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# Interactions between atmospheric composition and climate change - Progress in understanding and future opportunities from AerChemMIP, PDRMIP, and RFMIP

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**Abstract.** The climate science community aims to improve our understanding of climate change due to anthropogenic influences on atmospheric composition and the Earth's surface. Yet not all climate interactions are fully understood and diversity

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in climate model experiments persists as assessed in the latest Intergovernmental Panel on Climate Change (IPCC) assessment report. This article synthesizes current challenges and emphasizes opportunities for advancing our understanding of climate change and model diversity. The perspective of this article is based on expert views from three multi-model intercomparison projects (MIPs) - the Precipitation Driver Response MIP (PDRMIP), the Aerosol and Chemistry MIP (AerChemMIP), and the Radiative Forcing MIP (RFMIP). While there are many shared interests and specialisms across the MIPs, they have their own scientific foci and specific approaches. The partial overlap between the MIPs proved useful for advancing the understanding of the perturbation-response paradigm through multi-model ensembles of Earth System Models of varying complexity. It specifically facilitated contributions to the research field through sharing knowledge on best practices for the design of model diagnostics and experimental strategies across MIP boundaries, e.g., for estimating effective radiative forcing. We discuss the challenges of gaining insights from highly complex models that have specific biases and provide guidance from our lessons learned. Promising ideas to overcome some long-standing challenges in the near future are kilometer-scale experiments to better simulate circulation-dependent processes where it is possible, and machine learning approaches for faster and better sub-grid scale parameterizations where they are needed. Both would improve our ability to adopt a smart experimental design with an optimal tradeoff between resolution, complexity and simulation length. Future experiments can be evaluated and improved with sophisticated methods that leverage multiple observational datasets, and thereby, help to advance the understanding of climate change and its impacts.

## 1 Introduction

A central aim of climate science is to advance our understanding of how the Earth system responds to human activities. This endeavor involves the assessment of numerous aspects in the Earth system, which consists of multiple, interacting components. On a timescale of several decades, the Earth's temperature is controlled by a balance between the net amount of absorbed sunlight (solar radiation) and the radiation emitted by the planet and its atmosphere (terrestrial radiation). A perturbation of this balance is called a "radiative forcing" - a concept embedded in the study of the physical basis of climate (Ramaswamy et al., 2019). Radiative forcing may be caused by changes in atmospheric composition, including for instance concentrations of greenhouse gases such as carbon dioxide and methane and the burden of aerosols, as well as by changes in surface albedo or irradiance.

Changes to atmospheric composition have distinct effects on the Earth's energy budget and climate, which are classified into radiative forcing, climate response, and feedbacks. The direct impact of a change in atmospheric composition on radiation fluxes is the instantaneous radiative forcing (IRF). Re-equilibration occurs when the system responses yield a new surface temperature at which the net top-of-atmosphere fluxes are in balance when averaged over several decades. Climate responses can be amplified or weakened via positive or negative feedbacks that are induced by changes in physical and chemical processes. Balancing the system after an initial perturbation can take several hundred years depending on the magnitude of the perturbation because of the slow response of ocean temperature. There are also fast processes influencing the flux differences that arise from a change in atmospheric composition, even in the absence of surface temperature changes. Such changes, known as

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rapid adjustments, occur in the atmosphere. Examples are stratospheric cooling due to increasing carbon dioxide concentrations (Manabe and Wetherald, 1967), changes in clouds due to circulation changes (e.g. Gregory and Webb, 2008; Bretherton et al., 2013; Merlis, 2015), and chemical adjustments due to changes in emissions of reactive trace gases (Thornhill et al., 2021b; O'Connor et al., 2021). Effective Radiative Forcing (ERF) is the sum of the IRF and the rapid adjustments, whereas diagnosing climate responses requires an assessment of changes in the fully coupled atmosphere-ocean response contributing to surface temperature changes. These steps in the perturbation-response paradigm of climate science are schematically depicted in Figure 1.

Understanding and quantification of the different steps in the perturbation-response paradigm of climate science are typically derived through experiments with Earth System Models (ESMs, e.g., Heavens et al., 2013). Modern ESMs vary in their design and their level of complexity for representing physical, chemical, and biological processes. For example, some ESMs prescribe aerosol properties while models with additional process-complexity can simulate aerosol and their precursor emissions, transport, and deposition of aerosols (Figure 1). The climate modeling community collaborates regularly to produce multi-model ensembles of a common set of ESM experiments. However, even if ESM experiments have the same perturbation applied, the modeled climate response can differ. This diversity in response may be due to differences in process complexity within the respective ESMs, and/or may be due to the design and coupling of different model components. Ensembles of ESM experiments following the same experimental protocol aim to understand reasons for the diversity in climate responses and feedbacks, and to create future climate projections.

Results from multi-model intercomparison projects (MIPs) are widely used for both advancing scientific understanding and for informing stakeholders on climate change. The most prominent example is the experimental protocols of the Coupled Model Intercomparison Project (CMIP, Meehl et al., 2000) that have informed the assessment reports of the Intergovernmental Panel on Climate Change (IPCC), e.g., the sixth phase of CMIP (CMIP6, Eyring et al., 2016) informed the sixth IPCC assessment report (IPCC-AR6). The principal idea of MIPs is also used for different foci either outside of or endorsed by the CMIP consortium. For example, the Aerosol Model and Measurement Comparisons (AeroCom) focuses on the role of aerosols in the climate system(e.g., Glißet al., 2021; Textor et al., 2006), the Chemistry-Climate Model Initiative (CCMI) on the interactions between atmospheric chemistry and climate change (e.g., Morgenstern et al., 2017; Abalos et al., 2020), and the Precipitation Driver Response Model Intercomparison Project (PDRMIP, Myhre et al., 2017) on the role of anthropogenic and natural drivers for different precipitation responses. Several MIPs were endorsed during CMIP6, such as the Aerosol and Chemistry MIP (AerChemMIP, Collins et al., 2017) and the Radiative Forcing MIP (RFMIP, Pincus et al., 2016). While the specific foci for AerChemMIP and RFMIP varied, both the MIPs were driven by the common goal of better characterizing the preindustrial to present day radiative forcing and determining climate responses to these forcings.

The aims of this article are to synthesize and emphasize what has been learned on the experimental design, conceptual thinking, and diagnostic tools through connecting the scientific communities of the three MIPs: AerChemMIP, RFMIP and PDRMIP, under one umbrella named TriMIP (Figure 2). Each of the MIPs had their own perspective on how to accomplish their goals, but sufficient similarities inspired a series of joint TriMIP meetings. Similar conceptual understanding and overlap in diagnostic tools helped to build common ground across the community that proved useful to contribute to the same overarching

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goal – the advancement in understanding of our planet's changing climate. In addition to synthesizing the scientific advances made through TriMIP, the article discusses the challenges with understanding multi-model climate responses and identifies potential opportunities to make further advances in this area.

## 2 Advancement through MIP's cross-linkages

The three MIPs sought to advance the understanding of modern climate change due to anthropogenic influences considering structural differences between ESMs. While the MIPs share the conceptual idea of the perturbation-response paradigm (Figure 1), they focus on different components in the paradigm. RFMIP focuses on an improved understanding of the role of radiative forcing diversity for the climate response to anthropogenic perturbations in atmospheric composition (e.g., Smith et al., 2020a), and PDRMIP on precipitation responses to idealized atmospheric composition changes (e.g., Richardson et al., 2018). AerChemMIP also focuses on quantifying radiative forcing and responses. However, AerChemMIP starts earlier in the paradigm since all participating models simulate atmospheric composition based on emissions, transport, chemical transformations, and deposition, making these models more complex in their process representation and interactions (e.g., Thornhill et al., 2021a). The three MIPs use, to some extent, similar experimental strategies inspired by each other, but developed and adopted their own experimental protocol with a certain class of model in mind. Taken together, this has led to ensembles of ESM experiments of different complexity, model resolution, number, and length in the three MIPs.

A major advancement from the synergy between the three MIPs was the widespread adoption of the same method to quantify radiative forcing that facilitated easier comparisons across CMIP6. Estimates of ERF are key in the perturbation-response paradigm through characterizing the magnitude of a perturbation in the radiation budget, yet a consistent calculation of ERF was not possible in CMIP5 (Collins et al., 2017). Specifically, RFMIP helped to establish a consistent practice for diagnosing ERF for CMIP6 and related activities, building on experiences from PDRMIP (Forster et al., 2016). Amongst several approaches to quantify forcing, graphically summarized in Figure 3, there are two methods widely used now to estimate ERF from models. Firstly, ERF can be estimated by extrapolating the relationship between the radiation imbalance and temperature change in coupled atmosphere-ocean model experiments subject to abrupt concentration increases of the forcing agent (Regression method, Gregory et al., 2004). Secondly, ERF can be determined by suppressing ocean-temperature changes in atmosphere-only experiments and calculating the ERF as the radiation imbalance relative to an experiment without the forcing agent (Fixed sea-surface temperature method, Hansen et al., 2005). In this context, the parallel use of pre-industrial atmosphere-only control experiments in RFMIP and AerChemMIP, i.e., experiments where the atmospheric composition represents the values in 1850, proved valuable as a common reference to estimate ERFs from ESMs in CMIP6. RFMIP further requested results from diagnostic calls to the radiation schemes, also known as double radiation calls, that enabled calculations of the IRF (Chung and Soden, 2015). Such model diagnostics for IRF helped to quantify the contribution of adjustments to ERF estimates in CMIP6 models (e.g., Smith et al., 2020a).

The three MIPs benefited from being embedded in a landscape of other initiatives, with close connections to CMIP on the one hand and specialist MIPs like AeroCom on the other hand. The community of PDRMIP, AerChemMIP, and RFMIP can therefore be seen as a bridge between the global climate modeling community of CMIP6 and the specialized communities for aerosols and atmospheric chemistry that do not participate in CMIP. This setting allows CMIP to benefit from expert knowledge that would otherwise be missing. One example is PDRMIP, which began before CMIP6, and had a guiding role for the later MIPs concerning the practice of estimating ERF, the parallel use of fully coupled and fixed SST experiments, the choice of perturbation magnitudes and experiment length to quantify forcing and response, as well as the introduction of new model diagnostics. Another example is AerChemMIP, which adopted recommendations for the diagnostic requests and experimental design (e.g., Young et al., 2013; Archibald et al., 2020) from previous non-CMIP6 initiatives.

Finally, coming together of the three MIP communities under the TriMIP umbrella facilitated efficient communication of knowledge gaps and coordination of analysis of multi-model output to address these gaps resulting in publications in peer reviewed journals. Since several authors of the IPCC-AR6 also participated in TriMIP, the MIP-based publications were more relevant to the needs of the IPCC WGI AR6 including analysis of ERF (Smith et al., 2020a; Thornhill et al., 2021b), non-$CO_2$ biogeochemical feedbacks (Thornhill et al., 2021a), and climate (Allen et al., 2020, 2021) and air quality responses (Turnock et al., 2020) to changes in short-lived climate forcers.

## 3 Challenges in the MIP's research

A major challenge to further advance the understanding of climate change is that model differences in the components along the perturbation-response paradigm are not independent of each other. For instance, ESMs can have a different forcing for the same change in atmospheric composition, and forcing differences imprint on the climate response in addition to various feedback mechanisms influencing the response. This challenge is addressed by the three MIPs through suppressing interactions for one component in the perturbation-response paradigm to advance the understanding in another component. In this regard, the joint strength across the three MIPs is the restriction of model diversity in some parts in order to better characterize and ultimately understand model diversity in others. Methods to separate out some of these model differences include experiments using, for instance, prescribed aerosols (e.g. Fiedler et al., 2019) or reactive trace gases (e.g. Checa-Garcia et al., 2018), which makes the assessment of the contribution of different processes to model diversity more tractable. Specifically, PDRMIP provided aerosol information that models prescribed to circumvent some aerosol-related sources of model-to-model diversity. Such an experimental design allows a deeper exploration of a subset of components contributing to model diversity - in this case, the translation of aerosol concentration to radiative forcing and the climate response, by removing other sources of model differences. Along similar lines, AerChemMIP allows for chemical processing of aerosols and reactive gases, but removed feedbacks by performing atmosphere-only experiments. Finally, RFMIP aimed to understand how much of the climate response to a perturbation is due to changes in atmospheric composition rather then due to feedbacks. To that end, RFMIP only simulates the atmosphere and aerosols with prescribed sea-surface conditions to obtain precise model estimates of ERF. The three MIPs, therefore, addressed model differences arising from different components of the perturbation-response paradigm in a complementary manner.

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### 3.1 Computational Capacity Abyss

Available computational capacity affects MIP design and priorities at modeling centers performing many model experiments for diverse MIPs on short timescales. Not all experimental choices are explicitly defined by the MIP's experiment protocols, giving modelers room to make their own choices. Taken together, there were inevitable tradeoffs in the exact experimental designs. Such choices can be categorized along the three axes of (1) *model complexity* addressing how many process interactions ESMs allow or how much fidelity processes have, (2) *model resolution* referring to the grid spacing of the model, and (3) *simulation length* covering the length and number of simulations in an ensemble of experiments per ESM. These axes, schematically depicted in Figure 4, span a triangle in the complexity - resolution - length space. The area of the triangle scales with the computational demand of the experiments for an ESM and is limited by the computation capacity abyss, i.e., the available computing capacity at the modeling center. The computational demand does not scale linearly along all axes such that the abyss is more quickly reached for some experimental designs. Doubling the simulation length or number doubles the required computational resources that are needed, but this is not true for the model resolution and complexity. Increasing the model resolution by a factor of two for instances requires computational resources that are an order of magnitude larger.

Although increasing computing power has become available, tradeoffs along the three axes of experimental design are necessary, especially in light of the computational cost of interactive chemistry and/or competition for priority of experiments at modeling centers. Experiments with the most complex ESMs are necessary to understand interactions of chemical species in concert with climate change, for example, the carbon cycle or atmospheric composition-climate interactions. To that end, ESM experiments are performed that have interactive aerosol and chemistry schemes in addition to the fully coupled atmosphere-ocean-land system, making these models the most complex. Their computational demand for simulating many processes restricts the scope for increasing computing resources along the other two axes of experimental design: performing a large number of experiments, which allows the impact of model-internal variability to be reduced; and choosing a fine enough spatial resolution, which explicitly resolves more physical processes on the model grid. For some research questions, the complexity of ESMs can be reduced to a degree, while retaining sufficient process detail for the scientific problem that is to be studied. It makes creating large ensembles of ESM experiments possible such that model-internal variability can be separated from the mean radiative forcing (e.g. Forster et al., 2016; Fiedler et al., 2017), climate responses (e.g. Maher et al., 2019; Deser et al., 2020), and impacts on air quality (e.g. Garcia-Menendez et al., 2017; Fiore et al., 2022).

High process complexity, although desirable and needed, but also poses a challenge for reducing uncertainty in the assessment of the climate response to various forcings. AerChemMIP emphasized two such challenges. One challenge concerns the diversity in the level of complexity included in the ESMs, which arises due to choices made for the number of interacting processes, the representation of chemistry and aerosols, as well as the specification of the spatial resolution. As an example, this diversity is clearly evident in the complexity of aerosol processes with some CMIP6 models simulating emissions, transport, and deposition of different aerosol species (e.g. Mulcahy et al., 2018), while other models prescribe aerosols such as the spatial distribution of their optical properties (e.g. Mauritsen et al., 2019). Such differences in model capabilities and experimental setup have implications for understanding why their results differ (e.g. Wilcox et al., 2013). The second challenge

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comes from the consideration of model diversity in the design of a MIP, i.e., some models can simulate processes that others cannot. MIPs already have a specific class of models in mind, yet the experimental protocols do not necessarily specify the desired target range for model resolution or a guide for all processes to be switched on or off. These choices are made by the modelers. To some degree, this freedom is well justified since ESMs might otherwise not be able to participate and an ensemble of ESMs is needed to sample climate responses considering structural model differences. A full exploration of the role of climate-composition feedbacks, however, remains an outstanding challenge due to this difficulty.

### 3.2 Process Understanding Abyss

There are limits in advancing climate science with today's most complex ESMs since not all processes are represented or known. This process understanding abyss additionally restricts what can be simulated with even the most comprehensive ESMs (Figure 4). Some known chemical reactions and species, as well as their interactions, are not represented represented differently across ESMs, such that their relevance for climate is difficult to assess. For example, nitrate aerosols are not represented by all ESMs in CMIP6 but are available in some models (e.g., Zaveri et al., 2021). AerChemMIP showed that including previously missing interactive sources of chemical species in an ESM has the potential for surprising results in estimates of forcing (Morgenstern et al., 2020). Primary organic aerosols can be represented by some ESMs (e.g., Burrows et al., 2022a), but marine volatile organic compounds (VOCs) other than dimethyl sulfide (DMS) are not. Also, natural primary biological aerosol particles (PBAPs), such as bacteria, pollen, fungi and viruses (Szopa et al., 2021), are not simulated by ESMs, although PBAP emissions might increase with future warming (Zhang and Steiner, 2022) with potential health impacts. Both DMS and PBAPs are thought to aid in forming clouds; effects of such ice-nucleating aerosols on clouds is an area where more progress is needed (Burrows et al., 2022b).

Of the three MIPs, AerChemMIP played a unique role in the quantification of non-$CO_2$ biogeochemical feedbacks (Thornhill et al., 2021a), illustrated in Figure 5. Almost all non-$CO_2$ biogeochemical feedbacks are negative and therefore counteract warming. The only exception is the positive feedback from methane wetland emissions that amplifies warming and is the largest in magnitude compared to the other non-$CO_2$ biogeochemical feedbacks. The positive feedback from wetland emissions may be partly offset by the negative feedback of the methane lifetime. However, due to their potential magnitude, these methane feedbacks are important yet uncertain. Together with the large model-dependent feedback for biogenic VOCs, the multi-model mean feedback is negative, but the uncertain methane feedbacks give rise to the large spread in the total non-$CO_2$ biogeochemical feedbacks ranging from positive to negative. Not all potentially relevant chemistry-climate feedbacks are yet simulated, e.g., climate-induced changes in fire activity. Although some CMIP6 models represented fire dynamics, they did not fully include the interaction with atmospheric chemistry (e.g., Teixeira et al., 2021). And of those feedbacks that are simulated, model consensus and smaller in magnitude might suggest they are irrelevant but this could be misleading. Dust is one such example. The small dust feedback shows little model-to-model difference, but dust trends differ considerably across ESMs so much so that they are of opposite signs (e.g., Kok et al., 2023). This points towards insufficient process-based understanding of dust-aerosol changes with warming.

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Of those processes that are simulated, a large driver in model diversity for atmospheric composition is thought to stem from the representation of natural processes (e.g., Séférian et al., 2020; Zhao et al., 2022). ESMs simulate, for instance, different historical trends for $O_3$ and aerosols (Mortier et al., 2020; Griffiths et al., 2021). AerChemMIP further points to model differences in the concentrations of secondary organic aerosols (Turnock et al., 2020) and CMIP6 models show historical trends of different signs and magnitudes for desert-dust aerosols (Bauer et al., 2020; Thornhill et al., 2021a). A better understanding of trends in aerosol species can help to unravel model diversity in the evolution of aerosol forcing over time, and how it is related to time-dependent temperature biases in CMIP6 models (Flynn and Mauritsen, 2020; Smith and Forster, 2021; Smith et al., 2021; Zhang et al., 2021). In particular, a better understanding of natural aerosols in the rapidly warming Arctic may be a key factor in resolving the puzzle of Arctic amplification (Schmale et al., 2021), where diversity across ESMs for short-lived climate forcers is large (Whaley et al., 2022). Another example of the crucial role of representing natural processes is the ability of ESMs to simulate aerosol properties and weather on regional scales. Circulation is a grand challenge for ESMs (Bony et al., 2015), affecting the spatio-temporal distribution of aerosols. Desert-dust aerosols are, for instance, emitted and transported by winds, with a persistently large diversity across ESMs (e.g. Evan et al., 2014; Checa-Garcia et al., 2021; Zhao et al., 2022; Kok et al., 2023). The ability to accurately simulate atmospheric circulation is also relevant to the challenge of realistically simulating clouds and rainfall (e.g. Sperber et al., 2013; Stevens and Bony, 2013; Fiedler et al., 2020; Wilcox et al., 2020), influencing how aerosols can affect clouds and how aerosols are removed from the atmosphere. Sparse or missing observations are also a contributing factor, since they enable freedom in choosing model settings, e.g., used for tuning ESMs to reproduce observables (Mauritsen et al., 2012). Moreover, aerosol optical properties are partially biased (e.g., Brown et al., 2021), the size distributions of different aerosol species are not sufficiently understood (Mahowald et al., 2014; Croft et al., 2021), and model diversity in aerosol optical depth persists across different phases of CMIP and AeroCom (Wilcox et al., 2013; Vogel et al., 2022).

## 4 Methodological Opportunities

There are several opportunities to advance the understanding of climate responses to perturbations in emissions, atmospheric composition, and/or the land surface. These are opportunities to augment traditional ESM experiments through (1) the use of emulators where they are informative, i.e., where a climate response to a perturbation is expected to fall within the solution space of existing ensembles of ESM experiments, (2) the use of novel global kilometer-scale experiments where they are possible in light of the tradeoffs along the complexity - resolution - length axes, (3) the development and application of machine learning across the paradigm to speed up and improve processes in complex ESMs where it is needed, and and finally (4) new and sophisticated model evaluation and analysis methodologies that leverage multiple observational datasets to understand the causes of model diversity. Moreover, there is the opportunity to improve diagnostics of ESM experiments. These concern the further development of the method for radiative forcing calculations, and the revision or new implementation of ESM diagnostics to allow more synergies with impact assessments.

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## 4.1 Augmented ESMs

### 4.1.1 Emulators where informative

Another class of models, known as emulators (e.g., Meinshausen et al., 2011; Leach et al., 2021), reduces computational demand and helps to prioritize new ESM experiments. Emulators are informed by results from existing experiments with ESMs, of which there are now many from several CMIP phases and other MIPs. Unlike ESMs, emulators mimic the behaviour of ESM experiments through fast calculations instead of numerical integration of non-linear physical and chemical equations over time on a three-dimensional grid. Both techniques allow spatially resolved predictions of temperature and other variables, but emulators can do it at massively reduced computational costs compared to ESMs (Beusch et al., 2020; Watson-Parris et al., 2021, 2022). Once established, emulators can be used to explore the climate response to different forcings, e.g., to inform experimental designs of future emission scenarios in CMIP6 (O'Neill et al., 2016). Training emulators requires a broad range of ESM experiments such that they interpolate rather than extrapolate into unseen climate conditions. This training data could be made up of CMIP experiments, an ensemble of idealized experiments (Westervelt et al., 2020), or perturbed parameter ensembles where several ESM experiments with systematically different settings in parameterizations are performed to study sources of model-internal uncertainties (e.g., Johnson et al., 2018; Regayre et al., 2018; Wild et al., 2020). Emulators have been used for some time (Murphy et al., 2004; Lee et al., 2013, 2016; Yoshioka et al., 2019; Johnson et al., 2020; Watson-Parris et al., 2020; Wild et al., 2020) and modern techniques also utilize machine learning to allow validation against observations (Watson-Parris et al., 2021). Emulators can incorporate model spreads similar to output from classical MIPs with ESMs but with substantially less computing costs. Although the difficulty of accounting for non-parametric biases of CMIP models in emulators remains (Jackson et al., 2022), emulators are seen as useful to sample parametric differences and explore climate responses to different forcing agents.

### 4.1.2 Kilometer-scale experiments where possible

Much finer spatial resolutions with horizontal grid spacings of a few kilometers hold the potential to overcome some of the long-standing challenges concerning the representation of clouds, precipitation, and circulation in global climate simulations, which can be enabled by a step change in collaboration between climate science and high-performance computing (Slingo et al., 2022). Such high spatial resolution naturally improves the representation of clouds and precipitation, at least in part, due to better resolved orographic effects on atmospheric dynamics and the explicit simulation of convective cloud systems along with the meso-scale circulation (Oouchi et al., 2009; Berckmans et al., 2013; Heinold et al., 2013; Klocke et al., 2017; Satoh et al., 2019; Hohenegger et al., 2020), although not all model biases are eliminated (Caldwell et al., 2021). For some research questions on atmospheric composition and the associated climate response, kilometer-scale experiments are already used, e.g., for better understanding aerosol-cloud interactions (Simpkins, 2018), which is one of the key uncertainties in ERF from ESMs (e.g., Smith et al., 2020a). The coupling of atmospheric processes with the land improves in kilometer-scale experiments. It can reduce biases in the simulated temperature and precipitation (Barlage et al., 2021), which can help to better understand regional climate change that involve land-mediated feedbacks. Moreover, better resolved ocean dynamics holds the potential

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for surprises in understanding climate responses (Hewitt et al., 2022). Kilometer-scale experiments therefore allow new insights into processes in the Earth system following the perturbation-response paradigm, and can leverage the experiences made with regional kilometer-scale climate experiments for different world regions (e.g., Prein et al., 2015; Liu et al., 2017; Kendon et al., 2019). Although kilometer-scale experiments are presently only possible for limited area domains or globally for restricted time periods of a few weeks to years, they are promising to better simulate clouds, precipitation and circulation. Given the role of resolution in maintaining concentrated emissions, non-linearities in chemistry, and atmospheric transport of pollutants, more kilometer-scale climate change experiments might prove valuable to advance the understanding of climate and air quality interactions. Such experiments within limited area domains would also help to alleviate the computational cost of both high process complexity and high spatial resolution.

#### 4.1.3 Machine Learning where needed

New opportunities arise from machine learning approaches for improving or speeding up process representations in ESMs, as well as designing smart tools for postprocessing and evaluating ESM output. Machine learning is particularly attractive where kilometer-scale experiments are beyond what is feasible for high-complexity ESMs. One example is using deep learning for the design of new parameterizations (e.g., Rasp et al., 2018; Eyring et al., 2021; Veerman et al., 2021). Proofs of concept from single ESMs exist. Atmospheric chemistry parameterizations can, for instance, be replaced by fast representations based on machine learning (Keller and Evans, 2019; Shen et al., 2022). The causes of multi-model diversity highlighted in previous studies (Young et al., 2018; Mortier et al., 2020; Griffiths et al., 2021) can also be elucidated using machine learning. There is an increase in the availability of globally gridded fused model-observation data products (e.g., Randles et al., 2017; Buchard et al., 2017; Inness et al., 2019; Betancourt et al., 2021; van Donkelaar et al., 2021; Betancourt et al., 2022) that can be used as benchmarks in model evaluation of atmospheric composition. Novel aspects of such benchmarks include providing data relevant to health impacts (e.g., DeLang et al., 2021) and using machine learning techniques for global mapping of atmospheric composition (e.g., Betancourt et al., 2022). Liu et al. (2022) used deep learning to quantify the sensitivity of surface $O_3$ biases to different input variables in a CMIP6 model (UKESM1), thereby providing new understanding of biases without the computational cost of perturbed parameter ensembles and enabling postcorrection for surface $O_3$. Similarly, such approaches have been used to improve our understanding of model diversity in other aspects of atmospheric composition, e.g., surface particulate matter (PM, Anderson et al., 2022). Including necessary variables for such algorithms in the model output of future MIPs can enable a multi-model intercomparison of different contributions to model biases and provide bias-corrected data for future projections of changes that can be tailored towards impact studies, e.g., concerning future air quality and human health.

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### 4.2 Improved diagnostics and analyses

#### 4.2.1 Radiative Forcing Calculation

The concept of radiative forcing is central to the perturbation-response paradigm for understanding climate change (e.g. Sherwood et al., 2015), in which a radiative forcing is eventually balanced by a temperature response mediated by feedback processes. Definitions of radiative forcing have evolved over time to allow an increasingly wide range of different forcing agents or contexts for perturbations to be considered interchangeably (Figure 3). Early definitions of radiative forcing focused on changes in the net radiation at the tropopause, ideally after the stratosphere had adjusted to a new radiative equilibrium in the presence of the forcing agent (stratospherically adjusted radiative forcing, Hansen et al., 1997). These definitions have been generalized in the concept of ERF (ERF, see Sherwood et al., 2015). ERF, measured at the atmosphere's boundaries, encompasses both the effects on radiation fluxes due to a forcing agent (IRF) and the contributions from rapid adjustments, that refer to flux-modulating changes in the system driven by IRF in the absence of surface temperature changes.

Quantifying IRF for ESMs is desirable, even if the IRF of the forcing agent is constrained by other methods. There is little fundamental uncertainty for IRF of $CO_2$ changes, as indicated by errors on the order of a fraction of a percent from the most accurate line-by-line radiative transfer models (Pincus et al., 2020). However, due to the high computational demand, ESMs do not compute the radiation transfer with a line-by-line model. Instead, they rely on parameterizations, speeding up the computation at the expense of accuracy. Consequently, a model spread in IRF occurs despite so little fundamental uncertainty. For instance, a spread in $CO_2$ IRF has persisted across CMIP phases and accounts for a majority of the model spread in the $CO_2$ ERF (Chung and Soden, 2015; Soden et al., 2018; Kramer et al., 2019; Smith et al., 2020a). Quantifying the model's IRF, e.g., with double calls of the radiative transfer calculations, is particularly relevant in light of the model-state dependence of IRF (Stier et al., 2013; Huang et al., 2016), referring to ESM differences in atmospheric conditions that affect the radiative transfer. Both CMIP6 models (He et al., 2022) and theoretical arguments (Jeevanjee et al., 2021) suggest that $CO_2$ IRF is correlated with temperature, i.e., $CO_2$ IRF increases as the surface warms and the stratosphere cools. This feedback-like effect on forcing is thought to account for a $\sim$10% increase in $CO_2$ IRF for present-day against pre-industrial (He et al., 2022) and $\sim$30% for quadrupled $CO_2$ (Smith et al., 2020b). It requires clarity in the experimental design and reporting of resulting ERF estimates to disentangle the contributions of forcing from feedbacks in future experiments.

There are several methods to quantify ERF across ESMs that can be further improved and standardized (Figure 3). ERF is often computed from atmosphere-only model experiments using prescribed sea-surface temperatures and sea ice (fixed-SST method; Forster et al., 2016), e.g., adopted in RFMIP (Figure 6). The fixed-SST method has two advantages compared to the traditional approach based on coupled atmosphere-ocean experiments (Gregory et al., 2004). Firstly, the impact of internal variability on ERF estimates is reduced through sufficiently long atmosphere-only experiments (Forster et al., 2016; Fiedler et al., 2017) that are computationally less expensive. Secondly, the use of fully coupled experiments to estimate ERF relies on a linear relationship between ERF and temperature, now known not to be true in general (Armour, 2017; Rugenstein et al., 2020; Smith and Forster, 2021), and can lead to ERF estimates that differ from the results of fixed-SST experiments (Forster et al., 2016). One weakness of fixed-SST experiments to estimate ERF is the adjustment of land-surface temperatures. A change

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in land-surface temperatures affects the energy budget, leading to biased estimates of ERF at the order of 10% (Smith et al., 2020a). Such unwanted influences on the ERF estimate can be post-corrected (Tang et al., 2019; Smith et al., 2020a). If the capability of fixed land-surface temperatures (Andrews et al., 2021) was facilitated in more ESMs, biases in ERF arising from surface temperature adjustments would be eliminated in the future. By adopting the fixed sea- and land-surface temperature method (Figure 3), the change in the radiation budget would then be equal to the change in the energy budget of the system, which overcomes limitations of other methods for estimating ERF.

Model diversity in simulated aerosol burden and optical properties is yet another area where improved diagnostics and sophisticated ways of analysis would be useful for advancing the scientific understanding for the climate response to forcing. As discussed in Section 3, the significant diversity across ESMs in the simulated distributions of aerosol burden, optical properties, radiative effects, and the resulting climate responses, including temperature and precipitation, limit building confidence in model projections of climate change. Analysis of relevant and correlated model diagnostics together with observations in sophisticated ways can shed light on the source of diversity in the full cause-and-effect-chain and inform improvements in the treatment of aerosols in models. For example, Samset (2022) underscores the diversity in aerosol absorption as the dominant cause of model diversity in historical precipitation changes in CMIP6.

#### 4.2.2 Synergies with Impact Assessments

There is the opportunity to increase synergies with impact assessments of climate change through improved model diagnostics, specifically for $O_3$ and PM. A common tropopause diagnostic, included for the first time in CMIP6 models due to AerChem-MIP, was available for the calculation of tropospheric $O_3$ burden in a consistent manner. However, the tropopause height was found to vary across the models, and as $O_3$ is found in large concentrations in the stratosphere and upper troposphere, the tropopause definition contributed to the model spread in the calculated tropospheric $O_3$ burden. Through TriMIP, it was identified that a tropopause defined by the $O_3$ mixing ratio results in a smaller model spread in $O_3$.

Moreover, not all ESMs currently output diagnostics for PM, and those models that calculate PM use different formulas and combination of species. Such differences make any intercomparison of PM between models and observations difficult. To circumvent this issue, AerChemMIP tested (Allen et al., 2020; Turnock et al., 2020; Allen et al., 2021) estimating PM from model output following Fiore et al. (2012), but associated uncertainties are hard to quantify. Future MIPs could standardize calculations for $PM_{2.5}$ and $PM_{10}$ across experiments, e.g., consistent with air quality assessments following the standards of the World Health Organization. It would allow the use of PM measurements for air quality monitoring as an independent validation data set and could create a bridge to health impact studies.

Another opportunity to connect more with impact-oriented research can arise from ESM experiments for additional future socio-economic and mitigation-based pathways such that uncertainty in emission developments, including mitigation, can be systematically explored. Potential examples include a MIP on future methane removal (Jackson et al., 2021) in support of potential climate solutions or fire emission developments possibly accounting for the new capability to represent fire feedbacks (Teixeira et al., 2021). Such future considerations could provide a better link between climate change, weather extremes, air quality, and health impact assessments. Stronger interactions with the Vulnerability, Impacts, Adaptation and Climate Ser-

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vices (VIACS, Ruane et al., 2016) and the Inter-Sectoral Impact Model Intercomparison Project (ISIMIP, Frieler et al., 2017) community could enhance the usage of MIPs for societally relevant problems.

## 5 Conclusions

The existence of TriMIP was coincidental, yet the joint community of three MIPs has proven valuable for advancing the research field on atmospheric composition and associated climate responses. RFMIP helped to establish a consistent practice for diagnosing radiative forcing from CMIP6 models, and having preindustrial experiments across AerChemMIP and RFMIP facilitated the comparability of results. Challenges in advancing the understanding with Earth System Models following the perturbation - response paradigm remain. For instance, the approach works well for understanding temperature responses to radiative forcing, but it seems less satisfying for precipitation responses. In part, this is related to the grand challenge of representing clouds and circulation, which can be addressed with newly evolving capabilities. Moreover, model-state dependencies affecting radiative forcing and climate responses can potentially be reduced or even resolved in the near future. Promising ideas are the use of:

- prescribed land and ocean surface temperatures in Earth System Model experiments to quantify the effective radiative forcing free of artifacts arising from temperature adjustments on land,
- kilometer-scale model experiments with resolutions of a few kilometers to improve the understanding of interactions of atmospheric composition with circulation, clouds, and precipitation which are long-standing challenges in climate modeling with coarse-resolution ESMs,
- novel machine learning approaches to speed up and improve the representation of aerosols and chemistry that will still require parameterizations in kilometer-scale model experiments, and
- sufficiently long or many experiments to distinguish climate responses to atmospheric composition changes from internal variability that avoids ambiguity in responses to anthropogenic perturbations.

The optimal choice for new ESM experiments along the three axes of resolution, complexity, and length is specific to the research question and can be a challenge for modeling centers, especially when several MIPs are simultaneously requesting new experiments. Computationally efficient model code for new computer architectures and smart experimental designs are therefore important when we move outwards along any of the three axes in experimental design. The former can be addressed by even closer collaborations with computer science, which is needed to translate existing ESM codes for use on exascale machines (e.g., Fuhrer et al., 2018). The latter can benefit from information from computationally fast models that emulate results from existing ESM experiments, of which there are now many due to several CMIP phases. Exploiting results from emulators can help to prioritize new ESM experiments, e.g., to perform those that are needed for questions for which the answer is not expected in the solution space of existing experiments. Future model improvements may arise from the generalized aerosol/chemistry interface (GIANT, Hodzic et al.,, in prep.) initiative, which aims at a deeper understanding of dependencies among different components of ESMs.

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The three MIPs have built an international vibrant community that goes on to tackle new research endeavors. First new MIP proposals emerging from the TriMIP community have already been made. The new Regional Aerosol MIP (RAMIP, Wilcox et al., 2022) will explore the role of regional aerosol changes in near-future climate change, drawing on earlier experimental designs to be directly comparable with CMIP6. In parallel to RAMIP, atmosphere-only and coupled atmosphere-ocean experiments with two global models are conducted, in which emissions of nine short-lived climate forcers are varied for sixteen regions. The experiments are unique in the sense that a coarse-resolution model (Tatebe et al., 2019) is compared against a kilometer-scale model (Satoh et al., 2014), both using interactive aerosol and chemistry (Takemura et al., 2005; Sudo and Akimoto, 2007). Such comparisons can help to decide on the level of fidelity needed to study regional responses of air quality and climate to emission changes, in addition to the value of information on mitigation options for stakeholders.

The community of the three MIPs continues to maintain the exchange across the community and interdisciplinary boundaries under the new Composition Air quality Climate inTeractions Initiative (CACTI, cacti-committee@geomar.de). CACTI has the aim to quantify and better understand the global and regional forcing, the climate and air quality responses, and the Earth system feedbacks due to atmospheric composition and emission changes. Through joint strengths and diverse expertise, CACTI strives to contribute to the advancement of the understanding of anthropogenic climate change by adopting the established school of thought of the perturbation-response paradigm with novel methodological tools.

*Data availability.* Data of RFMIP and AerChemMIP are publicly available via ESGF, and data of PDRMIP via WDCC.

*Author contributions.* All authors contributed to the discussion and writing of the manuscript. V.N., F.M.O'C., C.J.S., and S.F. designed the figures. V.N., F.M.O'C., C.J.S., R.P., T.T., W.J.C., M.S., G.M., and S.F. participated in the organization of TriMIP meetings that contributed to the manuscript content. S.F. led the synthesis and writing.

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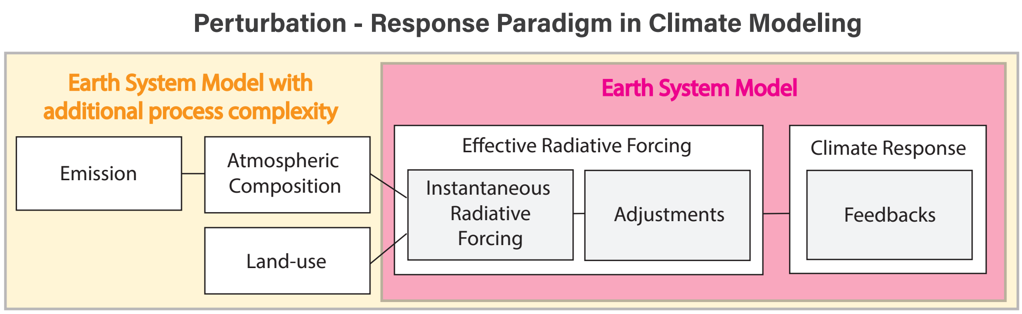


**Figure 1.** Schematic depiction of the perturbation-response paradigm in understanding and quantifying climate changes to perturbations. Marked are Earth System Models (pink) that simulate forcing and response based on prescribed perturbations in the atmospheric composition and land use from experiments of Earth System Models. Earth System Models with additional process complexity for instance prescribe emissions of pollutants and allow for biogeochemical feedbacks (yellow).

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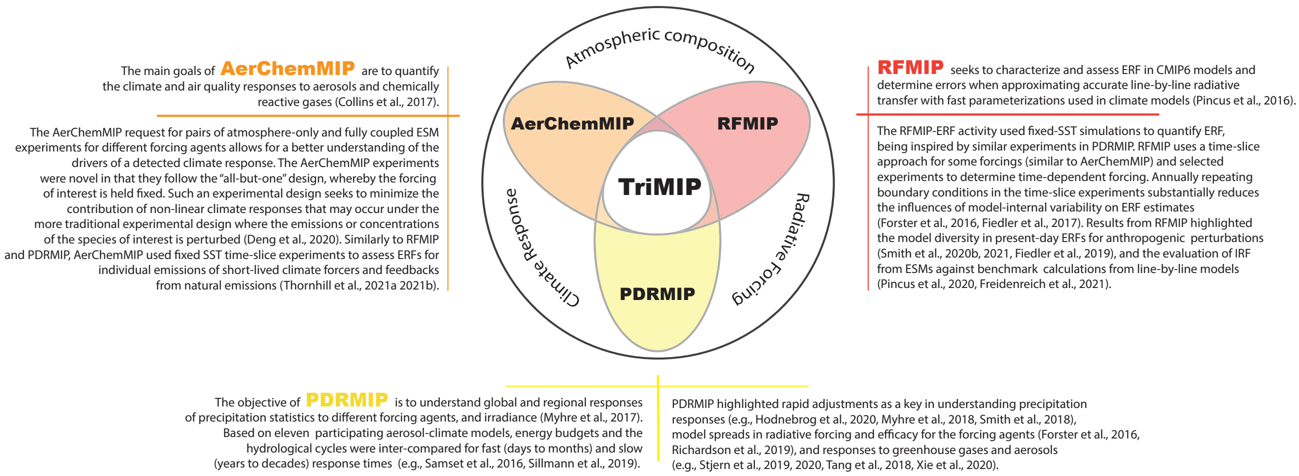


**Figure 2.** Overview of the MIPs and their topics in TriMIP.

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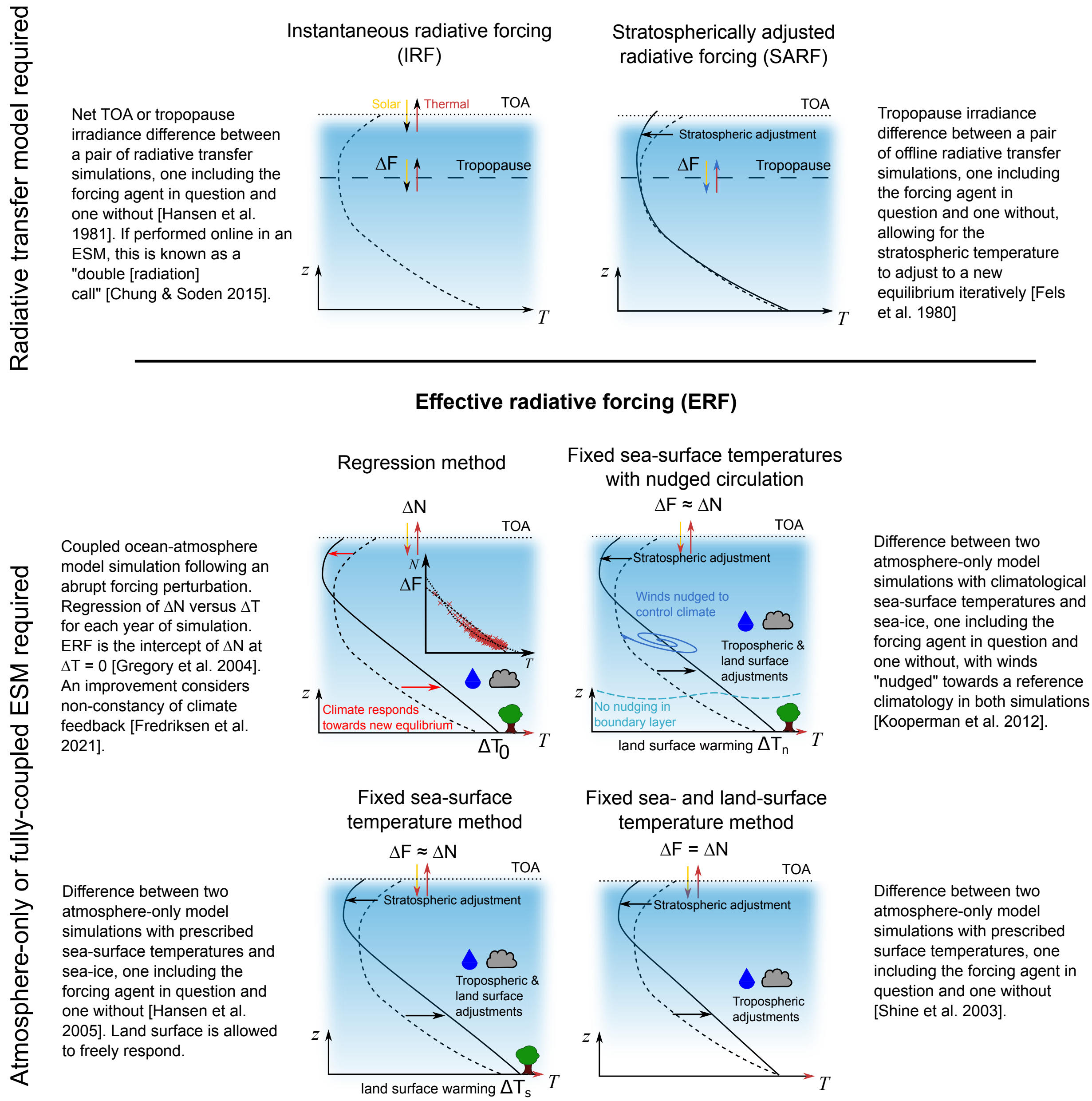


**Figure 3.** Methods for calculating radiative forcing. Shown are graphical depictions of the different methods for calculating radiative forcing based on (top) radiative transfer models and (bottom) general circulation models (GCMs). The latter have substantially developed over time by including more biological, physical and chemical processes, resulting in today's most comprehensive Earth System Models (ESMs). The methods differ in accuracy indicated by the differences between changes in the radiation ($\Delta F$) and energy budgets ($\Delta N$) at the top of the atmosphere (TOA) that arise from method-dependent temperature changes ($\Delta T$). ERF of ESMs in CMIP6 was for instance quantified with the fixed sea-surface temperature method. More accurate results might be obtained with the fixed sea- and land-surface temperature method in future experiments.

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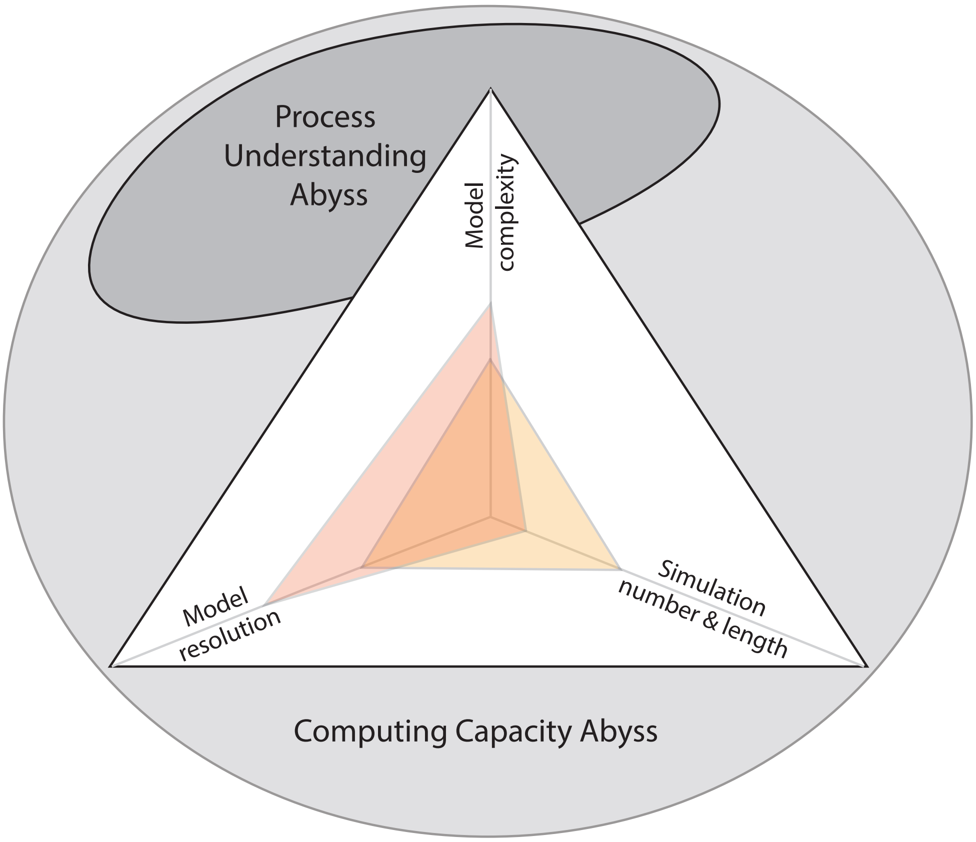


**Figure 4.** Tradeoffs in model configuration. Shown are the aspects in model experiment choices along the three axes: complexity, resolution, and length. The triangles mark potential choices along these axes with the area of the triangle indicating the computational demand. The circles mark limits concerning the availability of computing resources (Computing Capacity Abyss) and the understanding of physical, chemical and biological processes (Process Understanding Abyss).

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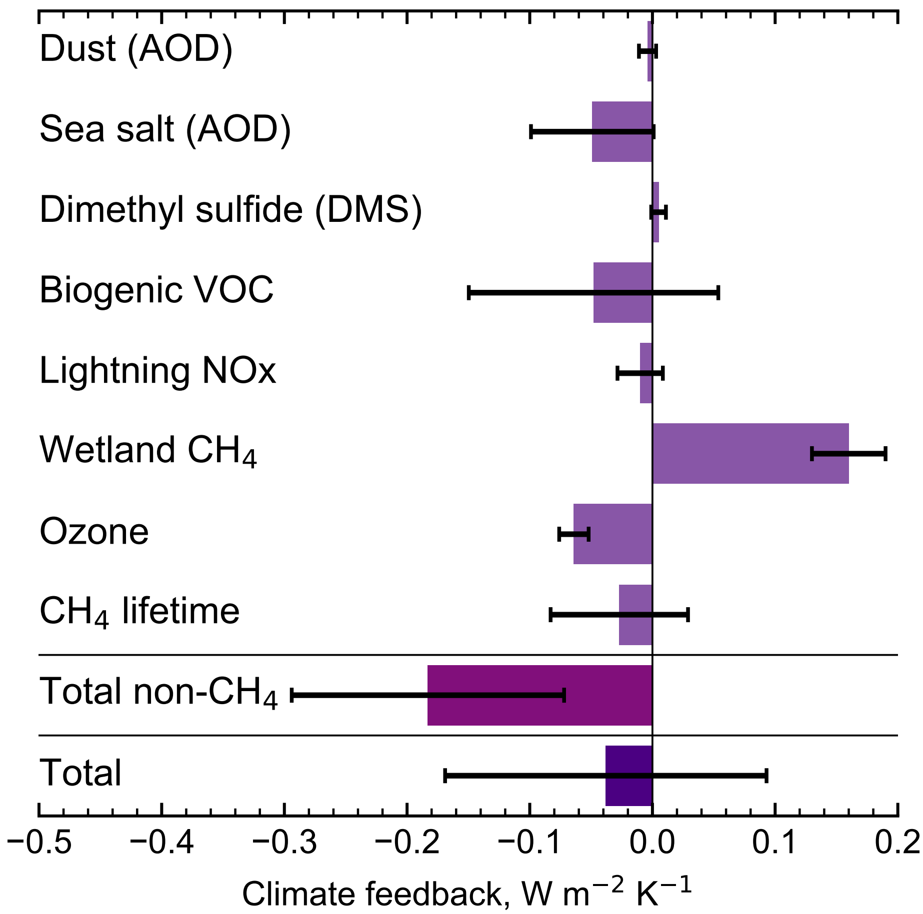


**Figure 5.** Feedback parameter, a measure of change in net energy flux at the top of the atmosphere for a given change in surface temperature, for chemistry and aerosol processes from AerChemMIP experiments (adjusted Figure 5 from Thornhill et al., 2021a, under the Creative Commons Attribution 4.0 International License - CC BY 4.0). Shown are the multi-model means (bars) and the model-to-model standard deviations (lines). Totals are sums of the individual feedbacks. Dust and sea-salt feedbacks are measured by their aerosol optical depth (AOD). Details on the calculation are given in Thornhill et al. (2021a).

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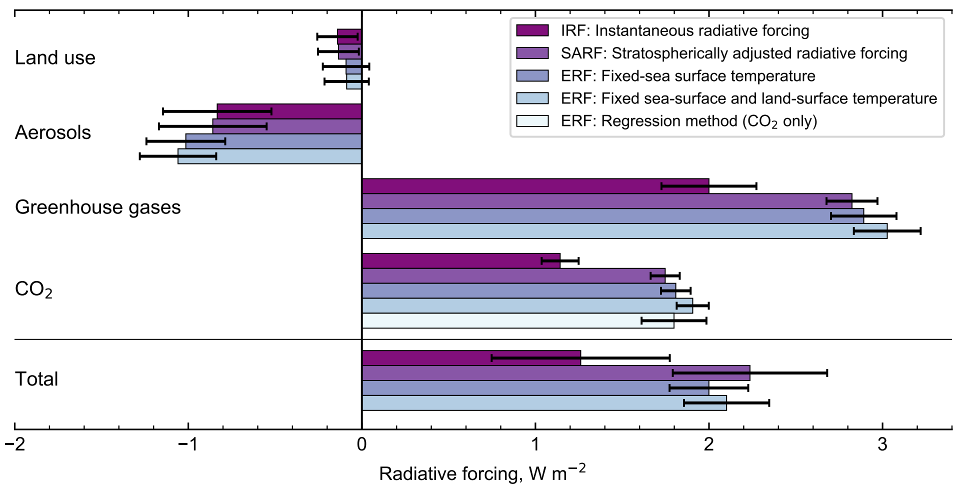


**Figure 6.** Radiative forcing for 2014 relative to 1850 from RFMIP experiments (adjusted Figure 1 from Smith et al., 2020a, under the Creative Commons Attribution 4.0 International License - CC BY 4.0). Shown are the multi-model means (bars) and the model-to-model standard deviations (lines) following different methods, graphically depicted in Figure 3. Details on the calculations are given in Smith et al. (2020a).