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COMMENTARY
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This article is a commentary on Fiore et al. (2022), https://doi.org/10.1029/2021JD035985.

Key Points:

- Several main sources of uncertainty impact projections of future air quality
- Earth System Models with interactive chemistry are computationally expensive, hence understanding which sources of uncertainty are most important is crucial
- The capability to explore and partition the uncertainty in multi-model responses for future air quality assessments is being realized

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Projections of Future Air Quality Are Uncertain. But Which Source of Uncertainty Is Most Important?

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Abstract Understanding how air pollution events may change in the future is of key importance to decision makers. Multi-model intercomparison projects focusing on atmospheric chemistry and air quality have been performed to inform the latest IPCC assessments. Future anthropogenic emission changes have generally been the foci of such model experiments, envisaged as the dominant driver of future atmospheric composition. The latest model assessments such as AerChemMIP utilize multi-model ensembles but also have limited individual model ensembles which permit different sources of uncertainty to be characterized. The recent study by Fiore et al. (2022, https://doi.org/10.1029/2021JD035985) specifically considers a multi-model and multi-member ensemble approach. It adds to the quantification of uncertainty in future projections through delineating uncertainty due to model diversity and due to internal or natural climate variability within the climate system, for mean and high PM2.5 air pollution events over the Eastern USA in the 21st century. Exploring the separate roles of internal climate variability and model diversity adds further value to the important research issue of quantifying how future anthropogenic climate change impacts air quality. Future multi-model intercomparisons need to balance the additional knowledge gained from research into understanding multiple sources of uncertainty that can inform decision making vs. the resource costs of performing these experiments using Earth System Models with interactive chemistry.

Plain Language Summary Climate change projections are inherently uncertain. There are several main sources of uncertainty that arise from: (a) future changes in emissions related to human activity, (b) models responding differently to climate change and/or emission changes, (c) natural variability in the climate system. To determine the impact of these sources of uncertainty on future air quality, complex Earth System Models are required which are extremely resource intensive. Therefore, most studies to date have not been able to evaluate all the relevant sources of uncertainty because multiple models and multiple simulations are required to do this. The study by Fiore et al. (2022, https://doi.org/10.1029/2021JD035985) uses results from multiple models and multiple simulations to explore uncertainty due to different model responses and to natural climate variability. In doing so, this study provides a more robust assessment of climate change impacts on air pollution events in the 21st century for the Eastern USA and sets out a framework for understanding how uncertainty impacts on future projections of atmospheric composition.

1. Introduction

Air pollution has adverse impacts on human health. Fine particulate matter ($PM_{2.5}$) and ozone (O_3) are the two main regional pollutants of concern for decision makers who seek to maintain or improve current regional and urban air quality standards now, and into the future. For emission control measures to be effective, the responses of these air pollutants to changes in both emissions and climate and the changing chemical environment need to be ascertained. Models such as global and regional-scale composition-climate models or full Earth System Models (ESMs) can quantify these responses. However, it is important not only to provide a deterministic estimate as our "best guess" of future levels of air pollution but to establish the uncertainty associated with future air pollutant projections by considering the different sources of uncertainty. A burning question is: which sources of uncertainty are the most important for future risk assessment and air quality management?

Anthropogenic and natural emissions together with synoptic-scale meteorology are the major drivers of regional-scale concentrations of both O_3 and $PM_{2.5}$. The latter comprises different inorganic and organic aerosols species with sizes less than 2.5 μm in aerodynamic diameter, whose size has its origins in measurement

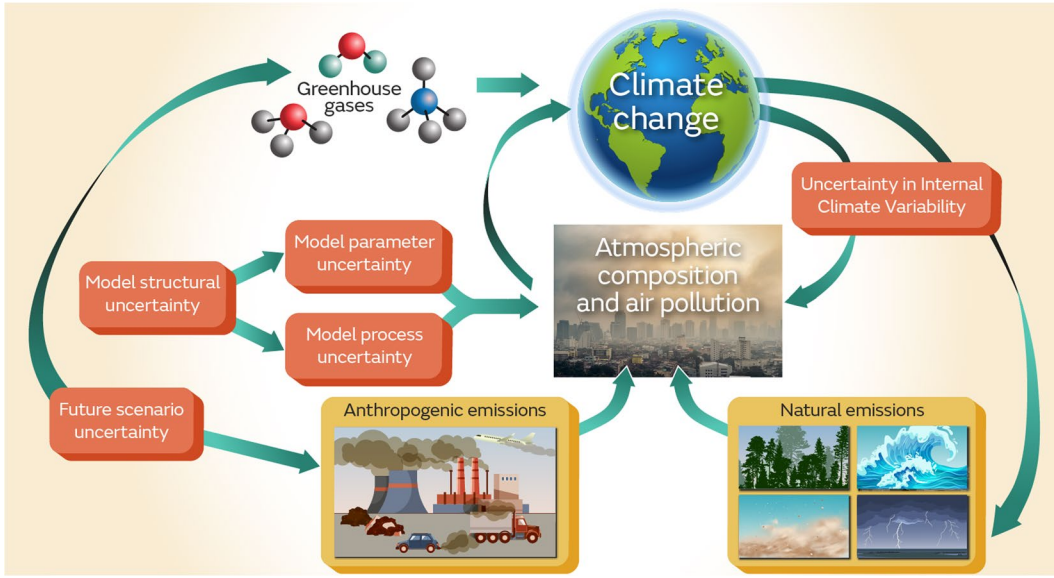

Figure 1. Schematic of atmospheric composition within the climate system and how the different types of uncertainty interact within this system.

capabilities. Its concentrations in the atmosphere arise from primary emissions and secondary formation from gas-phase species through a combination of chemical and physical processes. Meteorology and composition are tightly coupled. Besides controls on composition through large-scale and regional transport and local mixing, numerous chemical processes of production and destruction and deposition rates are influenced by meteorology (e.g., Brasseur & Kumar, 2021). Natural emission rates too are often influenced by meteorology, oceans and the biosphere. Hence, changes in climate influence many different chemistry and aerosol processes (e.g., Fiore et al., 2012; Jacob & Winner, 2009). Changes in composition in turn affect radiative forcing (O'Connor et al., 2021; Thornhill, Collins, Kramer, et al., 2021) and ultimately climate (Allen et al., 2021), with climate change in turn having a feedback on natural emission source strengths (Thornhill, Collins, Olivié, et al., 2021). All these interactions and feedbacks mean that future air quality projections are uncertain.

2. The Main Types of Uncertainty

In climate modeling, coupled model intercomparison projects (CMIPs; e.g., Eyring et al., 2016) designed to inform IPCC assessments, have allowed for advances in the characterization of uncertainty because multiple models performed simulations over the same period and with the same set of emissions scenarios (Lehner et al., 2020). These intercomparisons also provided initialized ensembles. These are simulations performed with the same model and same emissions scenario but varying initial conditions often minutely to enable an assessment of variability arising naturally (internally) in the climate system. The number of models producing initialized ensembles has expanded with more recent CMIP5/6 models (Deser et al., 2020). Hence CMIPs allow for the characterization of uncertainty due to three different sources: scenario uncertainty (sometimes called radiative forcing uncertainty), model or structural uncertainty and uncertainty from internal climate variability (Figure 1). These sources of uncertainty were compared for global and regional mean surface temperature and precipitation from CMIP3 models (Hawkins & Sutton, 2009, 2011) and CMIP5/6 models made available recently (termed Large Ensembles—LEs, Deser, 2020; Deser et al., 2020; Lehner et al., 2020). Whilst there were differences in magnitudes between CMIPs, for global mean temperature, the dominant source of uncertainty by 2100 was scenario uncertainty, whilst for precipitation model uncertainty was dominant except in the near term (Hawkins & Sutton, 2011; Lehner et al., 2020).

Scenario uncertainty arises because of different emission scenarios or pathways constructed to represent a range of technological and economic futures and mitigation (e.g., O'Neill et al., 2014; Rao et al., 2017; Riahi et al., 2017). This affects atmospheric composition directly through emission changes and climate through radiative forcing, which in turn feeds back on composition (e.g., Fiore et al., 2012; von Schneidemesser and Monks, 2013).

Structural uncertainty or the diversity in model responses to any given future emissions scenario occurs because of uncertainty in the representation of complex chemical processes that may depend on chemistry-aerosol-meteorology interactions. This type of uncertainty arises from our incomplete understanding of physical processes (known as process uncertainty) as well as imperfect representation of known processes also known as parameter uncertainty (Deser, 2020). To address the latter, a perturbed parameter ensemble (PPE) approach has been used, in which parameters within a single model are varied and the model response constrained by observations to produce a range of feasible model realizations. These approaches have been used for studies investigating climate sensitivity and aerosol radiative forcing (e.g., Johnson et al., 2020; Murphy et al., 2004; Sanderson et al., 2008). Due to computational challenges, these have not yet been widely used in CMIPs. However, recently, these techniques have been applied to atmospheric composition to study scenario and parameter uncertainty in a small number of models. By using a PPE and Gaussian emulation approach, albeit exploring a small number of model parameters and model inputs, Wild et al. (2020) found different models had different rankings of the most important processes driving tropospheric ozone concentrations. With a greater focus on air pollution, a recent assessment constructed a perturbed emissions ensemble to constrain NOx emissions estimates for Beijing (Yuan et al., 2022).

The other main source of uncertainty, internal climate variability, as outlined above is derived using individual members of an initial condition ensemble. Such an ensemble allows the potential contributions from climate change and natural climate variability on projected climate and air quality to be estimated. Another major application of initialized ensembles is to determine a robust anthropogenic climate change signal, and hence inform approaches in the field of the detection and attribution of anthropogenic climate change. At its simplest, the anthropogenic climate change "signal" is represented by the ensemble mean whilst the internal climate variability "noise" emanates from the initialized ensemble range (Fiore et al., 2022).

When modeling future air quality, studies have largely focused on scenario uncertainty and uncertainty due to diversity in model response. Brasseur and Kumar (2021) explain that future air quality studies have tended to neglect uncertainty due to internal climate variability because traditional atmospheric chemistry transport models do not exhibit chaotic behavior. The complexity of chemistry schemes with hundreds of species and chemical reactions have meant that only recent advances in computing capabilities have led to ESMs with detailed interactive chemistry being used increasingly. The climate component of the ESM is based on numerical weather prediction for which the dynamical equations representing the atmospheric conservation laws are nonlinear and chaotic, resulting in them being highly sensitive to initial conditions. Hence ESMs too exhibit internal climate variability due to chaos in the atmosphere. This means that for modeling future climate and air quality, ESM studies should also consider uncertainty due to internal climate variability, in addition to scenario and model uncertainty.

3. Uncertainty Assessment in the Latest Model Intercomparisons of Atmospheric Composition

The Aerosol and Chemistry Model Intercomparison Project (AerChemMIP) uses the latest generation of ESMs to assess the impact of aerosols and chemically reactive gases on air quality and climate (Collins et al., 2017). AerChemMIP was set up to investigate the impact of future policies on air quality and climate, the historical radiative forcing from anthropogenic emissions and the importance of climate feedbacks. The experimental design and research undertaken builds on previous multi-model ESM intercomparison projects (e.g., Aerocom—Textor et al., 2006; TF-HTAP—Doherty et al., 2013; ACCMIP—Lamarque et al., 2013; CCMI—Eyring et al., 2013). In terms of uncertainty assessment, the results from AerChemMIP have focused on uncertainties due to different emission scenarios and due to model diversity (i.e., scenario and structural uncertainty) often providing a combined uncertainty estimate. A more limited assessment of the uncertainties associated with internal climate variability has been undertaken. Due to the number and range of experiments proposed, as well as the inclusion of chemistry and aerosols within ESMs means AerChemMIP was one of the most computationally demanding model intercomparison projects endorsed by CMIP6. Each climate modeling center made decisions on how much time and resources to commit to each set of AerChemMIP experiments, potentially limiting the number of models participating, experiments conducted and availability of data with which to investigate the different types of uncertainty. Nevertheless, in the case of the coupled simulations for AerChemMIP, at least three initial condition ensemble members were requested (Collins et al., 2017).

Multi-model results from AerChemMIP simulations have been used to study the change in chemical composition and air pollution over the historical and future time periods. Studies on tropospheric O_3 and OH responses found inter-model differences or structural uncertainty in both the historical and future responses was due to differences in tropospheric chemistry and physics schemes (Griffiths et al., 2021; Stevenson et al., 2020). When analyzing surface air quality changes, Turnock et al. (2020) showed that CMIP6 models exhibited considerable structural uncertainty in both historical and future responses, especially for certain regions. Large model diversity was also reported in these models when calculating effective radiative forcings (ERFs) from reactive gases and aerosols, as well as the climate feedbacks on composition, highlighting the large structural uncertainty in processes between models (Thornhill, Collins, Kramer, et al., 2021; Thornhill, Collins, Olivié, et al., 2021). In these studies, the different sources of uncertainty were not intrinsically separated. Specifically considering uncertainty in internal climate variability, Allen et al. (2020, 2021) studied the response of atmospheric composition and climate to the mitigation of near-term climate forcers in ESM simulations across a maximum of 3 ensemble members from each model that conducted them, finding that structural uncertainty is comparable to the uncertainty in internal climate variability across individual ensemble members in the near term (2050s) and at the end of the century (2100). However, this was a limited sample as not all models provided data from multiple ensemble members.

There exists the potential to further exploit results from multi-ensemble AerChemMIP simulations to provide a quantification of the uncertainty in internal model variability in both past and future projections, in addition to scenario and structural uncertainty. One of the major issues arising from these large multi-model intercomparison exercises is how to design an appropriate set of experiments that can extract the relevant useful information for risk assessment, as well as a comprehensive assessment of all the uncertainties. In particular, the time, cost (computational, people and data storage) and feasibility of participating in these projects needs to be considered by individual modelling centers. Hence there is a trade-off to be considered in terms of adequate simulations for the characterization of different sources of uncertainty and the number of experiments that can be performed.

4. New Perspectives on Uncertainty for Air Quality Events

The assessment of uncertainty associated with internal climate variability has not been the primary focus of previous multi-model studies focusing on atmospheric composition, although it has started to emerge in recent studies (e.g., Allen et al., 2020, 2021; East & Garcia-Menendez, 2020; Fiore et al., 2022; Garcia-Mendenez et al., 2017; Pienkosz et al., 2019). The Fiore et al. (2022) study explores the impact from both structural uncertainty and internal climate variability on future mean and high air pollution levels in the Eastern USA using ensembles from two models. As the authors state, the use of multi-models with multi-ensemble members to gauge the significance of changes in high pollution events in light of internal climate variability is novel. They find that the structural uncertainty is large over much of the Eastern USA leading to differences in the direction of change in summertime mean $PM_{2.5}$ concentrations except in the northeast region between the two models. In this northeast region, the climate change signal is significant compared to internal climate variability, with increases in $PM_{2.5}$ levels of 1–4 μgm-3 in 2100 compared to present-day across the two models. The authors highlight the complexity of meteorology-chemistry interactions that may lead to structural uncertainty, finding that $PM_{2.5}$ concentration changes do not scale with temperature and precipitation changes, possibly because of different responses of individual $PM_{2.5}$ components to meteorological variables.

Changes in high $PM_{2.5}$ events are also determined. Fiore et al. (2022) find an increase in high $PM_{2.5}$ days (defined as the upper quartile of $PM_{2.5}$ concentrations) only occurs in both models for the northeast region of the Eastern USA. For this region, one model projects the number of high $PM_{2.5}$ events lasting more than 3 days is significant compared to climate variability for all of the last three decades of the 21st century whilst the other model suggests an increase in 3-day high $PM_{2.5}$ events only occurs in the 2090s. The authors also tentatively explore the impact of using the larger 12-member ensemble on model responses in comparison to using a smaller ensemble size. They suggest internal climate variability plays a role in the discrepancy in model responses to climate change in other studies, hence that structural uncertainty may be overestimated. Despite study limitations, Fiore et al. (2022) conclude that there is considerable model structural uncertainty between these two models, leading to fundamental differences in their responses to climate change.

Overall, Fiore et al. represents one of the few systematic assessments of the impacts due to climate change on regional-scale air pollution that considers uncertainty due to climate variability. However, due to the computational cost of running multi-model, multi-member ensembles, the number of models included was limited to

two, the ensemble size was limited to three members for one of the models, and there were some limitations in model capability. Nevertheless, the two-model ensemble offers scope for further analysis, including improved process-based understanding of differences in the model responses and the different drivers in the two models. In light of evidence that climate change may extend poor air quality events over the Eastern US beyond the traditional summer period (e.g., Zhang & Wang, 2016), there is also potential for extending the analysis into the spring and autumn periods, as well as extending the approach to other regions and other pollutants (e.g., O_3).

5. Outlook and Future Needs

Most studies of atmospheric composition and air quality to date have focused on scenario and structural uncertainty as the main sources of uncertainty for future air quality. But few studies have assessed uncertainty due to internal climate variability due to the limited availability of multi-member ensemble model simulations. The Fiore et al. (2022) study shows a promising step forward toward quantifying separately structural uncertainty and uncertainty due to internal climate variability, to understand the likely impact and potential ranges of climate change impacts on regional air quality events. It also highlights the challenges in experimental design when balancing computational costs against the need for sufficient characterization of uncertainty to be useful for decision makers. In their study, two models were used to explore structural uncertainty but only one of these included interactive chemistry, and for that model only three ensemble members were available. This contrasted to a relatively large 12-member ensemble for the other model with prognostic aerosol but no interactive chemistry. But this was enough to gain insights into regions of the Eastern USA where robust assessments of climate change impacts could be made, as well as regions where model process insights and developments to reduce uncertainty were needed.

Whilst the Fiore et al. study quantified robust changes relative to the uncertainty from climate variability, key uncertainties in future air quality projections remain through structural uncertainties and limitations in current ESM capability. One such limitation is nitrate aerosol—a key component of secondary inorganic aerosol in air quality and climate forcing (e.g., Hauglustaine et al., 2014)—which was generally not included in CMIP6 models. Other limitations in the Fiore et al. study, and more generally in the current generation of ESMs, are the omission of interactive wildfires, and the treatment of other natural emissions (e.g., biogenic volatile organic compounds, dimethyl sulfide, dust), whose responses to climate change have been found to differ both in magnitude and sign across models (e.g., Thornhill, Collins, Olivié, et al., 2021). By advancing model capability through the inclusion of new processes and improving the understanding of these processes, structural uncertainty can potentially be reduced (e.g., Hawkins & Sutton, 2009). However, Carslaw et al. (2018) argue that faster progress would be made if structural uncertainty quantification approaches were incorporated into model development. Although multi-model large PPEs may not be tractable for multi model ESM studies, rapidly growing novel machine learning approaches (e.g., Liu et al., 2022) may be more feasible.

Therefore, the pertinent question for future model intercomparisons is what sources of uncertainty are most important? There is no easy answer. It depends on the future time horizon, the magnitude of the climate change signal, the air pollutant species of interest, the region of study, and model capabilities. For air quality and atmospheric composition, there is the substantial difficulty associated with the computational cost of models using detailed chemical mechanisms compared to physical climate models. This high computational burden demands well thought out experimental designs that question the number of models and the number of ensemble members required for robust atmospheric composition and air quality assessments. There is also the added challenge of how multiple lines of evidence from future climate and air quality assessments, such as CMIP7, can be combined together from multi-model ensembles, single-model PPEs, and other tools such as emulators. Nevertheless, as Fiore et al. state, multi-model multi-member ensembles could transform the scientific community's capacity to develop probabilistic assessments of future changes in regional-scale pollution event frequency and duration, thereby helping to address policy maker and societal needs.

Data Availability Statement

Data were not used, nor created for this research.

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