

4.2 Particulate matter

Compared to O_3 , we find fewer 21st century projections of PM distributions over the next century. Those that exist are difficult to compare due to variations in study region and reported PM metrics, with some studies reporting only specific PM components (sulfate, nitrate, black carbon, organic) and sizes.^{31,186,241,242,244–246,267,295–301} Future PM, like O_3 , is expected to be driven by changes in both emissions and climate.

PM changes driven by local-to-regional anthropogenic emissions are complex, depend on regional oxidant levels (e.g., ref. 32 and 248), and cannot simply be scaled to changes in regional emissions. Nevertheless, changes in sulfate aerosol concentrations generally follow changes in SO_2 emissions. Changes in NO_x emissions influence nitrate aerosols to a lesser

extent than the SO_2 -sulfate relationship due to competition between sulfate and nitrate for ammonium, such that nitrate aerosol is inversely dependent on sulfate. Increases in ammonia (NH_3) emissions in conjunction with SO_2 reductions enhance nitrate aerosol, offsetting some of benefit from SO_2 controls.²⁴¹ Continued reductions in SO_2 emissions alongside rising NH_3 emission could lead to nitrate aerosol levels equivalent to or larger than sulfate aerosol levels in some regions over the 21st century.^{295,301} Projected changes in U.S. aerosol for 2050 are dominated by anthropogenic emission changes except in regions where large precipitation changes are projected^{186,241,243,245} or those that are heavily impacted by wildfires.²⁰⁴ Future growth in anthropogenic aromatic emissions could lead to a larger anthropogenic contribution to secondary organic aerosol.¹⁸⁷ Carmichael *et al.*²⁹⁹ project that changes in BC and SO_2 emissions from Asia over the period 2000 to 2030 will result in increased $\text{PM}_{2.5}$ concentrations over the region.

Regional $\text{PM}_{2.5}$ projections over the 21st century obtained with the CMIP5 and ACCMIP CCMs and CTMs are shown in Fig. 6. Over most of the globe, and in some industrial regions (North America, Europe, and Central Eurasia), an overall decline in $\text{PM}_{2.5}$ is projected, with little difference across the four scenarios. The particularly noisy projections over Africa, the Middle East, and to some extent Australia,

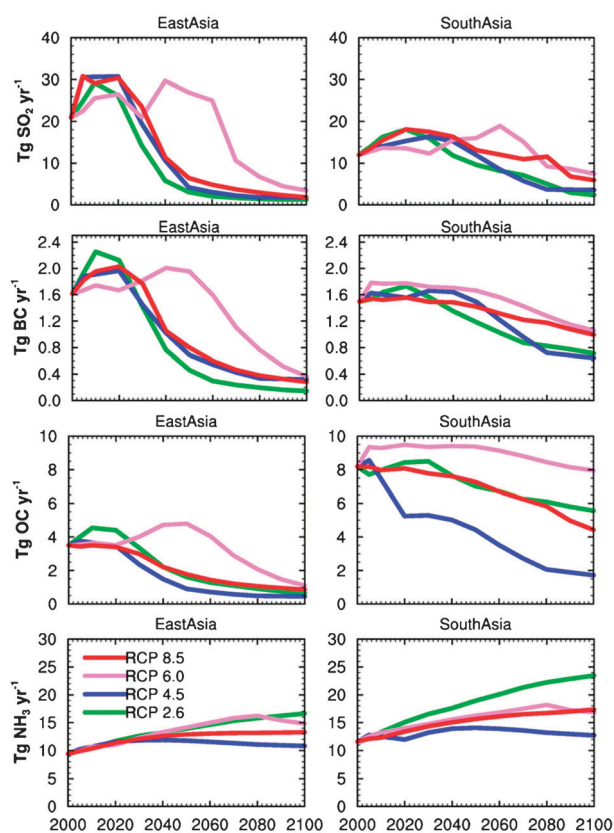


Fig. 7 RCP emissions (2000–2100) of aerosols and precursor emissions from anthropogenic plus biomass burning sources, averaged over two of the regions in Fig. 6 and 7 (East Asia, left column; South Asia, right column).

reflect the dominance of dust sources in these regions and the strong dependence on inter-annual meteorological variability. Over South and East Asia, the $\text{PM}_{2.5}$ projections do not rank according to the RCP RFs. Instead, RCP6.0 represents the high end of the $\text{PM}_{2.5}$ range and RCP4.5 represents the low end. Over South Asia, the $\text{PM}_{2.5}$ projections are strongly tied to the OC emissions (Fig. 7). South Asian SO_2 emissions decline similarly under all of the RCPs, with the exception of RCP6.0 in which SO_2 emissions may contribute to raising $\text{PM}_{2.5}$ relative to the other RCP scenarios from 2050 to 2070 (Fig. 7). Over East Asia, the different $\text{PM}_{2.5}$ projections likely reflect a combination of the changes in SO_2 and carbonaceous aerosols (Fig. 7).

5. Bridging knowledge gaps

Multi-decadal and multi-century simulations with fully coupled chemistry–climate models as summarized in Fig. 5 and 6 offer a new approach to study interactions between climate and air quality. In particular, these models provide new opportunities 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. Some of the models also include a well-resolved stratosphere, enabling assessment of the impact of stratospheric changes, including ozone depletion and recovery, on tropospheric ozone and the tropospheric oxidizing capacity. 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.*, ref. 30, 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, with the full model range spanning a factor of two.⁶⁶ Narrowing this range likely requires better constraints on global NO_x distributions;^{38,58} new space-based approaches may be particularly useful.^{302–308}

Black carbon mitigation is another approach under consideration to improve air quality and lessen global warming, with several emission control strategies identified³⁰ 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.^{38,309,310} 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 salient.

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 (Fig. 3–6). Growing record lengths of historically observed relationships between relevant meteorological variables and air quality provide useful information for evaluating models (*e.g.* ref. 194, 205, 226, 311–314) 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 O_3 and aerosols could aid in characterizing model bias in RF estimates.^{76,83,315,316} These observational approaches, combined with multi-model analysis that seeks to link differences across models to specific processes^{317–319} 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 O₃ to climate change is strongly dependent on the treatment of organic nitrates (RONO₂), specifically those from isoprene.^{6,7,320} The rate at which RONO₂ reacts to recycle NO_x is poorly understood^{321,322} and ranges from 0–100% are used in CCMs and CTMs; this range affects the sign of the O₃ response to changes in BVOC emissions.^{323–326} Chemical mechanisms that include RONO₂ formation without recycling NO_x *via* subsequent reaction display little biogenic VOC-ozone-climate sensitivity,⁸ and mechanisms that ignore RONO₂ altogether show large biogenic VOC-ozone-climate sensitivity.²⁴² 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.*,³²⁴ and newer findings^{327,328}). Knowledge of BVOC oxidation and subsequent SOA formation is advancing rapidly but uncertainties remain.^{54,132,140,329} Aerosol–oxidant interactions also require further study as they may determine PM air quality in some regions.^{32,272}

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 both pre-industrial and present-day O₃ budgets and tropospheric oxidizing capacity.^{330–335} Tropospheric halogen chemistry is not included in the model projections reviewed here (Fig. 3–6).

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 models^{87,130} 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 evaluate 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 directions.³³⁶

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.*, ref. 6, 7 and 337). Improved understanding of the spatially resolved global climate response as compared with the regional climate response to locally changing O₃ and aerosols from specific emission control strategies (including effects on the hydrologic cycle, temperature and circulation) is needed, particularly for the NTCFs in order to bypass current limitations in the global 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 impossible.³³⁸

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 patterns.^{108,111,112,339–341} 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.³⁴² Regional precipitation may be particularly sensitive (both sign and magnitude) to aerosol forcing location.¹³¹

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.*, ref. 13 and 199). The attribution of O₃ and PM air pollution between “anthropogenic” and “biogenic” sources is complicated by chemistry that involves both anthropogenic and biogenic precursors for both O₃ and aerosol,^{141,343} and by land-use changes which alter biogenic sources (*e.g.*, ref. 344 and 345). A major uncertainty regarding BVOC feedbacks is the opposing influences of rising CO₂ *versus* rising temperature.³⁴⁶ Sources from agriculture and livestock sectors are generally poorly constrained but non-negligible, particularly for CH₄³⁴⁷ and NH₃.³⁴⁸ Human-driven changes in land-use and land cover, such as urbanization or shifts between forests and agriculture, could dramatically alter future O₃ and aerosol precursor emissions from the biosphere^{187,189,279} as well as dry deposition,¹⁸⁸ and should be considered in future assessments.

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