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The Multi-Scale Infrastructure for Chemistry and Aerosols (MUSICA)

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ABSTRACT: To explore the various couplings across space and time and between ecosystems in a consistent manner, atmospheric modeling is moving away from the fractured limited-scale modeling strategy of the past toward a unification of the range of scales inherent in the Earth system. This paper describes the forward-looking Multi-Scale Infrastructure for Chemistry and Aerosols (MUSICA), which is intended to become the next-generation community infrastructure for research involving atmospheric chemistry and aerosols. MUSICA will be developed collaboratively by the National Center for Atmospheric Research (NCAR) and university and government researchers, with the goal of serving the international research and applications communities. The capability of unifying various spatiotemporal scales, coupling to other Earth system components, and process-level modularization will allow advances in both fundamental and applied research in atmospheric composition, air quality, and climate and is also envisioned to become a platform that addresses the needs of policy makers and stakeholders.

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Empirical and modeling studies have provided strong evidence of dynamical and chemical coupling across the range of spatial and temporal scales inherent in the Earth system (e.g., Prinn 2012). Current chemical transport models, however, inadequately account for the two-way coupling of atmospheric chemistry with other Earth system components over the range of urban/local to regional to global scales and from the surface up to the top of the atmosphere.

As a result, the predictability of local air quality from regional models is currently limited by the insufficient representation of large-scale feedbacks and prescribed land and ocean state, while predictability of future atmospheric trace constituents is hampered by either the coarse resolution of global chemical climate models or the initial and boundary impacts affecting limited area regional models (e.g., National Research Council 2012; Bellucci et al. 2015; Huang et al. 2017; Neal et al. 2017; Im et al. 2018). Also, predictions of weather and climate often neglect the two-way interactions with atmospheric trace constituents.

However, in order to establish warning systems and develop adaptation and mitigation strategies, decision-makers need accurate predictions of atmospheric composition, weather, and climate on time scales from hours to weeks to seasonal to decadal, along with reliable quantification of their uncertainties. To meet future challenges, future modeling systems need to have the ability to 1) change spatial scales in a consistent manner, 2) resolve multiple spatial scales *in a single simulation*, 3) couple model components that represent different Earth system processes, and 4) easily mix and match model components as needed for a specific application. This requires moving away from the fractured modeling activities of the past and bridging the gap between regional chemical weather models and global chemical climate models. It requires coupling from the local emission scale all the way up to the global forcing scale within a single framework.

This vision motivates the development of Multi-Scale Infrastructure for Chemistry and Aerosols (MUSICA). MUSICA is not a single model, but a set of infrastructure concepts and requirements with rigorously defined standards that will enable studying atmospheric composition across all relevant scales. It describes a unified and modular framework with consistent scale-aware modeling approaches, i.e., approaches that are not dependent on model resolution and that can be applied to any Earth system model. It will account for feedbacks between all components of the Earth system, thereby permitting the exploration of interactions between atmospheric chemistry and weather and climate. Evaluation and data assimilation will be essential components of MUSICA.

To date, the lack of coordination between the different atmospheric chemistry models has made it difficult to accurately assess the benefits of different model components (Fast et al. 2011). MUSICA's flexible modular design will break down the problem of simulating atmospheric composition into the representation of individual processes that are described by separate modules. MUSICA can be configured in different ways appropriate for the scientific question on hand. This design will facilitate direct intercomparison of individual modules in a single framework, opening the way to quantify uncertainties of individual processes and enable co-development of individual components.

MUSICA will enable exploration of new science topics (Table 1) addressing important, frontier issues by overcoming constraints posed by limited scale awareness of current models and frameworks. MUSICA is envisioned to become a central tool for research but needs to be applicable to a wide range of users and should also be suitable for operational applications. This requires the system to be open source, transferable, efficient, and user friendly.

These new approaches to a modular unified framework are too large a challenge to be achieved by a single organization. MUSICA and its components are developed by the atmospheric research community under the coordination of the National Center for Atmospheric Research (NCAR) Atmospheric Chemistry Observations and Modeling (ACOM) Laboratory and in support of the National Science Foundation (NSF) Atmospheric and Geospace Sciences (AGS) Atmospheric Chemistry program. The current status and the future design plans for MUSICA are shared here with the entire community, with an invitation to actively engage in the design and development so that the framework is built to address the needs of the wider community. Several strategic partnerships with prominent research groups and organizations in the United States and elsewhere in the world are being established to foster this community development. MUSICA is guided by a steering group with committed representatives of the broad research community and by working groups each led by three co-chairs, who represent the wider research community including national and international universities, and research organizations (see www2.acom.ucar.edu/sections/musica-governance for a list of members). The working groups are open and invite members from the community to join them. The NCAR ACOM Laboratory serves as the lead organizer and coordinator of MUSICA. It is intended that MUSICA development will connect with other unified atmospheric chemistry modeling initiatives and leverage ongoing activities. Examples include the Modular Earth Submodel System (MESSy; Jöckel et al. 2010; Garny et al. 2019), the Multiscale Online Nonhydrostatic Atmosphere Chemistry (MONARCH) model (Badia et al. 2017), and NOAA's Unified Forecast System (UFS) community effort, which is aimed at producing a multiscale operational global numerical forecast system.

Infrastructure design

The specific goal of MUSICA is to produce a new modular and flexible infrastructure that will enable chemistry and aerosols to be simulated over all relevant atmospheric scales in a single, coherent fashion. Coupling of a stand-alone chemistry component to other atmospheric processes (e.g., aerosol–cloud interactions or convective scavenging) and Earth system

Table 1. Examples of new science applications enabled by MUSICA.

Past and Current Approach	Future Approach including MUSICA
Impacts of the Asian monsoon on weather and climate	
Hemispheric to global impacts without resolving convection or surface air quality over the monsoon region.	More realistic predictions by resolving local air quality and convection in monsoon region consistently with global impacts.
Exploiting the future constellation of geostationary satellites for atmospheric composition	
Global analysis at resolutions coarser than that of observations or regional analysis without considering the benefits of the entire constellation and those of polar orbiting satellites.	Matching measurement resolution over the key regions together with global feedbacks results in more seamless prediction of long-range transport and more accurate source attributions between regions of the globe.
Subseasonal to seasonal predictions of air quality and weather	
Hampered by global simulations not fully resolving regional climate-relevant phenomena and with limited accuracy and information content at human impact scales.	Global coupled predictions with strategically placed high resolution over key areas (e.g., El Niño region) improves simulated regional climate variability, air–sea coupling, and exposure-relevant scales.
Stratospheric intrusions	
Global models with coarse resolution near the upper troposphere–lower stratosphere or regional models dependent on boundary conditions.	Higher horizontal and vertical resolutions over tropopause fold regions will allow better representation of frontal passages and the filaments associated with intrusions.
Long-range pollution transport and urban air quality	
Global models providing boundary conditions to regional models only consider one-way transport and are inconsistent in nature.	Seamless two-way feedback from local to global and global to local scales.
Intercontinental-/global-scale transport of chemical layers	
Coarse vertical and spatial resolution and associated numerical diffusion prevents simulation of the layered structure of the troposphere.	Adaptive model resolution preserves chemical layers and enables assessment of their global impacts.
Aerosols seeding extreme events (e.g., hurricanes)	
High resolution over impact regions but coarse resolution over aerosol source regions and/or from lateral boundary conditions leads to poor aerosol prediction and affects feedback on extreme event predictions.	High resolution enabled over impacts and aerosol source regions in a consistent framework with fully enabled feedback of meteorology, chemistry, and dynamics and between ocean and atmosphere.
Feedback loop of climate change on trace gas and aerosol gas concentrations	
Global simulations with coarse resolution over high-emissions regions impact the accuracy of simulated pollutant life cycles and land–sea–atmosphere exchange.	Global feedbacks with increased spatial resolution over high-emission regions better represent the life cycles of short-lived pollutants and land–sea–atmosphere exchange.
Gravity wave processes impacting stratosphere and mesosphere temperature and mixing	
Global simulations with general circulation, chemistry, and climate dependent on parameterized wave sources, characteristics, and transport; or costly high-resolution "nature runs."	Better resolution of the gravity wave spectrum within the refined region and a more internally consistent gravity wave parameterization on the global grid.
Effect of megacities on global atmospheric composition and climate	
Disconnected spatial and temporal scales, separate models for local/regional and global impacts.	A fully coupled system accounts for detailed chemistry/emissions over megacities, and enables quantifying their impacts on remote regions (e.g., Arctic) and the global atmosphere.
Chemical data assimilation (CDA) and evaluation	
CDA in models is generally done separately from meteorological DA and limited to updating atmospheric concentrations. Evaluation tools have been developed separately from DA.	CDA will be co-developed as integral part of MUSICA with the objective of updating concentrations and inputs (e.g., emissions) efficiently. Commonalities between DA and evaluation will be addressed in parallel.
Air quality (AQ) under a changing climate	
Use downscaling methods to provide meteorological and chemical initial and boundary conditions to a regional AQ model.	Conduct an ensemble of simulations under various future scenarios with a single self-consistent model with sufficient resolution to simulate key AQ metrics.

Table 1. (*Continued*).

Co-benefits of greenhouse gas–emission reduction policies	
Conduct separate inconsistent simulations using regional or global AQ models and global climate models to investigate impacts on AQ and climate change from the move toward a net-zero-emissions future.	Conduct simulations under net-zero future scenarios with a single self-consistent model with sufficient resolution to simulate key AQ metrics and accurately simulate climate change.
Top-down emission estimates	
Either coarse resolution or inconsistency in modeling and emissions when constraining sources and sinks of long-lived species.	Improved accuracy and consistency by simulating transport and chemistry of long-lived species consistently across all scales.
Land surface coupling	
Coarser-resolution climate models are limited in their representation of land–atmosphere couplings, such as biogenic emissions and dry deposition of atmospheric constituents. Many regional models lack full coupling between land and atmosphere processes.	Land–atmosphere coupling and regionally finer resolution improves representation of meteorology, biogenic emissions and wet and dry deposition (e.g., simulating effect of acid rain on vegetation).

components (e.g., land–sea atmospheric exchange) will leverage community-developed software tools. The Earth System Modeling Framework (ESMF, https://sourceforge.net/projects/esmf/) and National Unified Operational Prediction Capability (NUOPC, www.earthsystemcog.org/projects/nuopc/refmans) provide flexible and tested tools for coupling models and grid interpolation. The Common Community Physics Package (CCPP, https://dtcenter.org/community-code/common-community-physics-package-ccpp) (Theurich et al. 2016), which will be adopted by MUSICA, includes a metadata standard that allows a model build system to check that all required fields are present. This infrastructure can be used to make sure all required fields are either provided by the host model or are read in from a data file.

MUSICA design will enable its components to be connected to any three-dimensional (3D) global or regional atmosphere model, or to any 1D single column or 0D box model through the CCPP. It will also have the capability to be driven by atmospheric reanalyses. The infrastructure will provide and extend the functionality of existing models [e.g., the Community Atmosphere Model with Chemistry (CAM-chem) and Whole Atmosphere Community Climate Model (WACCM), both embedded in the Community Earth System Model (CESM; Hurrell et al. 2013), GEOS-Chem (Bey et al. 2001), or the Weather Research and Forecasting Model with Chemistry (WRF-Chem; Grell et al. 2005; Fast et al. 2006)], but will also incorporate and advance models of chemistry and aerosols down to turbulence-resolved scales (e.g., Kim et al. 2012). In a first configuration, MUSICA will be configured within the CESM framework ("Example implementations and current status" section).

At the heart of MUSICA is the Chemistry and Aerosol Suite (Fig. 1), which provides updates to chemical states of gases and aerosols within the broader atmospheric simulation framework. The suite is being designed to support a range of gas phase schemes ("Chemical schemes" section) and aerosol representations ("Aerosols" section) and includes all associated processes (e.g., photolysis-rate calculations, nucleation, coagulation, thermodynamics) in separate modules. It is to be incorporated through the CCPP to enable its connection to any physical component that is compliant with the interface. Any atmospheric dynamics model that uses CCPP will be able to use those physical components. This modular structure with a standardized interface simplifies the intercomparison of individual modules and opens the way to quantifying uncertainties of individual processes, e.g., directly assessing the impact of different aerosol schemes on radiative forcing.

MUSICA will be capable of coupling to biogeochemistry treated in land and ocean models and provide a platform for both short-term air quality predictions and fully coupled Earth system simulations. Closely tied to the development of MUSICA is the development of community tools for processing input data such as emissions onto flexible model grids ("Emissions and deposition" section), a common model evaluation and diagnostics framework, and data assimilation capabilities ("Model evaluation and data assimilation" section).

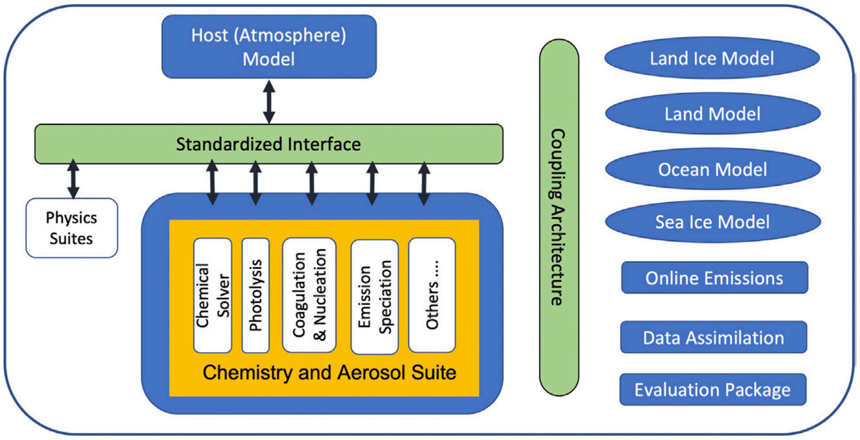

Fig. 1. MUSICA defines a framework to integrate chemistry into any atmospheric host model. It includes coupling to emissions, evaluation, and data assimilation tools and enables linking to other Earth system components, all through well-defined standardized interfaces. All physics and chemistry processes operate on individual columns; cross-column processes have to be handled by the host model.

MUSICA components and working groups

The seven existing MUSICA working groups are established and invite members from the community to become active members. The following sections provide a brief overview of the working group topics and discuss expected developments, challenges and scientific gains expected from the new infrastructure, and how limitations in current modeling systems (Table 2) can be overcome.

Model architecture. MUSICA needs to be fundamentally flexible to allow individual components to be used in multiple atmospheric host models and data needs to be communicated between individual modules through well-defined standardized interfaces. Data requirements from each module are communicated during the time of model configuration and configuration tests are being implemented to ensure that a valid and compatible set of model components is selected. Users of MUSICA will be able to select parameterizations in a user-friendly way. A dictionary of chemical properties will ensure that physical and chemical quantities are defined consistently between all components of the framework. For MUSICA, monolithic codes will be broken into separate interoperable modules (Fig. 1), thus a user can choose a set of modules during configuration and also change the order of module calls (e.g., test calling transport routines before or after chemistry routines).

Emissions and observational data or any other input data will be accessed and remapped to the model grid during runtime. A remapping tool is responsible for the full 3D conservative remapping required by the user specified model grid and the source data grids. A clearly defined layer will provide a connection for code related to the evaluation of the model, modification of model data through assimilation ("Model evaluation and data assimilation" section), and delivery of emissions data ("Emissions and deposition" section). These processes abstract the data sources from the model data, thus minimizing problems such as storing data on alternative grids, communication costs, and load balancing. It will eliminate the need for users to implement regridding solutions themselves.

MUSICA will simplify the user experience, clarify data dependencies between parameterizations, make the configurations of scientific model runs and corresponding datasets more traceable, support unit testing and integration testing, broaden and simplify model evaluation, and simplify updating and adding new schemes and parameterizations. Important goals include ensuring that MUSICA can be run on many different computer systems, designing clear processes for code improvements, and satisfying many different types of users. Last, MUSICA and all its components must guarantee open access and provide comprehensive benchmark scenarios and user guides.

Table 2. Limitations in current modeling systems as they relate to the different MUSICA Working Group topics.

Model architecture

- Physical and chemical processes are typically packaged together into a single monolithic code base with complex data interdependencies, but this slows down scientific advancement and also prevents fair comparisons and rigorous assessments of different process treatments. This inflexible approach further complicates modifying or replacing different process modules in models.

Emissions and deposition

- Current models require extensive preprocessing of emissions inventory files to match the model horizontal resolution, the model chemical scheme, etc. This also consumes disk space, challenges incorporating feedbacks of dynamics and meteorology on emissions, and often requires users to develop their own regridding tools.
- Using offline inventories introduces inconsistencies between land use and vegetation information used in the model and that used to generate the different types of anthropogenic and natural emissions
- Current dry deposition schemes crudely parameterize the dependence on meteorology and biophysics, leading to poor understanding of the impact of dry deposition on atmospheric chemistry and vegetation.

Chemical schemes

- Current chemical mechanisms are not flexible and targeted toward a limited scale and application.
- Mechanism development, which is based on a set of handwritten equations, starting simple and adding complexity as needed (Sarwar et al. 2008; Carter 2010; Stockwell et al. 1997; Emmons et al. 2010; Bates and Jacob 2019; Wang et al. 2019) is becoming more challenging as complexity increases.

Aerosols

- Modules that calculate aerosol processes are typically strongly tied to the aerosol representation of the host model, which complicates porting aerosol process treatments between host models with different aerosol representations. The required level of complexity increases when considering aerosol–cloud interactions or interstitial and cloud-borne aerosol species.

Physics, transport, subgrid processes

- Subgrid physics processes can be sensitive to the grid mesh sizes and therefore are often tuned within a limited range of spatial scales for optimizing model results.
- Most chemistry transport models assume that each grid cell is well mixed, although it has been shown with large-eddy simulations that reactants can be segregated causing effective reaction rates to be different from those in the well-mixed grid cell (e.g., Ouwersloot et al. 2011; Kim et al. 2016).
- Currently, boundary layer parameterizations are employed by the chemistry transport model and constituents are transported as passive tracers. However, fast chemistry alters trace gas vertical gradients, which affects their transport efficiency.

Whole atmosphere

- Gravity waves are a dominant driver of the vertical constituent transport and circulation of the middle and upper atmosphere, connecting the whole atmosphere to solar processes and Earth’s surface (Fritts and Alexander 2003; Grygalashvyly et al. 2012; Smith et al. 2013; Gardner and Liu 2016; Hendrickx et al. 2018). These waves must be parameterized as their 10–100-km scales are too small to be resolved on a global mesh, but there is evidence their transports are severely underrepresented in models (Garcia et al. 2014; Smith et al. 2015; Huang et al. 2015; Millán et al. 2015; Yue et al. 2015; Gardner et al. 2016).

Model evaluation and DA

- Current evaluation packages are limited in scope and inflexible at incorporating new observational datasets.
- The inability to conduct a more advanced ensemble analysis and prediction via stochastic parameterization across scales and chemical species, leads to insufficient characterization of model skill.
- Not considering multiscale coupled chemical DA leads to suboptimal exploitation of information content in observations and in accounting for chemical feedback processes.
- Mismatches in the representation of observational and model data result in systematic errors in model evaluation and source attribution.

Emissions and deposition. A critical component of chemistry models is the specification of anthropogenic, biomass burning, and natural emissions. Historic and up-to-date emission inventories will be made available in MUSICA, in cooperation with international groups developing these inventories, and efforts will be made to achieve consistency among global, regional, and local emission inventories.

The spatial resolution of emission inventories is constantly improving: the resolution of some global inventories is currently on the order of 0.1° (e.g., Crippa et al. 2016; Granier et al. 2018), close to that of regional inventories at 7 km (Kuenen et al. 2014). Nevertheless, such resolutions might still be too coarse for modeling some scenarios, e.g., urban air quality at the city scale. Moreover, the proxies used to spatially allocate the emissions may often not be representative of the real-world spatial emission patterns (e.g., use of population density to distribute residential wood combustion emissions).

An online capability for regridding, applying temporal variation and vertical distribution of emissions, and combining different inventories (e.g., with different sectors and/or different resolutions) is needed. This will eliminate the need for creating model-configured emission files as a preprocessing step. More importantly, it will allow accounting for impacts of dynamics and meteorology on emissions such as changes in fugitive emissions with temperature or plume dispersion for point emissions. To accomplish this, we use the examples of the Harvard–NASA Emissions Component (HEMCO; Keller et al. 2014) and the High-Elective Resolution Modeling Emission System (HERMES; Guevara et al. 2019) emission models, including collaborating with these teams.

Conservative regridding tools that can handle unstructured grid meshes with regional refinement capabilities (Fig. 2) are needed. Applying diurnal, day-of-week, holiday, and seasonal variations, as well as vertical distribution (e.g., power plant stack height), to standard inventories is critical for high-resolution air quality modeling, but may not be necessary for coarse-resolution global modeling. Emission temporal profiles should account for spatial variations across countries and regions due to variable sociodemographic habits (e.g., influence of local crop calendars on the application of fertilizers) and climate conditions (e.g., relationship between outdoor temperature and residential combustion emissions).

An online emission capability for MUSICA should enable efficient updating of inventories such as the ability to predict preliminary emissions for the most recent years, not typically available in standard inventories (e.g., by applying sector and pollutant-dependent

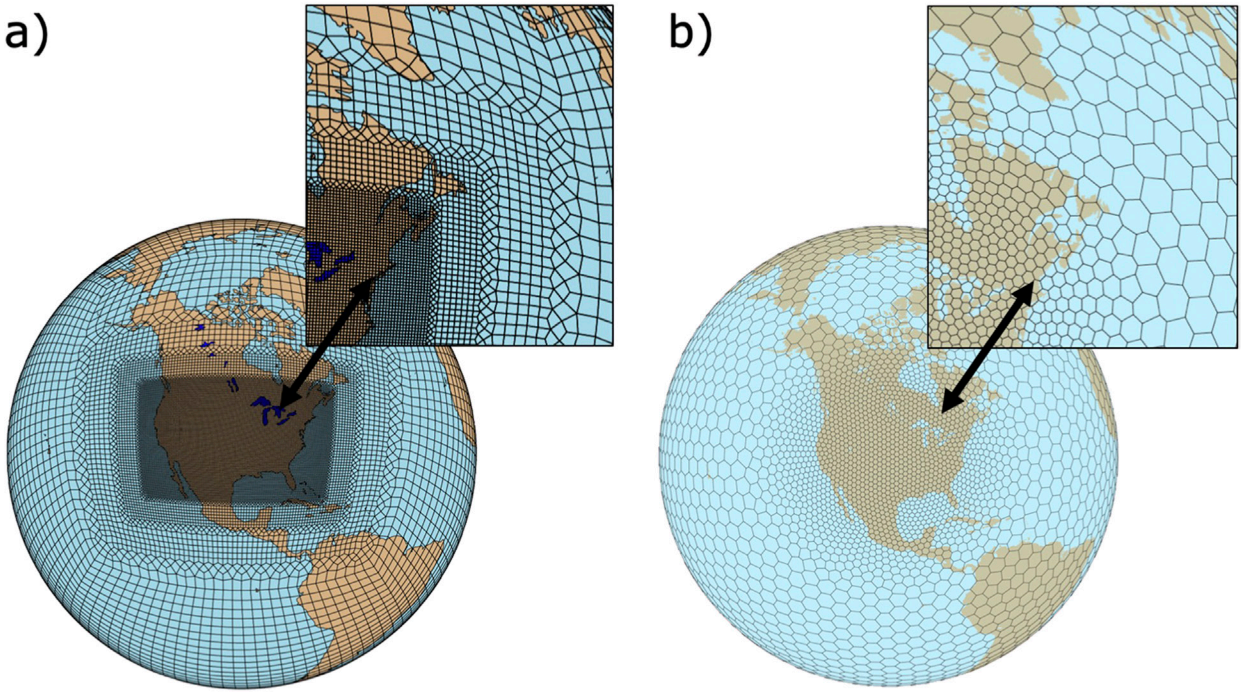

Fig. 2. Examples of unstructured grids with regional refinement over the contiguous United States: (a) the Spectral Element (SE) dynamical core (Zarzycki et al. 2014) and (b) the Model for Prediction Across Scales (MPAS; Michaelis et al. 2019) dynamical core.

extrapolations) or adding emission sectors that are not captured in inventories (e.g., traffic dust resuspension emissions). Detailed maps of vegetation and land use will be necessary for the spatial distribution of anthropogenic emissions linked to, e.g., agriculture practices, and for natural emissions (biogenic, dust, etc.), and should be consistent with those used in the development of fire emissions and the land type used in any other part of the simulation.

The integration of emission models into the emission tools, in addition to the inventories themselves, should be considered. Such models need to combine activity data and emission factors collected at a fine spatial scale (e.g., road level) with detailed emission estimation algorithms that represent the different factors influencing the emission processes. Moreover, these models should include functionalities for automatically manipulating and performing spatial operations on geometric objects (raster, shapefiles) so that they can be implemented in any region at high resolution.

Some emission sources are already calculated online in many models, such as biogenic emissions (Guenther et al. 2012; Hudman et al. 2012), lightning nitric oxide (NO) emissions ("Physics, transport, subgrid processes" section), or dust and sea salt emissions (Mahowald et al. 2006a,b). These emissions depend on environmental variables in nonlinear ways, requiring special care to make the underlying parameterization scale aware and linking to land and ocean models.

The degree to which depositional processes should be resolved for accurate simulation at different scales is uncertain at this stage. While recent work points to the value of coupling atmospheric and land models through terrestrial dry deposition (e.g., Paulot et al. 2018; Clifton et al. 2020), a major question is whether more complex models such as multilayer canopy models, including those with in-canopy ambient chemistry, more accurately capture dry deposition at regional scales. A common framework for testing different approaches for scale-aware dry deposition modeling would expedite advancing our understanding of dry deposition's role in the Earth system.

Chemical schemes. It is both a challenge and opportunity to provide accurate yet computationally efficient chemical mechanisms for diverse regions (local to global, troposphere to upper atmosphere) and time scales (hours to centuries). The development, testing, validation, and tracing of new chemical mechanisms will be made simpler in MUSICA's infrastructure design. New chemical mechanisms should be flexible and could be tailored to address different types of problems—e.g., a long-term global climate study might require a different mechanism than a short-term air quality study. Albeit a large challenge, the potential to apply different mechanisms at different locations/altitudes within a single model run (Santillana et al. 2010; Shen et al. 2020) should be explored.

Perhaps most exciting are the tools with which next-generation chemical mechanisms will likely be built in concert with the traditional mechanisms and methods alluded to in Table 2. Models like the Generator of Explicit Chemistry and Kinetics of Organics in the Atmosphere (GECKO-A; Aumont et al. 2005) and the Statewide Air Pollution Research Center (SAPRC) Mechanism Generation (MechGen) system (Carter 2019) use structure–activity relationships (SARs) to estimate kinetic parameters for unstudied reactions, and may contain millions of chemical species and reactions. These fully explicit mechanisms could replace traditional methods for mechanism development and could be reduced through some combination of machine learning and more traditional approaches (Szopa et al. 2005) to lumped/parameterized mechanisms of magnitude and content appropriate to answer a specific science problem.

As with other aspects of MUSICA, modularity is critical. There is a clear need for an adaptive approach (Shen et al. 2020) or other methods that allow for facile switching between different mechanisms including user-provided mechanisms. Benchmark mechanisms need to be provided that meet the needs of the user community, with respect to their ability to address scientific problems of current scope and interest as well as possible limitations in computing resources. In addition, seamless connections to other infrastructure components must exist (e.g., connections to solvers and radiative transfer codes). Connecting the chemical mechanisms to emission and deposition modules—i.e., providing protocols that allow connections to be made between emitted/deposited and mechanism species—needs to be given full consideration.

Various unresolved challenges associated with mechanism development remain. For example, as fossil fuel emissions, which currently are the dominant source over much of the developed world, are decreasing as a result of mitigation policies, other sources such as biomass burning or personal care products gain in importance. Including these new emission sources requires identifying appropriate compounds that represent the chemistry, developing reaction pathways for these identified compounds, and applying reduced-chemistry approaches. In many cases, the representative compounds are still being identified; for many of the compounds that have been identified, the available kinetic and mechanistic data are insufficient to develop reliable reaction pathways.

Mechanisms must include gas-phase chemistry, but also must consider condensed-phase partitioning and chemistry in aerosols and clouds. Chemical mechanisms for aerosol and cloud chemistry, which are continually developing, could follow the new paradigm of utilizing SARs to represent organic chemistry in the aqueous phase. The next-generation chemical mechanisms must address the need to represent a broader range of atmospheric processes, in addition to the range of scales and conditions previously described.

Aerosols. The representation of aerosol populations in current models ranges from the simplest and computationally most efficient bulk approach that assumes a single or fixed size bin (e.g., Colarco et al. 2010) to modal (e.g., Binkowski and Roselle 2003; Liu et al. 2012; Neale et al. 2010; Rasch et al. 2019), and sectional approaches (Wexler et al. 1994; Bessagnet et al. 2004; Zaveri et al. 2008). Fully explicit approaches, which simulate individual particles using a Monte Carlo method (e.g., Riemer et al. 2009) are becoming available. In contrast with the other approaches, the fully explicit approach does not impose any assumptions regarding the size distribution and mixing state of aerosol compositions, yet they are generally too computationally expensive to be included in 3D models. MUSICA will be designed to accommodate any type of aerosol treatment.

In addition to treating the aerosol number and size distribution, the complexity of an aerosol model also results from the treatment of processes that affect their life cycle such as nucleation, coagulation, gas-to-particle partitioning, removal mechanisms, and hygroscopic and optical properties. MUSICA and its underlying modularity will permit greater interoperability of processes within an aerosol model, therefore facilitating advances in aerosol process treatments that can be documented over time. For example, many types of thermodynamic modules (e.g., Jacobson et al. 1996; Nenes et al. 1999; Zaveri et al. 2005) with varying complexity and computational expense have been developed and implemented in various aerosol models. However, in a modular framework, one aerosol model could be chosen for different thermodynamic modules allowing a more direct assessment of errors associated with the treatment of gas-to-particle partitioning. Also, a flexible aerosol framework should lead to a better quantification of impacts of a more accurate representation of the aerosol mixing state (e.g., by allowing a better transition between freshly emitted aerosols that are treated as externally mixed and aged aerosols that are treated as internally mixed in remote regions).

Hundreds of prognostic variables are often needed to represent the size, composition, mixing state, and volatility associated with the multitude of chemical pathways responsible for secondary aerosol formation. In addition, it is often desirable to track anthropogenic, biomass burning, biogenic, and other natural aerosols separately, which further increases the computational burden. MUSICA will enable increased flexibility so that more complex and realistic representation of the aerosol life cycle can be included in air quality simulations in parallel with a computationally more efficient representation for climate simulations.

Physics, transport, subgrid processes. Physics processes and subgrid-scale transport encompass turbulence, gravity waves, convective transport, vertical mixing, and wet scavenging of trace gases and aerosols in stratiform and convective clouds. This includes chemical transformations within subgrid-scale processes, especially parameterized convection. Wet scavenging and lightning nitrogen oxides (NO_x) generation is included here, rather than the "Emissions and deposition" section, because of the reliance of these two processes on the properties of convection as well as resolved clouds.

While some numerical weather prediction and climate models already have the capability of regional refinement where scale-aware parameterizations have been evaluated (e.g., Fowler et al. 2016, 2020), an important activity is testing and evaluating parameterizations of processes at grid meshes ranging from hundreds of kilometers in size to ~1 km in size. Ultimately, the parameterizations need to ensure that mass, momentum, and energy are conserved in the transition from explicitly resolved to parameterized scales. However, there are several specific challenges for atmospheric chemistry that we highlight here.

One specific challenge is the vertical resolution (see also the "Whole atmosphere" section). With varying horizontal resolution, the vertical resolution and aspect ratios will also need to change to accurately resolve turbulent vertical mixing of atmospheric constituents, the role of convection and its effect on aerosol processing as well as finescale chemical processes. This may include how to best adapt and modify variable vertical grid structures for boundary layer parameterizations. Additionally, for representing the long-range transport of plumes or troposphere–stratosphere exchange, a vertical resolution of approximately 100 m is needed (Zhuang et al. 2018) and the question remains to what degree this will be feasible in a refined global mesh.

Another specific challenge is seamlessly estimating lightning flash rate. A coarse grid will contain a convective parameterization representing one or many convective storms, while the fine grid will resolve the convective storm. Considering that lightning flash-rate parameterizations are based on bulk properties of a single storm, there will be a need to transition from using storm properties in one grid cell to those in several neighboring grid cells. To confront this challenge, continual evaluation and development of lightning schemes will be needed.

In addition, constituent transport by shallow convection (Grell and Freitas 2014), a process often omitted in models, will need to be incorporated at grid scales larger than hundreds of meters. Shallow convective transport is a means of moving trace gases and aerosols to the free troposphere and simultaneously reducing surface mixing ratios. The MUSICA framework will allow further explorations of subgrid-scale issues such as the segregation of trace gases on small scales through its ability to connect to large-eddy simulation models or via regional to local grid refinement.

Whole atmosphere. Gravity waves couple the whole atmosphere through momentum and constituent transport and drive the general circulation of the middle atmosphere, while the Asian monsoon and tropopause folds transport constituents between the troposphere and the stratosphere. These phenomena are not adequately resolved in global models, and it is costly to simulate them with a global high-resolution mesh.

Regional refinement in MUSICA will resolve mesoscale gravity waves, reducing the dependence of circulation and transport in the middle and upper atmosphere on parameterizations. Since gravity waves can occur anywhere, adaptive grids would be ideal but are not likely to be realized in the near future. The primary challenges will be to 1) develop a "scale aware" gravity wave scheme that adjusts the parameterized wave spectrum within the grid refinement, 2) redevelop existing parameterizations to be consistent with the resolved gravity wave transports, and 3) address any spurious momentum forcings at the edges of the refinement where the newly resolved waves will decay (due to the coarsening grid) and potentially introduce artifacts in dynamics and transport.

In spite of these challenges, regional refinement in MUSICA presents new opportunities for research and model development. "Nature run" simulations with strategically placed refined meshes, in addition to remote sensing observations, can be used to develop more self-consistent parameterizations of gravity wave dynamics and transport for the global grid (e.g., Gardner et al. 2019). Regional refinement will better resolve key ocean–atmosphere coupling in subseasonal-to-seasonal forecasting regions, like the east Pacific, as well as finer horizontal transport in the Asian monsoon region (Table 1). However, vertical resolution remains the primary limit to modeling whole atmosphere coupling, and there is a need to explore the possibility of vertical refinement (Holt et al. 2016; Daniels et al. 2016; Garcia and Richter 2018).

Model evaluation and data assimilation. The integration with observations will be a critical aspect of MUSICA. MUSICA needs to be capable of supporting field campaign design and analyses, satellite calibration and validation, retrieval algorithm development, and correlative analysis between different atmospheric quantities as well as data assimilation and inverse modeling. By appropriately accounting for the differences in the representation of processes at multiple scales, MUSICA will allow reducing uncertainties in the representativeness of the modeled and observed states of chemical constituents (e.g., Janjíc et al. 2017), and as a consequence, will improve the utility of model evaluation and interpretation. As such, MUSICA will be essential for fully exploiting available observational constraints and gains from, for example, the upcoming geostationary satellite constellation (Judd et al. 2018; Chance et al. 2019) (Table 1).

Available evaluation tools should provide basic statistical metrics for evaluating the ability of the model to fit large observational datasets, for analyzing complex time series and for establishing statistical relations between the concentrations of different species. A database of evaluation datasets will be compiled, with the ability for a user to select which datasets are appropriate for the model evaluation (e.g., satellite curtains and maps, aircraft tracks, surface stations, sonde profiles) Given the selected datasets, output will be sampled at the appropriate co-temporal and collocated points and written to files for easy comparison.

Data assimilation tools will build upon current capabilities, e.g., the Data Assimilation Research Testbed (DART; Anderson et al. 2009; Gaubert et al. 2016), the Community Gridpoint Statistical Interpolation (GSI) tool (Shao et al. 2016), or the Joint Effort for Data Assimilation Integration (JEDI, www.jcsda.org/jcsda-project-jedi). Similarly, evaluation and diagnostic tools will complement existing tools, e.g., the CESM diagnostics package (www.cesm.ucar.edu/working_groups/Atmosphere/amwg-diagnostics-package/), the ObsPack Diagnostics (Masarie et al. 2014) or the Aerosol Modeling Testbed (Fast et al. 2011). MUSICA requires more advanced evaluation and assimilation tools that span a wide range of spatial and temporal scales and needs to provide interfaces that make it easy for users to digest their own data. Evaluation tools also need to enable direct model to model intercomparison.

Chemical data assimilation faces specific challenges. These include high dimensionality and nonlinearity, irregular frequency distribution, representativeness, mass conservation issues, model errors, parameter uncertainties, and more importantly the need to account for emissions (e.g., surface boundary conditions). Methods will also have to account for the tight coupling within the chemical system (between multiple constituents of varying lifetimes) and across Earth system components (e.g., Carmichael et al. 2008; Sandu and Chai 2011; Bocquet et al. 2015).

The development of tangent linear and adjoint capabilities, along with efforts to improve and refine the ensemble forecast capability, will be an integral part of MUSICA's development and not ex post facto as has been the case in the past. This offers a more effective and efficient development strategy, which can minimize many of the pitfalls (e.g., use of ad hoc approaches, high computational costs, redundant use of resources). Although still considered a frontier question, multiscale data assimilation (e.g., Nadeem et al. 2018) will be a target for MUSICA.

Technical challenges remain when assimilating chemical data on unstructured grids with variable resolution, particularly with regard to mass-conserving interpolation and grid transformation, similar to the challenges faced with emissions data ("Emissions and deposition" section). Development of scale-aware and consistent observation operators (i.e., transformation of states and parameters from model to observation spaces) is important. More advanced algorithms on localization and inflation (for ensemble Kalman filter and hybrid methods) and more flexible assimilation windows to specify different length scales (for 4D-Var) need to be developed to fully exploit the multiscale nature of the infrastructure.

Example implementations and current status

MUSICA is designed to be implemented under different configurations with, for example, different dynamical cores and different formulations of physical and chemical processes. This could include offline systems such as the GEOS-Chem High Performance model (Eastham et al. 2018), or online systems such as the ECMWF Integrated Forecast System (Huijnen et al. 2019). Once the models are adapted for compliance with CCPP and the principles underlying MUSICA, code and knowledge exchange could be significantly accelerated. However, a first implementation (MUSICA V0) will be achieved by configuring MUSICA within the CESM framework, which enables full interactions with ocean, ice, and land models.

As part of the System for Integrated Modeling of the Atmosphere (SIMA) project, CESM is being extended to support cross-scale simulations and includes the hydrostatic Spectral Element (SE) dynamical core with variable resolution in the atmosphere model (Lauritzen et al. 2018). This system allows global simulations with regional refinement down to a few kilometers. The addition of the Model for Prediction Across Scales–Atmosphere (MPAS-A; Skamarock et al. 2012, 2018) dynamical core and its physics parameterizations are underway and will extend the infrastructure to nonhydrostatic scales. A gas-phase-only version of the Chemistry and Aerosol Suite—the Model Independent Chemistry Module (MICM)—is being included in the early version of the model infrastructure and the inclusion of aerosols is underway. In a first release, the suite of Model for Ozone and Related Chemical Tracers (MOZART) mechanisms (Emmons et al. 2020) is integrated through MICM and a gas-phase chemistry box model version based on MICM (MusicBox) will be released to the community together with MUSICA V0 in summer 2020. Other chemical schemes are planned to be added to the framework by and with their developers, such as the mechanism used in the GEOS-Chem model (Bey et al. 2001).

Figure 3 shows surface ozone from a 1° uniform global-resolution simulation, which is compared to preliminary results from a MUSICA V0 simulation with a regionally refined grid of ~14-km resolution over the contiguous United States embedded in a 1° global resolution (as shown in Fig. 2a). Despite the fact that both simulations are nudged to meteorological reanalyses, the regional refinement leads to changes not only over the higher-resolution region, but also affects the outflow and leads to differences over the downwind regions. Evaluation of these first MUSICA simulations is still underway but the infrastructure is now at a stage where assessment of individual processes and of scale awareness is possible and the increased value of a multiscale framework can be tested. It also provides a framework conducive for active engagement with the community.

An offline mass-conserving emission processing tool for mapping a variety of inventories to unstructured grid meshes with regional refinement will become available together with MUSICA V0. Development of a single online and offline emission tool for MUSICA has started and involves restructuring of the HEMCO emission module (Keller et al. 2014). The current CESM evaluation and diagnostics tool is being adapted to work with the framework. Sharing these beta versions with the wider community provides a framework for co-development and testing.

As for data assimilation, the adoption of MUSICA Version 0 with existing capabilities (e.g., DART) is envisaged to be straightforward and will require less than two years to develop and implement. We also foresee that a development of an observation package can be leveraged across different activities (model development, evaluation and testing, data assimilation).

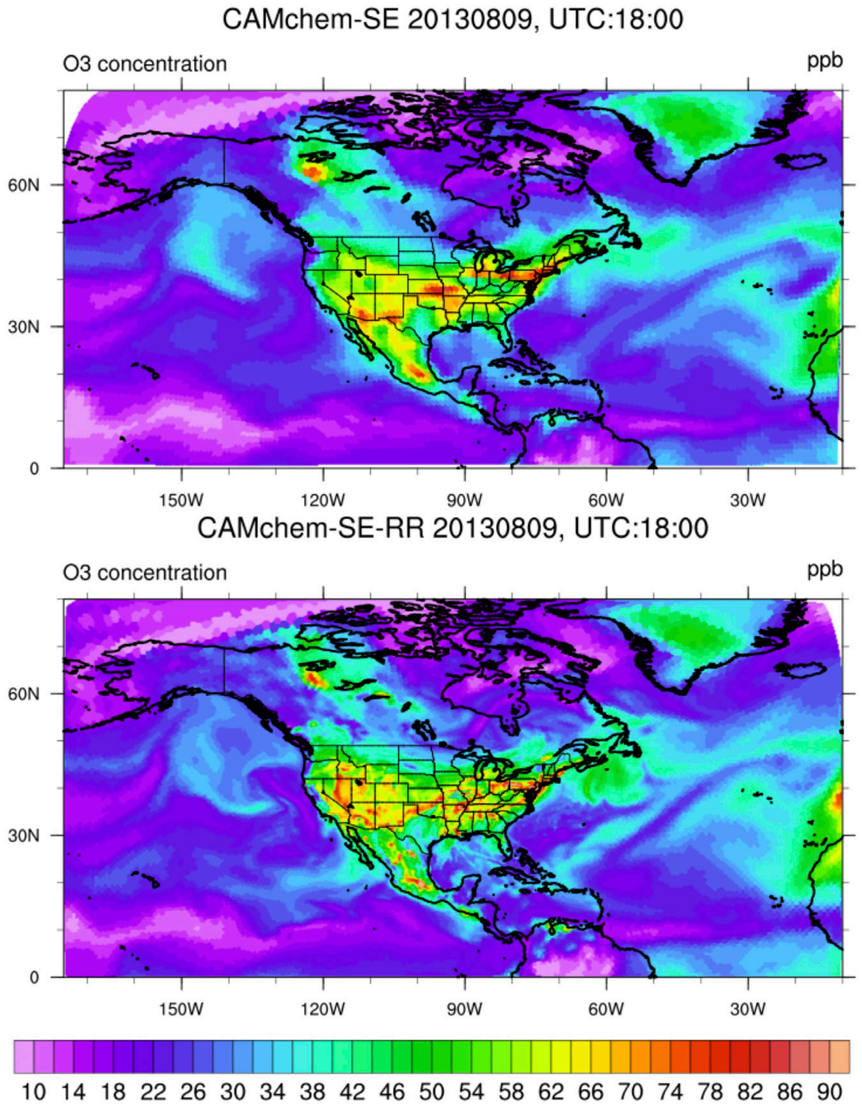

Fig. 3. Surface ozone for 1800 UTC 9 Aug 2013 (top) simulated at uniform global 1° horizontal resolution and (bottom) with a preliminary MUSICA V0 global 1° simulation that refines to 14-km resolution over the contiguous United States.

Community involvement and outlook

The development of a multiscale infrastructure that meets the scientific needs of the entire user community represents an exciting challenge that necessitates strong community involvement and partnerships among different organizations and different disciplines ranging from laboratory studies and field experiments to statistics and computational sciences and from molecular chemistry to space physics. The community is invited at an early stage to engage in the development and design of the MUSICA framework. The new capabilities represented by MUSICA will deepen existing, and establish new, working relations of the research community with a variety of users ranging from the research community to stakeholders. MUSICA will contribute to both advancing the science and to providing relevant and actionable information for the development of mitigation policies or warning systems.

Within the next few years, MUSICA will gradually replace the current suite of community chemistry models supported by NCAR and is envisioned to also integrate the capabilities of other modeling capabilities in the community. It will provide efficiencies through consolidation of model development and training efforts and provide a single point of entry for the majority of end-users. The transition phase will be dictated by the progress of MUSICA in providing at a minimum the capabilities of current models. The transition will be accompanied by educational activities including user guides and in-person and online tutorials. MUSICA User Tutorials will occur annually, but we envision the community taking part in offering training opportunities (e.g., through regional MUSICA "hubs").

After the challenges posed by the multiscale nature of the new infrastructure are met, the atmospheric community will be able to leverage MUSICA to advance our understanding of the multiscale couplings across the range of spatial scales, throughout the whole atmosphere, and across different components of the Earth system. The MUSICA framework, with its inherent modularity and flexibility and ability to tailor configurations to specific issues, as well as its emphasis on global community participation, is an excellent vehicle to address modern, multiscale, and complex problems. MUSICA is envisioned to become an engine of convergent research by not only advancing chemistry/aerosol/air quality modeling capabilities but by providing a framework for advancing research in the entire Earth system sciences.

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References

Anderson, J. L., T. Hoar, K. Raeder, H. Liu, N. Collins, R. Torn, and A. Avellino, 2009: The data assimilation research testbed: A community facility. *Bull. Amer. Meteor. Soc.*, **90**, 1283–1296, https://doi.org/10.1175/2009BAMS2618.1.

Aumont, B., S. Szopa, and S. Madronich, 2005: Modelling the evolution of organic carbon during its gas-phase tropospheric oxidation: Development of an explicit model based on a self generating approach. *Atmos. Chem. Phys.*, **5**, 2497–2517, https://doi.org/10.5194/ACP-5-2497-2005.

Badia, A., O. Jorba, A. Voulgarakis, D. Dabdub, C. Pérez García-Pando, A. Hilboll, M. Gonçalves, and Z. Janjic, 2017: Description and evaluation of the Multiscale Online Nonhydrostatic AtmospheRe CHemistry model (NMMB-MONARCH) version 1.0: Gas-phase chemistry at global scale. *Geosci. Model Dev.*, **10**, 609–638, https://doi.org/10.5194/GMD-10-609-2017.

Bates, K. H., and D. J. Jacob, 2019: A new model mechanism for atmospheric oxidation of isoprene: Global effects on oxidants, nitrogen oxides, organic products, and secondary organic aerosol. *Atmos. Chem. Phys.*, **19**, 9613–9640, https://doi.org/10.5194/ACP-19-9613-2019.

Bellucci, A., and Coauthors, 2015: Advancements in decadal climate predictability: The role of nonoceanic drivers. *Rev. Geophys.*, **53**, 165–202, https://doi.org/10.1002/2014RG000473.

Bessagnet, B., and Coauthors, 2004: Aerosol modeling with CHIMERE - Preliminary evaluation at the continental scale. *Atmos. Environ.*, **38**, 2803–2817, https://doi.org/10.1016/J.ATMOSENV.2004.02.034.

Bey, I., and Coauthors, 2001: Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation. *J. Geophys. Res.*, **106**, 232073–232095, https://doi.org/10.1029/2001JD000807.

Binkowski, F. S., and S. J. Roselle, 2003: Models-3 Community Multiscale Air Quality (CMAQ) model aerosol component: 1. Model description. *J. Geophys. Res.*, **108**, 4183, https://doi.org/10.1029/2001JD001409.

Bocquet, M., and Coauthors, 2015: Data assimilation in atmospheric chemistry models: Current status and future prospects for coupled chemistry meteorology models. *Atmos. Chem. Phys.*, **15**, 5325–5358, https://doi.org/10.5194/ACP-15-5325-2015.

Carmichael, G. R., A. Sandu, T. Chai, D. Daescu, E. Constantinescu, and Y. Tang, 2008: Predicting air quality: Improvements through advanced methods to integrate models and measurements. *J. Comput. Phys.*, **227**, 3540–3571, https://doi.org/10.1016/J.JCP.2007.02.024.

Carter, W. P. L., 2010: Development of the SAPRC-07 chemical mechanism. *Atmos. Environ.*, **44**, 5324–5335, https://doi.org/10.1016/J.ATMOSENV.2010.01.026.

——, 2019: Gateway to the SAPRC-16 Mechanism Generation System. http://mechgen.cert.ucr.edu/.

Chance, K., and Coauthors, 2019: TEMPO green paper: Chemistry, physics, and meteorology experiments with the tropospheric emissions: Monitoring of pollution instrument. *Proc. SPIE*, **11151**, 111510B., https://doi.org/10.1117/12.2534883.

Clifton, O. E., and Coauthors, 2020: Influence of dynamic ozone dry deposition on ozone pollution. *J. Geophys. Res. Atmos.*, **125**, e2020JD032398, https://doi.org/10.1029/2020JD032398.

Colarco, P., A. da Silva, M. Chin and T. Diehl, 2010: Online simulations of global aerosol distributions in the NASA GEOS-4 model and comparisons to satellite and ground-based aerosol optical depth. *J. Geophys. Res.*, **115**, D14207, https://doi.org/10.1029/2009JD012820.

Crippa, M., G. Janssens-Maenhout, F. Dentener, D. Guizzardi, K. Sindelarova, M. Muntean, R. Van Dingenen, and C. Granier, 2016: Forty years of improvements in European air quality: Regional policy-industry interactions with global impacts. *Atmos. Chem. Phys.*, **16**, 3825–3841, https://doi.org/10.5194/ACP-16-3825-2016.

Daniels, M. H., K. A. Lundquist, J. D. Mirocha, D. J. Wiersema, and F. K. Chow, 2016: A new vertical grid nesting capability in the Weather Research and Forecasting (WRF) Model. *Mon. Wea. Rev.*, **144**, 3725–3747, https://doi.org/10.1175/MWR-D-16-0049.1.

Eastham, S. D., and Coauthors, 2018: GEOS-Chem high performance (GCHP v11-02c): A next-generation implementation of the GEOS-Chem chemical transport model for massively parallel applications. *Geosci. Model Dev.*, **11**, 2941–2953, https://doi.org/10.5194/GMD-11-2941-2018.

Emmons, L. K., and Coauthors, 2010: Description and evaluation of the Model for Ozone and Related Chemical Tracers, version 4 (MOZART-4). *Geosci. Model Dev.*, **3**, 43–67, https://doi.org/10.5194/GMD-3-43-2010.

——, and Coauthors, 2020: The Chemistry Mechanism in the Community Earth System Model version 2 (CESM2). *J. Adv. Model. Earth Syst.*, **12**, e2019MS001882, https://doi.org/10.1029/2019MS001882.

Fast, J. D., W. I. Gustafson Jr., R. C. Easter, R. A. Zaveri, J. C. Barnard, E. G. Chapman, and G. A. Grell, 2006: Evolution of ozone, particulates, and aerosol direct forcing in an urban area using a new fully-coupled meteorology, chemistry, and aerosol model. *J. Geophys. Res.*, **111**, D21305, https://doi.org/10.1029/2005JD006721.

——, ——, E. G. Chapman, R. C. Easter, J. Rishel, R. A. Zaveri, G. Grell, and M. Barth, 2011: The Aerosol Modeling Testbed: A community tool to objectively evaluate aerosol process modules. *Bull. Amer. Meteor. Soc.*, **92**, 343–360, https://doi.org/10.1175/2010BAMS2868.1.

Fowler, L. D., W. C. Skamarock, G. A. Grell, S. R. Freitas, and M. G. Duda, 2016: Analyzing the Grell–Freitas convection scheme from hydrostatic to nonhydrostatic scales within a global model. *Mon. Wea. Rev.*, **144**, 2285–2306, https://doi.org/10.1175/MWR-D-15-0311.1.

——, M. C. Barth, and K. Alipathy, 2020: Impact of scale-aware deep convection on the cloud liquid and ice water paths and precipitation using the Model for Prediction Across Scales (MPAS-v5.2). *Geosci. Model Dev.*, **13**, 2851–2877, https://doi.org/10.5194/gmd-13-2851-2020.

Fritts, D. C., and M. J. Alexander, 2003: Gravity wave dynamics and effects in the middle atmosphere. *Rev. Geophys.*, **41**, 1003, https://doi.org/10.1029/2001RG000106.

Garcia, R. R., and J. H. Richter, 2018: On the momentum budget of the quasi-biennial oscillation in the whole atmosphere community climate model. *J. Atmos. Sci.*, **76**, 69–87, https://doi.org/10.1175/JAS-D-18-0088.1.

Garcia, R. R., M. Lopez-Puertas, B. Funke, D. R. Marsh, D. E. Kinnison, and A. K. Smith, 2014: On the distribution of CO_2 and CO in the mesosphere and thermosphere. *J. Geophys. Res. Atmos.*, **119**, 5700–5718, https://doi.org/10.1002/2013JD021208.

Gardner, C. S., and A. Z. Liu, 2016: Chemical transport of neutral atmospheric constituents by waves and turbulence: Theory and observations. *J. Geophys. Res. Atmos.*, **121**, 494–520, https://doi.org/10.1002/2015JD023145.

——, ——, and Y. Guo, 2016: Vertical and horizontal transport of mesospheric Na: Implications for the mass influx of cosmic dust. *J. Atmos. Sol. Terr. Phys.*, **162**, 192–202, https://doi.org/10.1016/J.JASTP.2016.07.013.

——, Y. Guo, and A. Z. Liu, 2019: Parameterizing wave-driven constituent transport in the upper atmosphere. *Earth Space Sci.*, **6**, 904–913, https://doi.org/10.1029/2019EA000625.

Garny, H., R. Walz, M. Nützel, and T. Birner, 2019: Extending the Modular Earth Submodel System (MESSy v2.55) model hierarchy: The ECHAM/MESSy idealized (EMIL) model set-up. *Geosci. Model Dev. Discuss*, https://doi.org/10.5194/gmd-2019-330.

Gaubert, B., and Coauthors, 2016: Towards a chemical reanalysis in a coupled chemistry-climate model: An evaluation of MOPITT CO assimilation and its impact on tropospheric composition. *J. Geophys. Res. Atmos.*, **121**, 7310–7343, https://doi.org/10.1002/2016JD024863.

Granier, C., N. Elguindi, and S. Darras, 2018: D81.2.2.3: CAMS emissions for all species for years 2000-2018, including documentation. ECMWF Copernicus Rep., 19 pp., accessed 27 December 2019, https://atmosphere.copernicus.eu/sites/default/files/2019-11/05_CAMS81_2017SC1_D81.2.2.3-201808_v2_APPROVED_Ver2.pdf.

Grell, G. A., and S. R. Freitas, 2014: A scale and aerosol aware stochastic convective parameterization for weather and air quality modeling. *Atmos. Chem. Phys.*, **14**, 5233–5250, https://doi.org/10.5194/ACP-14-5233-2014.

——, S. E. Peckham, R. Schmitz, S. A. McKeen, G. Frost, W. C. Skamarock, and B. Eder, 2005: Fully coupled 'online' chemistry in the WRF model. *Atmos. Environ.*, **39**, 6957–6976, https://doi.org/10.1016/J.ATMOSENV.2005.04.027.

Grygalashvyly, M., E. Becker, and G. R. Sonnemann, 2012: Gravity wave mixing and effective diffusivity for minor chemical constituents in the mesosphere/lower thermosphere. *Space Sci. Rev.*, **168**, 333–362, https://doi.org/10.1007/S11214-011-9857-X.

Guenther, A. B., X. Jiang, C. L. Heald, T. Sakulyanontvittaya, T. Duhl, L. K. Emmons, and X. Wang, 2012: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): An extended and updated framework for modeling biogenic emissions. *Geosci. Model Dev.*, **5**, 1471–1492, https://doi.org/10.5194/GMD-5-1471-2012.

Guevara, M., C. Tena, M. Porquet, O. Jorba, and C. Pérez García-Pando, 2019: HERMESv3, a stand-alone multi-scale atmospheric emission modelling framework – Part I: Global and regional module. *Geosci. Model Dev.*, **12**, 1885–1907, https://doi.org/10.5194/GMD-12-1885-2019.

Hendrickx, K., L. Megner, D. R. Marsh, and C. Smith-Johnsen, 2018: Production and transport mechanisms of NO in the polar upper mesosphere and lower thermosphere in observations and models. *Atmos. Chem. Phys.*, **18**, 9075–9089, https://doi.org/10.5194/ACP-18-9075-2018.

Holt, L. A., M. J. Alexander, L. Coy, A. Molod, W. Putman, and S. Pawson, 2016: Tropical waves and the quasi-biennial oscillation in a 7-km global climate simulation. *J. Atmos. Sci.*, **73**, 3771–3783, https://doi.org/10.1175/JAS-D-15-0350.1.

Huang, M., and Coauthors, 2017: Impact of intercontinental pollution transport on North American ozone air pollution: An HTAP phase 2 multi-model study. *Atmos. Chem. Phys.*, **17**, 5721–5750, https://doi.org/10.5194/ACP-17-5721-2017.

Huang, W., X. Chu, C. S. Gardner, J. D. Carrillo-Sanchez, W. Feng, J. M. C. Plane, and D. Nesvorny, 2015: Measurements of the vertical fluxes of atomic Fe and Na at the mesopause: Implications for the velocity of cosmic dust entering the atmosphere. *Geophys. Res. Lett.*, **42**, 169–175, https://doi.org/10.1002/2014GL062390.

Hudman, R. C., N. E. Moore, A. K. Mebust, R. V. Martin, A. R. Russell, L. C. Valin, and R. C. Cohen, 2012: Steps towards a mechanistic model of global soil nitric oxide emissions: Implementation and space based-constraints. *Atmos. Chem. Phys.*, **12**, 7779–7795, https://doi.org/10.5194/ACP-12-7779-2012.

Huijnen, V., and Coauthors, 2019: Quantifying uncertainties due to chemistry modelling: Evaluation of tropospheric composition simulations in the CAMS model (cycle 43R1). *Geosci. Model Dev.*, **12**, 1725–1752, https://doi.org/10.5194/GMD-12-1725-2019.

Hurrell, J. W., and Coauthors, 2013: The community Earth system model: A framework for collaborative research. *Bull. Amer. Meteor. Soc.*, **94**, 1339–1360, https://doi.org/10.1175/BAMS-D-12-00121.1.

Im, U., and Coauthors, 2018: Influence of anthropogenic emissions and boundary conditions on multi-model simulations of major air pollutants over Europe and North America in the framework of AQMEII3. *Atmos. Chem. Phys.*, **18**, 8929–8952, https://doi.org/10.5194/ACP-18-8929-2018.

Jacobson, M. C., M. L. Shulman, R. J. Charlson, and G.-J. Roelofs, 1996: Sensitivity of droplet size distributions to organic cloud condensation nuclei constituents. *Nucleation and Atmospheric Aerosols 1996*, Pergamon, 952–955, https://doi.org/10.1016/B978-008042030-1/50235-3.

Janjíc, T., N. Bormann, M. Bocquet, J. A. Carton, S. E. Cohn, S. L. Dance, and P. Weston, 2017: On the representation error in data assimilation. *Quart. J. Roy. Meteor. Soc.*, **144**, 1257–1278, https://doi.org/10.1002/QJ.3130.

Jöckel, P., and Coauthors, 2010: Development cycle 2 of the Modular Earth Submodel System (MESSy2). *Geosci. Model Dev.*, **3**, 717–752, https://doi.org/10.5194/GMD-3-717-2010.

Judd, L. M., and Coauthors, 2018: The dawn of geostationary air quality monitoring: Case Studies from Seoul and Los Angeles. *Front. Environ. Sci.*, **6**, 85, https://doi.org/10.3389/fenvs.2018.00085.

Keller, C. A., M. S. Long, R. M. Yantosca, A. M. Da Silva, S. Pawson, and D. J. Jacob, 2014: HEMCO v1.0: A versatile, ESMF-compliant component for calculating emissions in atmospheric models. *Geosci. Model Dev.*, **7**, 1409–1417, https://doi.org/10.5194/GMD-7-1409-2014.

Kim, S.-W., M. C. Barth, and M. Trainer, 2012: Influence of fair-weather clouds on isoprene chemistry. *J. Geophys. Res.*, **117**, D10302, https://doi.org/10.1029/2011JD017099.

——, ——, and ——, 2016: Impact of turbulent mixing on isoprene chemistry. *Geophys. Res. Lett.*, **43**, 7701–7708, https://doi.org/10.1002/2016GL069752.

Kuenen, J. J. P., A. J. H. Visschedijk, M. Jozwicka, and H. A. C. Denier van der Gon, 2014: TNO-MACC_II emission inventory; A multi-year (2003–2009) consistent high-resolution European emission inventory for air quality modelling. *Atmos. Chem. Phys.*, **14**, 10963–10976, https://doi.org/10.5194/ACP-14-10963-2014.

Lauritzen, P. H., and Coauthors, 2018: NCAR release of CAM-SE in CESM2.0: A reformulation of the spectral element dynamical core in dry-mass vertical coordinates with comprehensive treatment of condensates and energy. *J. Adv. Model. Earth Syst.*, **10**, 1537–1570, https://doi.org/10.1029/2017MS001257.

Liu, X., and Coauthors, 2012: Toward a minimal representation of aerosols in climate models: Description and evaluation in the Community Atmosphere Model CAM5. *Geosci. Model Dev.*, **5**, 709, https://doi.org/10.5194/GMD-5-709-2012.

Mahowald, N., J. Lamarque, X. Tie, and E. Wolff, 2006a: Sea-salt aerosol response to climate change: Last Glacial Maximum, preindustrial, and doubled carbon dioxide climates. *J. Geophys. Res.*, **111**, D05303, https://doi.org/10.1029/2005JD006459.

——, D. Muhs, S. Levis, P. Rasch, M. Yoshioka, C. Zender, and C. Luo, 2006b: Change in atmospheric mineral aerosols in response to climate: Last glacial period, preindustrial, modern, and doubled carbon dioxide climates. *J. Geophys. Res.*, **111**, D10202, https://doi.org/10.1029/2005JD006653.

Masarie, K. A., W. Peters, A. R. Jacobson, and P. P. Tans, 2014: ObsPack: A framework for the preparation, delivery, and attribution of atmospheric greenhouse gas measurements. *Earth Syst. Sci. Data*, **6**, 375–384, https://doi.org/10.5194/essd-6-375-2014.

Michaelis, A. C., G. M. Lackmann, and W. A. Robinson, 2019: Evaluation of a unique approach to high-resolution climate modeling using the Model for Prediction Across Scales – Atmosphere (MPAS-A) version 5.1. *Geosci. Model Dev.*, **12**, 3725–3743, https://doi.org/10.5194/GMD-12-3725-2019.

Millán, L., S. Wang, N. Livesey, D. Kinnison, H. Sagawa, and Y. Kasai, 2015: Stratospheric and mesospheric HO_2 observations from the Aura Microwave Limb Sounder. *Atmos. Chem. Phys.*, **15**, 2889–2902, https://doi.org/10.5194/ACP-15-2889-2015.

Nadeem, A., R. Potthast, and A. Rhodin, 2018: On sequential multiscale inversion and data assimilation. *J. Comput. Appl. Math.*, **336**, 338–352, https://doi.org/10.1016/J.CAM.2017.08.013.

National Research Council, 2012: *A National Strategy for Advancing Climate Modeling*. National Academies Press, 294 pp., https://doi.org/10.17226/13430.

Neal, L. S., M. Dalvi, G. Folberth, R. N. McInnes, P. Agnew, F. M. O'Connor, N. H. Savage, and M. Tilbee, 2017: A description and evaluation of an air quality model nested within global and regional composition-climate models using MetUM. *Geosci. Model Dev.*, **10**, 3941–3962, https://doi.org/10.5194/GMD-10-3941-2017.

Neale, R. B., and Coauthors, 2010: Description of the NCAR Community Atmosphere Model (CAM5.0). NCAR Tech. Note NCAR/TN-486+STR, 268 pp., www.cesm.ucar.edu/models/cesm1.1/cam/docs/description/cam5_desc.pdf.

Nenes, A., C. Pilinis, and S. N. Pandis, 1999: Continued development and testing of a new thermodynamic aerosol module for urban and regional air quality models. *Atmos. Environ.*, **33**, 1553–1560, https://doi.org/10.1016/S1352-2310(98)00352-5.

Ouwersloot, H. G., J. Vilà-Guerau de Arellano, C. C. van Heerwaarden, L. N. Ganzeveld, M. C. Krol, and J. Lelieveld, 2011: On the segregation of chemical species in a clear boundary layer over heterogeneous land surfaces. *Atmos. Chem. Phys.*, **11**, 102681–102704, https://doi.org/10.5194/ACP-11-10681-2011.

Paulot, F., S. Malyshev, T. Nguyen, J. D. Crounse, E. Shevliakova, and L. W. Horowitz, 2018: Representing sub-grid scale variations in nitrogen deposition associated with land use in a global Earth system model: Implications for present and future nitrogen deposition fluxes over North America. *Atmos. Chem. Phys.*, **18**, 172963–172978, https://doi.org/10.5194/ACP-18-17963-2018.

Prinn, R., 2012: Development and application of earth system models. *Proc. Natl. Acad. Sci. USA*, **110**, 3673–3680, https://doi.org/10.1073/PNAS.1107470109.

Rasch, P. J., and Coauthors, 2019: An overview of the atmospheric component of the energy exascale Earth system model. *J. Adv. Model. Earth Syst.*, **11**, 2377–2411, https://doi.org/10.1029/2019MS001629.

Riemer, N., M. West, R. Zaveri, and R. Easter, 2009: Simulating the evolution of soot mixing state with a particle-resolved aerosol model. *J. Geophys. Res.*, **114**, D09202, https://doi.org/10.1029/2008JD011073.

Sandu, A., and T. Chai, 2011: Chemical-Data assimilation – An overview. *Atmosphere*, **2**, 426–463, https://doi.org/10.3390/ATMOS2030426.

Santillana, M., P. Le Sager, D. J. Jacob, and M. P. Brenner, 2010: An adaptive reduction algorithm for efficient chemical calculations in global atmospheric chemistry models. *Atmos. Environ.*, **44**, 4426–4431, https://doi.org/10.1016/J.ATMOSENV.2010.07.044.

Sarwar, G., D. Luecken, G. Yarwood, G. Z. Whitten, and W. P. L. Carter, 2008: Impact of an updated carbon bond mechanism on predictions from the CMAQ modeling system: Preliminary assessment. *J. Appl. Meteor. Climatol.*, **47**, 3–14, https://doi.org/10.1175/2007JAMC1393.1.

Shao, H., and Coauthors, 2016: Bridging research to operations transitions: Status and plans of community GSI. *Bull. Amer. Meteor. Soc.*, **97**, 1427–1440, https://doi.org/10.1175/BAMS-D-13-00245.1.

Shen, L., D. J. Jacob, M. Santillana, X. Wang, and W. Chen, 2020: An adaptive method for speeding up the numerical integration of chemical mechanisms in atmospheric chemistry models: Application to GEOS-chem version 12.0.0. *Geosci. Model Dev.*, **13**, 2475–2486, https://doi.org/10.5194/GMD-13-2475-2020.

Skamarock, W. C., J. B. Klemp, M. G. Duda, L. D. Fowler, S.-H. Park, and T. Ringler, 2012: A multiscale nonhydrostatic atmospheric model using centroidal Voronoi tesselations and C-grid staggering. *Mon. Wea. Rev.*, **140**, 3090–3105, https://doi.org/10.1175/MWR-D-11-00215.1.

——, M. G. Duda, S. Ha, and S. Park, 2018: Limited-area atmospheric modeling using an unstructured mesh. *Mon. Wea. Rev.*, **146**, 3445–3460, https://doi.org/10.1175/MWR-D-18-0155.1.

Smith, A. K., V. L. Harvey, M. G. Mlynczak, B. Funke, M. García-Comas, M. Hervig, and M. Kaufmann, 2013: Satellite observations of ozone in the upper mesosphere. *J. Geophys. Res. Atmos.*, **118**, 5803–5821, https://doi.org/10.1002/JGRD.50445.

——, M. López-Puertas, B. Funke, M. García-Comas, M. G. Mlynczak, and L. A. Holt, 2015: Nighttime ozone variability in the high latitude winter mesosphere. *J. Geophys. Res. Atmos.*, **119**, 132547–132564, https://doi.org/10.1002/2014JD021987.

Stockwell, W. R., F. Kirchner, M. Kuhn, and S. Seefeld, 1997: A new mechanism for regional atmospheric chemistry modeling. *J. Geophys. Res.*, **102**, 252847–252879, https://doi.org/10.1029/97JD00849.

Szopa, S., B. Aumont, and S. Madronich, 2005: Assessment of the reduction methods used to develop chemical schemes: Building of a new chemical scheme for VOC oxidation suited to three-dimensional multiscale HOx-NOx-VOC chemistry simulations. *Atmos. Chem. Phys.*, **5**, 2519–2538, https://doi.org/10.5194/ACP-5-2519-2005.

Theurich, G., and Coauthors, 2016: The Earth system prediction suite: Toward a coordinated U.S. modeling capability. *Bull. Amer. Meteor. Soc.*, **97**, 1229–1247, https://doi.org/10.1175/BAMS-D-14-00164.1.

Wang, X., and Coauthors, 2019: The role of chlorine in global tropospheric chemistry. *Atmos. Chem. Phys.*, **19**, 3981–4003, https://doi.org/10.5194/ACP-19-3981-2019.

Wexler, A. S., F. W. Lurmann, and J. H. Seinfeld, 1994: Modeling urban and regional aerosols, I, Model development. *Atmos. Environ.*, **28**, 531–546, https://doi.org/10.1016/1352-2310(94)90129-5.

Yue, J., J. Russell III, Y. Jian, L. Rezac, R. Garcia, M. López-Puertas, and M. G. Mlynczak, 2015: Increasing carbon dioxide concentration in the upper atmosphere observed by SABER. *Geophys. Res. Lett.*, **42**, 7194–7199, https://doi.org/10.1002/2015GL064696.

Zarzycki, C. M., C. Jablonowski, and M. A. Taylor, 2014: Using variable-resolution meshes to model tropical cyclones in the Community Atmosphere Model. *Mon. Wea. Rev.*, **142**, 1221–1239, https://doi.org/10.1175/MWR-D-13-00179.1.

Zaveri, R. A., R. C. Easter, and L. K. Peters, 2005: A computationally efficient Multicomponent Equilibrium Solver for Aerosols (MESA). *J. Geophys. Res.*, **110**, D24203, https://doi.org/10.1029/2004JD005618.

——, R. C. Easter, J. D. Fast, and L. K. Peters, 2008: Model for Simulating Aerosol Interactions and Chemistry (MOSAIC). *J. Geophys. Res.*, **113**, D13204, https://doi.org/10.1029/2007JD008782.

Zhuang, J., D. J. Jacob, and S. D. Eastham, 2018: The importance of vertical resolution in the free troposphere for modeling intercontinental plumes. *Atmos. Chem. Phys.*, **18**, 6039–6055, https://doi.org/10.5194/ACP-18-6039-2018.