The Community Regional Atmospheric Chemistry Multiphase Mechanism (CRACMM) Collaborative Effort

30 May 2024

Objective

The objective of this collaborative effort is to develop an advanced chemical mechanism to improve simulation and forecasting of air quality to better protect human health and the environment. Both the Environmental Protection Agency (EPA) and National Oceanic and Atmospheric Administration (NOAA) require advanced models to provide timely and accurate air quality forecasts as well as improve our understanding of the drivers of degraded air quality so that effective mitigation strategies can be developed. In air quality modeling, chemical mechanisms are used to represent atmospheric chemical reactions, and how accurately the model makes predictions depends in part on how well those chemical mechanisms approximate the actual chemical processes in the atmosphere. Existing chemical mechanisms are either not sufficiently detailed to accurately represent ozone, particulate matter, air toxics, and species of concern in one system or they are too detailed for select systems to be computationally feasible for forecasting and air quality management applications. For example, disconnects between emissions and chemistry have been identified as a key component hampering a proper representation of particulate matter (PM).[1] The joint activity described here will bring together the latest atmospheric chemistry science, efficient computational methods, and rigorous evaluation and iterative improvement using data from measurement campaigns and satellite-based remote sensing.

The Community Regional Atmospheric Chemistry Multiphase Mechanism[2] (CRACMM; pronounced CRACK-um, www.epa.gov/cmaq/cracmm) version 1 was developed by EPA and NOAA collaborators and released for the first time in October 2022. CRACMM predicts ozone (O_3), particulate matter (PM), chemical components of PM such as secondary organic aerosol (SOA), and air toxics like formaldehyde. EPA and NOAA will continue to collaborate to improve CRACMM by including newly discovered reactions, product yields, chemical properties, emitted species, and other new advances as the state-of-science develops.

A critically important and novel design feature of CRACMM is a focus on accurately simulating and conserving reactive organic carbon (ROC). Along with nitrogen oxides, ROC is the fuel for atmospheric chemistry and responsible for significant amounts of atmospheric reactivity, secondary PM, tropospheric ozone, and secondary carbon dioxide.[3] ROC emissions have also evolved over time as air quality regulations have been enacted. In the past, motor vehicles were dominant emitters of ROC. Today, the relative dominance of ROC emissions has shifted with sources like volatile chemical products (e.g., paints, solvents, adhesives)[4] and wood burning growing in relative importance. EPA and NOAA are also improving methods for chemically speciating ROC emissions and how their chemistry is represented in CRACMM. Recent field campaigns led by NOAA provide data to constrain both the inputs to CRACMM as well as transformation products.

This joint activity between EPA's Office of Research and Development (ORD) Center for Environmental Measurement and Modeling (CEMM) and NOAA's Chemical Science Laboratory (CSL) is consistent with the missions of both EPA and NOAA. EPA-NOAA collaborations on CRACMM fall under a Memorandum of Agreement (MOA) on Cooperation in Forecasting Air Quality (2021) which indicates: "NOAA and EPA agree to collaborate on scientific research… including, but not limited to: developing air quality models and tools for integrating meteorology and air chemistry, including ozone and precursor species, aerosols and their properties." CRACMM also addresses recommendations from the 2023 National Academies of Sciences, Engineering, and Medicine report on Transforming EPA Science to Meet Today's and Tomorrow's Challenges which encourages EPA to: "Improve the predictive capability of CMAQ" through "process-level knowledge" and have "continued collaboration with other organizations to develop measurement and modeling strategies for national air quality" (Recommendation 5-3).

Organizational Structure

The official, public versions of CRACMM will be housed within the CMAQ model released by the EPA ORD CEMM, and documentation will be ported to other locations for broader dissemination. CRACMM is intended to be informed by a broad scientific community with NOAA CSL being a core partner. Thus, NOAA will incorporate CRACMM versions into their work and contribute updates to the central code as part of this effort. See Figure 1 for connections between EPA and NOAA Models interacting with CRACMM. A core group of scientists from each organization will discuss objectives, challenges, preliminary work on a regular basis (at least quarterly each year), culminating in a yearly summary involving directors from both programs and other interested parties. Future directions will be examined on at least a biennial basis coinciding with release of new CRACMM versions. The core group of scientists will produce and revise yearly goals and future objectives within a planning documentation.

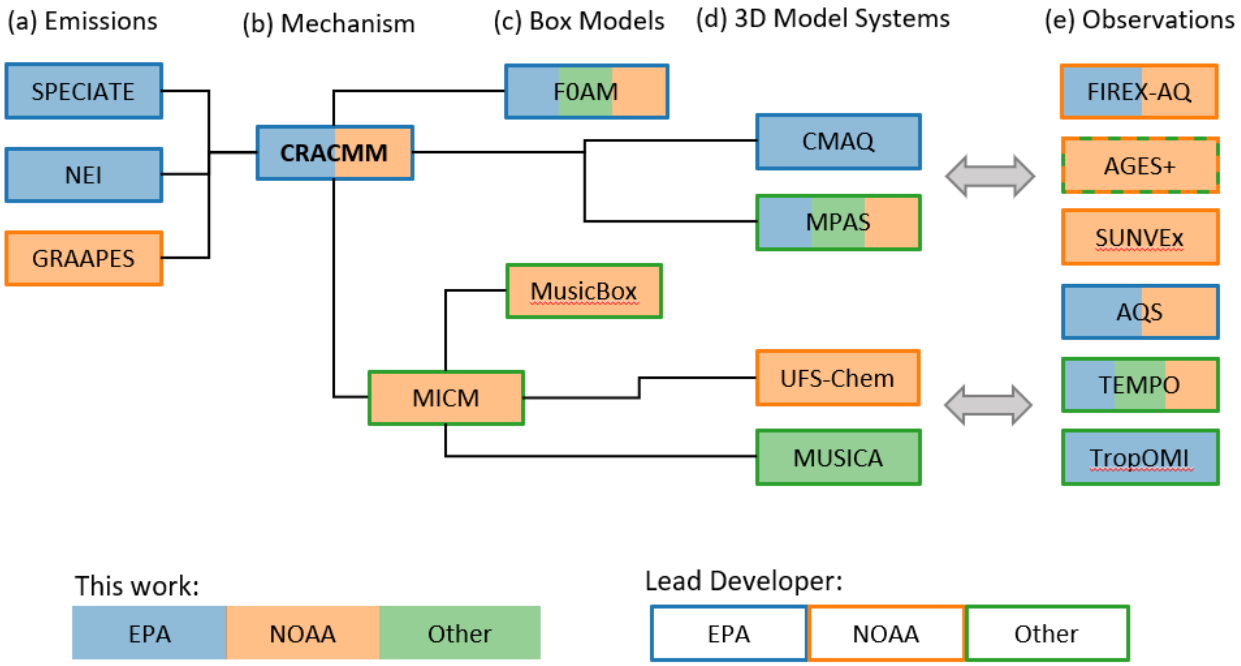

Figure 1: Connections between EPA and NOAA Models interacting with CRACMM. Emissions models and data (a) include: SPECIATE, the National Emissions Inventory (NEI), and the GReenhouse gas And Air Pollutants Emissions System (GRAAPES). The mechanism (b) is the Community Regional Atmospheric Chemistry Multiphase Mechanism (CRACMM). Box models (c) include: the Framework for 0-D Atmospheric Modeling (F0AM) and MusicBox. The Model-Independent Chemistry Module (MICM) is an interface allowing CRACMM to be incorporated in various model systems. 3-D models (d) include: the Community Multiscale Air Quality (CMAQ) modeling system, Model for Prediction Across Scales (MPAS), Unified Forecasting System with Chemistry (UFS-Chem), and Multi-Scale Infrastructure for Chemistry and Aerosols (MUSICA). Observational data (e) include measurements from: FIREX-AQ, AGES+, SUNVEx, AQS, TEMPO, and TropOMI. Outline colors indicate the primary developer of the model or data set. Shaded colors indicate who will lead that item as it relates to CRACMM. External developers include NCAR, NASA, and the scientific community.

Current Capabilities

EPA Center for Environmental Measurement and Modeling (CEMM)

CEMM conducts research to advance EPA's ability to model contaminants in the environment, including research to provide fundamental methods and models needed to implement environmental statutes. CEMM develops EPA's flagship Community Multiscale Air Quality (CMAQ) photochemical modeling system to help manage air quality at multiple scales. CMAQ can estimate the contributions of various air emission sources to air pollution and inform exposure and effects assessments. CEMM distributes CRACMM in CMAQ and as a standalone mechanism from US EPA's github repository (www.github.com/USEPA/CMAQ and www.github.com/USEPA/CRACMM). In addition, EPA CEMM

staff develop methods to estimate emissions for sources including volatile chemical products, mobile sources, and residential wood combustion. CEMM staff co-lead development of the EPA SPECIATE database which is the main source of emission speciation data for regulatory analyses and assessments.

Key EPA CEMM-related personnel:

- Havala Pye, EPA CRACMM lead – will provide overall project management and serve as EPA lead for volatile chemical product and wildfire emissions and chemistry.
- Ben Murphy, Staff Scientist – will lead updates for mobile and residential wood combustion sector emissions, provide support for algorithm development, testing, and documentation, and develop later generation (>1) chemistry for semi- and intermediate volatility compounds.
- Emma D'Ambro, Staff Scientist – will provide technical input on CRACMM chemistry.
- Golam Sarwar, Staff Scientist – will contribute to the development of condensed dimethyl sulfide chemistry for hemispheric simulations with CRACMM.
- Rob Pinder, EPA Supervisory Scientist – will represent the CRACMM effort in EPA management decision-making processes and facilitate overall directions.

NOAA Chemical Science Laboratory (CSL)

NOAA CSL has a long history of studying the sources and chemical evolution of urban, industrial, fire, and biogenic emissions. For example, recent work on urban emissions has assessed the role of consumer and industrial products - termed volatile chemical products (VCPs) - on urban ozone formation.[4] These emissions are less well constrained than some other sources, and the atmospheric chemistry and resulting air quality impacts of these sources remain an important topic of research. NOAA CSL is currently developing mechanisms that account for the reactivity and chemical transformation of the major molecules emitted from VCPs and other understudied urban sources, such as commercial and residential cooking.

NOAA CSL is interested in developing many components of CRACMM in collaboration with EPA including advancing the chemistry of VCPs, cooking, biogenic, and wildfire emissions, and capturing future atmospheric chemistry changes due to lower NO_x levels resulting from regulatory controls. Several NOAA CSL past and upcoming field campaigns have focused on the urban environment, including the Southwest Urban NOx and VOC EXperiment (SUNVEx) 2021, Atmospheric Emissions and Reactions Observed from Megacities to Marine Areas (AEROMMA) Study 2023, and the upcoming Utah Summer Ozone Study (USOS) 2024. These field campaigns will collect detailed gas and aerosol-phase field observations in multiple urban areas across the country, and the datasets will be archived on the NOAA CSL website (https://csl.noaa.gov/field.html). NOAA CSL scientists will work together with EPA scientists to evaluate and advance CRACMM using these detailed field observations under a variety of chemical regimes across the Continental US.

NOAA CSL plans to implement CRACMM into box and 3D modeling tools and use these models to better understand which emission sources contribute most to formation of common air pollutants such as ozone and secondary organic aerosol (SOA). NOAA CSL also plans to use CRACMM to evaluate and inform the design of reduced complexity mechanisms for use in computing-intensive experiments. EPA colleagues have implemented CRACMM into the Framework for 0-D Atmospheric Modeling (F0AM)[5]

and NOAA scientists are comparing this mechanism against other mechanisms used to simulate urban air quality (e.g., RACM-ESRL-VCP and the Master Chemical Mechanism). For 3D modeling, CRACMM will be implemented into NOAA's Unified Forecasting System with chemistry (UFS-Chem) through NCAR's Model Independent Chemistry Module (MICM). MICM will also connect to other community modeling tools including MusicBox and the Multi-Scale Infrastructure for Chemistry Modeling (MUSICA), which will encourage the use and development of CRACMM by a larger community of scientists. UFS-Chem plans to unify operational and research air quality and atmospheric composition forecasts at NOAA. Inclusion of the CRACMM mechanism in CMAQ and UFS-Chem could help facilitate transition of EPA/NOAA CSL research collaborations on chemical mechanism development into NOAA's air quality forecast operations (i.e., research-to-operations).

Key NOAA personnel:

- Rebecca Schwantes – NOAA CRACMM lead – will lead implementation of CRACMM in MICM/UFS-Chem.
- Matthew Coggon – will lead and facilitate connection of CRACMM with measurements from laboratory and field experiments.
- Chelsea Stockwell, Quazi Rasool, Siyuan Wang, Zachary Decker, Michael Robinson, & Carsten Warneke – will help to improve the CRACMM mechanism and evaluate against field observations.
- Brian McDonald, Atmospheric Composition Modeling program lead – will represent the CRACMM effort in CSL management decision-making processes and facilitate overall directions.
- Steve Brown, Tropospheric Chemistry program lead - will represent the CRACMM effort in CSL management decision-making processes and facilitate overall directions.
- Colin Harkins – will help to improve CRACMM emissions speciation.

University Partners

Collaborations with academic researchers can both contribute to the development and evaluation of CRACMM and provide a broader user base for research applications. For example, CRACMM incorporates an isoprene oxidation mechanism developed as part of the EPA STAR-funded automated atmospheric chemical mechanism reduction (AMORE) project by a team led by Faye McNeill at Columbia University. AMORE uses an algorithm based on graph theory to distill mechanisms too complex and detailed for practical use in large scale models such as CRACMM (e.g. the master chemical mechanism[6] or Caltech isoprene mechanism[7]) down to a manageable size without compromising key outcomes such as production rates of ozone, SOA, and air toxics. The AMORE isoprene scheme was extensively tested against other mechanisms and data from chamber experiments[8] and ambient measurements[9], also by university partners, before its incorporation into CRACMM. AMORE can be used in the future to develop other sub-mechanisms for CRACMM for chemical systems that require extensive reduction from their explicit mechanisms, such as those of monoterpenes, or to rapidly update the reduced mechanisms when new science becomes available.

Further collaborations will take advantage of the broad academic user bases of both F0AM (in which CRACMM and AMORE-isoprene are already available) and GEOS-Chem (a global chemical transport

model, which has already been used to test the AMORE isoprene mechanism). Both models are widely used by university groups to develop mechanisms, compare them against each other, and test them against ambient and experimental observations. Integrating CRACMM with these models opens an avenue to wide-ranging applications of CRACMM throughout the academic community, providing tools by which researchers can evaluate the mechanism themselves and develop changes and additions for further mechanism updates via community contributions. Current collaborators will serve as liaisons to facilitate greater involvement with university partners in the future.

Key university personnel:

- V. Faye McNeill, Columbia University -- leading development of the AMORE tool for mechanism reduction, including creation and testing of new model sub-mechanisms to be integrated into CRACMM.
- Kelvin Bates, University of Colorado Boulder -- integrating CRACMM into GEOS-Chem, performing mechanism intercomparisons on regional to global scales, and fostering partnerships in the GEOS-Chem user community.
- William R. Stockwell, University of Texas El Paso – original developer of underlying RACM mechanism providing technical guidance and continuity in mechanism development approaches.

Tangential Work

CRACMM implementation in NOAA model(s) will be performed in collaboration with the National Center for Atmospheric Research (NCAR) and is covered under a NCAR-NOAA MOA for Co-development of a Common Modeling Infrastructure (2018). CRACMM will also incorporate advancements from the broader scientific community, including, but not limited to, those resulting from the 2019 EPA Chemical Mechanisms to Address New Challenges in Air Quality Modeling Request for Applications (RFA).

Workflow

Activities during Year 1 include:

- Developing improved chemistry for secondary formaldehyde.
- Applying CRACMM to western U.S. wildfires during FIREX-AQ 2019.
- Intercomparing MCM, RACM_ESRL_VCP, RACM2B_VCP, and CRACMM1 in box model form for SUNVEx 2021.
- Facilitating dialogue between UFS-Chem and CMAQ infrastructure groups.
- Broadening participation and linking with the Generalized Aerosol/chemistry iNTerface (GIANT) effort at the International Aerosol Modeling Algorithms (IAMA) meeting.
- Hosting EPA Visitor at NOAA CSL 19-21 September 2023:
    - EPA to provide NOAA-CSL Seminar
    - NOAA to facilitate discussions at NOAA and NCAR
    - Meet with NOAA-CSL management
    - Start development of CRACMM logo

- Developing and posting of this document by the end of calendar year at www.epa.gov/cmaq/cracmm and elsewhere if needed.
- Preparing an EM article introducing atmospheric chemical mechanisms, their development, usage, and future needs.

In subsequent years, CRACMM2 will be publicly released (2024), CRACMM2 will be applied to the 2023 NOAA field campaigns, CRACMM will be implemented in MICM and UFS-Chem, and additional mechanism intercomparisons will be performed. EPA and NOAA will increasingly apply CRACMM to new domains under new conditions and coordinate updates for CRACMM3. A major coordination and planning effort, led by EPA and NOAA but open to a broader community, is anticipated in January-July 2025 to prepare for CRACMM3 in the 2026 CMAQ release. See the CRACMM Fact Sheet (https://www.epa.gov/system/files/documents/2023-10/cracmm-factsheet_20231012.pdf) for information on CMAQ release cycles.

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