A MUlti-Scale Infrastructure for Chemistry and Aerosols - MUSICA

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Abstract

To explore the various couplings across space, time and between ecosystems in a consistent manner, atmospheric modeling is moving away from the fractured limited-scale modeling strategy of the past towards 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 spatio-temporal scales, coupling to other Earth System components and process-level modularization will allow advances on topics ranging from fundamental research to air quality to climate and is also envisioned to become a platform that addresses the needs of policy makers and stakeholders.

Capsule: MUSICA will become a community infrastructure for simulating atmospheric chemistry. The unification of various spatio-temporal scales, coupling to other Earth System components and process-level modularization will allow significant scientific advances.

1. Introduction

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. Also, predictions of weather and climate often neglect the effects of 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 which represent different Earth system processes, and (4) easily mix-and-match model components. 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 is the motivation behind the 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 which 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 which 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 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 inter-comparison of individual modules in a single framework, thus will open the way to quantifying 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 due to 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 being 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. Several strategic partnerships with prominent research groups and organizations in the USA and elsewhere in the world are being established to foster the development. MUSICA is guided by a Steering Group (Appendix A) with committed representatives of the broad research community and by Working Groups each led by three co-Chairs (Appendix B), who represent the wider research community including national and international universities and research organizations. 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 (e.g., Jöckel et al., 2010; Badia et al., 2017).

2. 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 components (e.g. land/sea atmospheric exchange) is a challenge and requires MUSICA to apply modern software approaches (e.g. object oriented) and scientists and software engineers to closely work together throughout all stages of the design and development.

MUSICA design will enable its components to be connected to any 3-dimensional (3-D) global or regional atmosphere model, or to any 1-D single column or 0-D box model. It will also have the capability to be driven by atmospheric re-analyses. 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 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.

At the heart of MUSICA is the Chemistry and Aerosol Suite (Figure 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 (Section 3c) and aerosol representations (Section 3d) and includes all associated processes (e.g. photolysis rate calculations, nucleation, coagulation, thermodynamics, etc.) in separate modules. It is to be incorporated through a standardized and well-defined interface, thus can be connected to any atmosphere dynamics model and physical component that is compliant with the interface. This modular structure with a standardized interface simplifies the inter-comparison 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 (Section 3b), a common model evaluation and diagnostics framework, and data assimilation capabilities (Section 3g).

3. MUSICA components and working groups

The following Sections provide a brief overview of the currently seven working group topics, expected developments, and challenges and scientific gains expected from the new infrastructure. The working groups are in the process of establishing themselves and are inviting members from the community to become active members.

3a. Model architecture

The development of MUSICA will be enabled by novel thinking in modern computational architectures. 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 will be communicated during the time of model configuration and configuration tests will be 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.

In current models, physical and chemical processes are typically packaged together into a single monolithic code base with complex data interdependencies, but this slows down scientific advancement – if package A is better than package B for (e.g.) surface ozone but package B is better than A for predictions of carbonaceous aerosols, we want to be able to combine the best of both. For MUSICA monolithic codes will be broken into separate interoperable modules (Figure

1), thus a user can choose a set of modules during configuration and also change the order of the module call (e.g. test calling transport routines before or after chemistry routines).

In MUSICA, 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 3-D 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 (Section 3g), and delivery of emissions data (Section 3b). 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 re-gridding solutions themselves.

MUSICA will simplify the user experience, clarify data dependency between parameterizations, make the configurations of scientific model runs and corresponding data sets 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. Lastly, MUSICA and all its components must guarantee open access and provide comprehensive benchmark scenarios, and user guides.

3b. 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 degrees (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 re-gridding, 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 for running a model and reduces disk space needs. 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. In order to accomplish this, we will learn from the examples of the HEMCO (Keller et al., 2014) and HERMES (Guevara et al., 2019) emission models and collaborate with these teams.

Conservative re-gridding tools that can handle unstructured grid meshes with regional refinement capabilities (Figure 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 have been 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 more recent years (e.g., applying sector and pollutant-dependent 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.

It should be considered to integrate emission models into the emission tools. Such models should 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 NO emissions (Section 3e), or dust and sea salt emissions (Mahowald et al., 2006a; Mahowald et al., 2006b). These emissions depend on environmental variables in non-linear ways, requiring special care to make the underlying parameterization scale-aware and linking to land and ocean models.

Dry deposition of gases and aerosols depends on meteorology and biophysics, and needs to be fully consistent with the land and oceanic surface representation and meteorology determining surface fluxes of energy and emission of biogenic compounds. Current dry deposition schemes crudely parameterize the process, leading to poor understanding of the impact of dry deposition on atmospheric chemistry and vegetation. However, the degree to which depositional processes should be resolved for accurate simulation at different scales is uncertain. While recent work points to the value of coupling atmospheric and land models through terrestrial dry deposition (e.g., Paulot et al., 2018; Clifton, 2018), 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.

3c. 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). Therefore, chemical mechanisms should be developed to be more flexible and modular than is currently the case. Such mechanisms 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., 2019) should be pursued.

Perhaps most exciting are the tools with which next-generation chemical mechanisms will likely be built. Traditional methods for mechanism development, where mechanisms are hand-written, starting simple and adding complexity as needed, are a vital, proven tool, and the maintenance and development of these mechanisms (for example, Carbon Bond (Sarwar et al., 2008), SAPRC

(Carter, 2010), RACM2 (Stockwell et al., 1997), MOZART (Emmons et al., 2010), GEOS-Chem (Bates et al., 2019; Wang et al., 2019)) must continue. However, mechanisms developed on the basis of a new paradigm are also coming into existence. Models like GECKO-A (Aumont et al., 2005) and the SAPRC 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 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., 2019) 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 emissions modules – i.e., providing protocols that allow connections to be made between emitted 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.

3d. 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), and sectional approaches (Wexler et al., 1994; Bessagnet et al., 2004). MUSICA will be designed to accommodate any type of aerosol treatment.

Sectional aerosol models such as MOSAIC (Zaveri et al., 2008) represent aerosols in different internally mixed and discrete size bins. They are typically more computationally expensive than modal ones and thus used primarily for research purposes. Modal aerosol representation as frequently used in global climate models (e.g., Neale et al., 2010; Rasch et al., 2019) assumes a log-normal uniform aerosol composition within each size mode. 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. However, millions of particles are needed; therefore, the explicit approach is used primarily within a box-model like configuration although a few recent studies have integrated this approach into 3-D models (Curtis et al., 2017).

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 lifecycle such as nucleation, coagulation, gas-to-particle partitioning, removal mechanisms, and hygroscopic and optical properties. Currently, modules that treat 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.

This prevents fair comparisons and rigorous assessments of different aerosol treatments and also complicates modifying or replacing aerosol treatments in models. Determining which treatment for an aerosol process performs the best, and why, is nearly impossible because many parts of 0-D or 3-D model configurations are different. In addition, it is difficult to gauge which numerical representation of an aerosol process is most appropriate for a particular application or spatio-temporal scale.

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).

A goal of MUSICA is to enable increased flexibility so that more complex and realistic representation of the aerosol lifecycle can be included in air quality simulations and a computationally more efficient representation can be used in climate simulations. 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.

3e. Physics, transport, subgrid- processes

Physics processes and subgrid-scale transport encompass turbulence, gravity waves (Section 3f), convective transport, vertical mixing, and wet scavenging of trace gases and aerosols in stratiform and convective clouds. All of these processes can be sensitive to the grid mesh sizes and therefore are often tuned for optimizing model results. In addition, subgrid-scale processes can affect chemical reactivity and aerosol microphysics when model grid cells are not well mixed. Wet scavenging and lightning nitrogen oxides (NO_x) generation is included here, rather than Section 3b, 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; Fowler et al., 2019), an important activity that needs to be executed for physical processes affecting composition is testing and evaluating parameterizations at grid meshes ranging from 100s km 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 Section 3f). With varying horizontal resolution, the vertical resolution and aspect ratios will also need to change to accurately resolve vertical mixing and the role of convection and turbulence as well as fine-scale chemical processes. 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. Finally, many coarse resolution models have been tuned for lower vertical resolutions, and it needs to be considered how to adapt and modify a boundary layer routine where a variable vertical grid is needed to accurately capture the vertical mixing of atmospheric constituents.

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 100s m. Shallow convective transport is a means of moving trace gases and aerosols to the free troposphere and simultaneously reducing surface mixing ratios. 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. Furthermore, 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). The MUSICA framework will allow further explorations of these issues through its ability to connect to large-eddy simulation models or via regional to local grid refinement.

3f. Whole atmosphere

Gravity waves couple the full vertical extent of the atmosphere through momentum and constituent transport, impacting stratospheric ozone and the drag on low-Earth orbit satellites, while tropopause folds and the Asian monsoon lead to pronounced stratosphere-troposphere exchange of gases and aerosols. The characteristic scales of these circulations are generally finer than those found in global models, despite their global impacts. Regional refinement in MUSICA has the ability to resolve these phenomena, enhancing its value as a tool for supporting multi-scale whole atmosphere science - but it also faces a number of new challenges.

Convection, geostrophic adjustment associated with frontal systems, and flow over topography generate vertically-propagating gravity waves with horizontal wavelengths of tens to hundreds of kilometers. Their amplitudes grow exponentially as they propagate upward into the low densities of the mesosphere and lower thermosphere until they break, shedding their momentum and turbulently mixing the atmosphere. The sources of these waves, their characteristics, and their impacts on momentum, heat, and chemical transport are typically parameterized [Garcia et al. 2014]. These parameterized waves play a major role in driving the Quasi-Biennial and Semi-Annual Oscillations, the meridional circulation of the mesosphere, and vertical transport between the stratosphere, mesosphere, and lower thermosphere [e.g., Fritts and Alexander 2003; Grygalashvyly et al., 2012; Gardner and Liu, 2016].

In the thermosphere, high-energy processes such as photo-dissociation, photo-ionization, and energetic particle precipitation produce atomic oxygen and nitric oxide [Hendrickx et al. 2018]. Gravity waves mix these species downward into the mesosphere and stratosphere, with major impacts on airglow emissions, ozone, and oxidative species [Smith et al., 2013]. The transport of atomic oxygen creates a net transfer of chemical potential energy from the thermosphere to the mesosphere, while nitric oxide transport to the stratosphere may drive ozone destruction [Brasseur and Solomon, 2005]. Water vapor, methane, and carbon dioxide are similarly mixed upward, with long-lived greenhouse gases like sulfur hexafluoride mixed up to their loss regions [Kovács et al., 2017]. Observations indicate these gravity-wave-driven vertical transports throughout the mesosphere and thermosphere are severely underrepresented in models [Smith et al., 2015; Huang et al., 2015; Millán et al. 2015; Yue et al. 2015; Gardner et al., 2016b].

Regional refinement in MUSICA will resolve mesoscale gravity waves, reducing the dependence of many circulations and transports on parameterizations. Since these phenomena could occur anywhere, ideally adaptive grids would be needed 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 region, (2) redevelop the 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 likely evanesce due to the coarsening grid potentially introducing 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].

3g. 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, correlative analysis between different atmospheric quantities as well as data assimilation and inverse modeling. Evaluation is critical to characterize and reduce model errors associated with insufficient scientific understanding, such as uncertainties in input parameters, and model approximations.

By appropriately accounting for the differences in the representation of processes at multiple scales, MUSICA provides key opportunities to overcome some of the current limitations faced when confronting models with observations. It will reduce uncertainties in the representativeness of the modeled and observed states of chemical constituents (e.g., Janjic 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 data sets, for analyzing complex time series and for establishing statistical relations between the concentration of different species. A database of evaluation data sets will be compiled, with the ability for a user to select which data sets are appropriate for the model evaluation (e.g., satellite curtains and maps, aircraft tracks, surface stations, sonde profiles, etc.) Given the selected data sets, output will be sampled at the appropriate co-temporal and co-located points and written to files for easy comparison.

Data assimilation tools will build upon current capabilities, e.g., the Data Assimilation Research Testbed (Anderson et al., 2009), the Community Gridpoint Statistical Interpolation (GSI) tool (Shao et al., 2016), or the Joint Effort for Data assimilation Integration (JEDI -- https://www.jcsda.org/jcsda-project-jedi). Similarly, evaluation and diagnostic tools will complement existing tools, e.g., the CESM diagnostics package (http://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 non-linearity, 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, multi-scale 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 (section 3b). 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.

4. 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. 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 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. 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. Currently, MICM supports various versions of the MOZART gas phase chemistry (Emmons et al., 2020) and a gas-phase chemistry box model version based on MICM (MusicBox) will be released to the community together with MUSICA V0.

Figure 3 shows surface ozone from a 1-degree uniform global resolution simulation, which is compared to a MUSICA v0 simulation with a regional refined grid of ~14 km resolution over the contiguous U.S. embedded in a 1-degree global resolution (as shown in Figure 2a). Despite the fact that both simulations are nudged to meteorological re-analysis, 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.

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. Future developments will require close coordination among all working groups and other data assimilation efforts. We also foresee that a development of an observation package can be leveraged across different activities (model development, evaluation and testing, data assimilation). Inclusion of tangent linear and adjoint modeling capabilities will take devoted development and, like other efforts, continued support.

5. Community involvement and outlook

The development of a multi-scale infrastructure that meets the scientific needs of the entire user community represents an exciting challenge that necessitates strong partnerships among different organizations and different disciplines ranging from lab studies and field experiments to statistics and computational sciences and from molecular chemistry to space physics. 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 endusers. 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 model tutorials (e.g., through regional MUSICA "Hubs").

After the challenges posed by the multi-scale nature of the new infrastructure are met, the atmospheric community will be able to leverage MUSICA to advance our understanding of the multi-scale 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, multi-scale 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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Table 1: Examples of new science applications enabled by MUSICA

Past & 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 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.
Sub-seasonal 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 Nino region) will lead to improved regional climate variability and air-sea coupling and also allow information on exposure relevant scales.

Stratospheric intrusions	
Global models hampered by coarse resolution near UTLS or regional models that are hampered by boundary effects.	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 feedback 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 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, dynamics and between ocean and atmosphere.
Feedback loop of climate change on greenhouse 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; 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	A fully coupled system will allow to account for detailed chemistry/emissions over megacities, and quantify the associated effects on remote regions (e.g., Arctic) and global atmosphere.
Chemical data assimilation (CDA) and evaluation	
CDA in models is generally done separately from meteorological DA and limited to updating atmospheric concentrations.	CDA will be co-developed as integral part of MUSICA with the objective of updating concentrations and inputs (e.g. emissions)

Evaluation tools have been developed separately from DA.	efficiently. Commonalities between DA and evaluation will be addressed in parallel.
Air quality under a changing climate	
Use the Pseudo Global Warming method to provide initial and boundary conditions to a regional AQ model from 'bias-corrected' data from an ensemble mean of climate model simulations.	Conduct an ensemble of simulations under various future scenarios with a single self-consistent model with sufficient resolution to simulate key air quality metrics.
Co-benefits of greenhouse gas emission reduction policies	
Conduct separate inconsistent simulations using regional AQ models and global climate models to investigate impacts on AQ and climate change from the move towards a net-zero-emissions future.	Conduct simulations under net-zero future scenarios with a single self-consistent model with sufficient resolution to simulate key air quality 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)

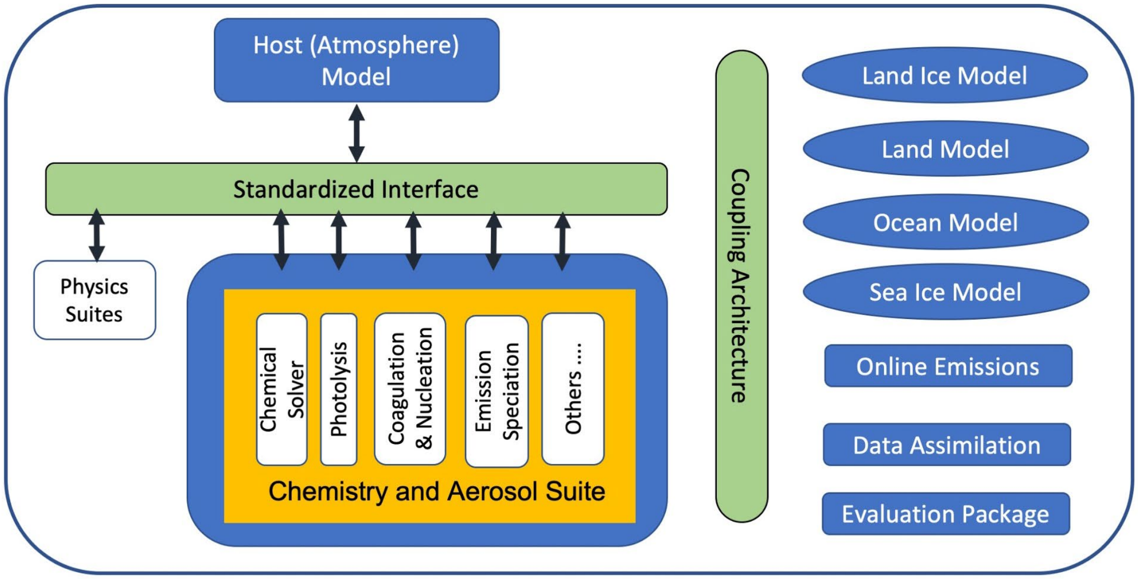

Figure 1: MUSICA defines a framework to integrate chemistry into any atmospheric host model. It includes coupling to emissions, evaluation and data assimilation tools and enables linking to other Earth System components, all through well-defined standardized interfaces. All physics and chemistry processes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

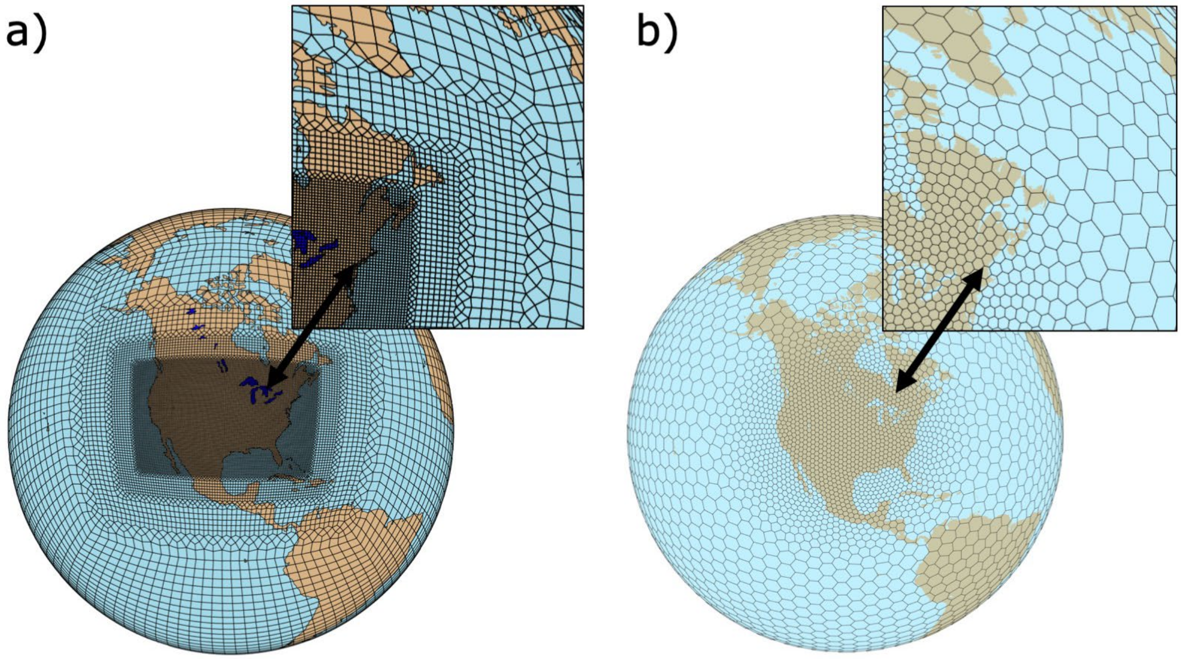

Figure 2: Examples of unstructured grids with regional refinement over the contiguous U.S. (a) the Spectral Element (SE) dynamical core (Zarzycki et al., 2014) and (b) the Model for Prediction Across Scales (MPAS) (Michaelis et al., 2019) dynamical core.

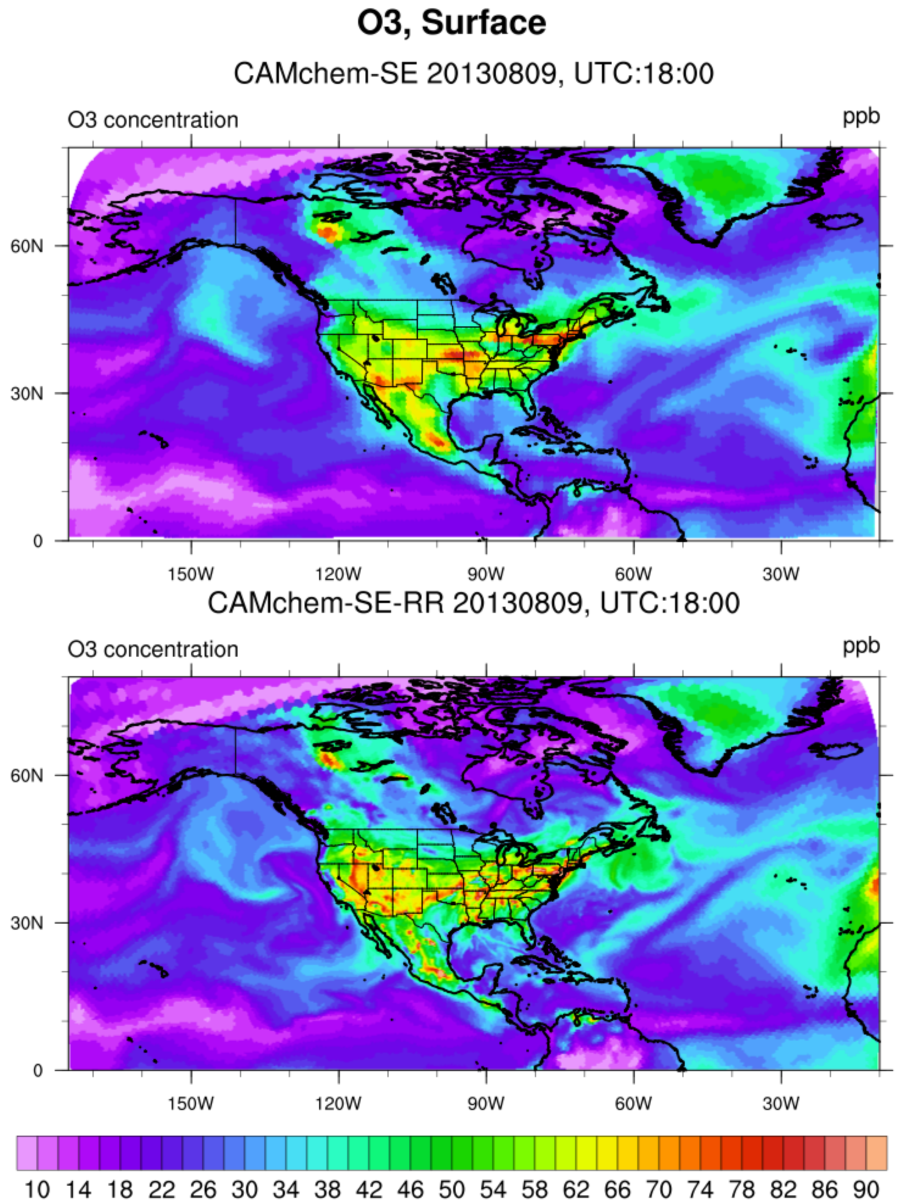

Figure 3: Surface ozone for 9 August 2013 18 UTC simulated at uniform global 1˚ horizontal resolution (top) and with a MUSICAv0 global 1˚ simulation that refines to 14 km resolution over the contiguous U.S. (bottom).