Final Report for "Cloud-Aerosol Physics in Super-Parameterized Atmospheric Regional Climate Simulations (CAP-SPARCS)" (DE-SC0002003) for 8/15/2009 through 8/14/2012

1. DOE Award Number: DE-SC0002003.
Name of Recipient: Scripps Institution of Oceanography, University of California, San Diego.
Project Title: Cloud-Aerosol Physics in Super-Parameterized Atmospheric Regional Climate Simulations (CAP-SPARCS)
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Collaborators: Steve Ghan (PNNL), Phil Rasch (PNNL).

2. There are no authorized distribution limitation notices.

3. Executive Summary

Description of the Project: Improving the representation of local and non-local aerosol interactions in state-of-the-science regional climate models is a priority for the coming decade (Zhang, 2008). With this aim in mind, we have combined two new technologies that have a useful synergy: (1) an aerosol-enabled regional climate model (Advanced Weather Research and Forecasting Model with Chemistry WRF-Chem), whose primary weakness is a lack of high quality boundary conditions and (2) an aerosol-enabled multiscale modeling framework (PNNL Multiscale Aerosol Climate Model (MACM)), which is global but captures aerosol-convection-cloud feedbacks, and thus an ideal source of boundary conditions. Combining these two approaches has resulted in an aerosol-enabled modeling framework that not only resolves high resolution details in a particular region, but crucially does so within a global context that is similarly faithful to multi-scale aerosol-climate interactions. We have applied and improved the representation of aerosol interactions by evaluating model performance over multiple domains, with (1) an extensive evaluation of mid-continent precipitation representation by multiscale modeling, (2) two focused comparisons to transport of aerosol plumes to the eastern United States for comparison with observations made as part of the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT), with the first being idealized and the second being linked to an extensive wildfire plume, and (3) the extension of these ideas to the development of a new approach to evaluating aerosol indirect effects with limited-duration model runs by “nudging” to observations. This research supported the work of one postdoc (Zhan Zhao) for two years and contributed to the training and research of two graduate students. Four peer-reviewed publications have resulted from this work, and ground work for a follow-on project was completed.

How this Research Adds to the Understanding of Climate Change: Understanding climate change on regional scales requires a broad scientific consideration of anthropogenic influences that goes beyond greenhouse gas emissions to also include the effects of aerosol. For instance, it is now clear that on small scales, human-induced aerosol plumes can exert microclimatic radiative and hydrologic forcing that rivals that of greenhouse gas–forced warming. This project has made significant scientific progress by investigating how both multiscale modeling frameworks and linked regional-global scale modeling can be used to improve prediction of how aerosols, clouds, and climate will mutually interact to produce climate change at regional scales in response to anthropogenic forcing.

4. Comparison of the Actual Accomplishments with the Objectives

The *project objectives* were to improve scientific models about the potential response of the Earth's climate to increased greenhouse gas levels by:

1. *Improving the methods available for simulating regional variability of aerosol-cloud interactions by developing a new formulation for ultra-high resolution modeling, namely a unified regional/global aerosol-climate modeling framework.* The actual accomplishments of the project include the development of the unified regional/global aerosol-climate modeling framework as described by Zhao et al. (2013).
2. *Improving the physical formulations of aerosol-cloud processes by intercomparing and evaluating regional climate simulations with observational data sets.* The actual accomplishments of the project include intercomparisons and evaluations of regional climate simulations with observational data sets, as described by Zhao et al. (2012) and Zhao et al. (2013).
3. *Improving simulations by the addition of better representations of aerosol types by incorporating global transport of aerosols to regional sub-domains and by allowing MACM-imposed aerosol transport to influence the WRF-Chem predictions of regional climate variability*. The actual accomplishments of the project include incorporating global transport of aerosol to regional sub-domains, as described by Zhao et al. (2013).

Overall, the project accomplished more detailed comparisons to the International Consortium for Atmospheric Research on Transport and Transformation (ICARTT) than were originally proposed, with the results contributing to two peer-reviewed publications (Zhao et al., 2012; 2013). As a result of our initial model evaluation studies, we decided that comparisons to observations during Northern California and the Cloud Indirect Forcing Experiment (CIFEX) and Indian Subcontinent during the Indian Ocean Experiment (INDOEX) would be insufficient to address the aerosol-cloud modeling problems we were targeting, and we instead expanded the scope of our empirical validation effort by developing a new method for global comparisons to observations (Kooperman et al., 2012). This new method uses nudging to dramatically reduce the run time required for quantifying aerosol-cloud indirect effects, hence making it possible to use the MACM improvements on aerosol representation for global radiative forcing changes (despite the intensive CPU requirements that limit runs to a few years rather than the decades required for accurate simulations of climate change). This advance was well beyond what was proposed, but will clearly be a lasting impact from our project.

5. Summary of Project Activities

This research supported the work of one postdoc (Zhan Zhao) for two years and contributed to the training and research of two graduate students. Four peer-reviewed publications have resulted from this work, and ground work for a follow-on project was completed.

Completed Studies

Zhao et al. (2012) investigated the importance of vertical positioning and composition of nonlocal aerosol plumes at WRF-Chem lateral boundaries in regional climate simulations with explicit convection. The potential impacts of nonlocal aerosol plumes on WRF-Chem simulated clouds, precipitation, temperature, as well as the particle scavenging of convection on aerosol plumes, were also explored. Using the Weather Research and Forecasting model with Chemistry (WRF-Chem), we explored the impacts of nonlocal aerosol plumes transported at three different altitudes on a summertime convective system developed in a clean environment over the northeastern United States. Idealized aerosol plumes from forest fire and volcano emissions, which are known to be frequently transported in this region, were prescribed at three separate altitudes on the upstream boundary of WRF-Chem. The low-altitude (1.5–2.5 km) plume characteristic of forest fire emissions intersects the water clouds, resulting in optically thicker clouds and about a 30% decrease in accumulated precipitation. The precipitation response to the idealized aerosol plume is attributed to the aerosol “second indirect effect” and aerosol-induced enhancement in evaporation efficiency. Convection also significantly impacted this low-altitude aerosol plume, because wet removal scavenges up to 70% of plume aerosols over regions where deep convection and precipitation occur. In stark contrast, mid-altitude (5.6–6.6 km) and high-altitude (11.5–12.5 km) plumes exerted a negligible effect on clouds and precipitation. The apparent highly nonlinear sensitivity of simulated convection to the vertical positioning of nonlocal aerosol plumes is explained in terms of the dominant controls influencing this convection regime and limitations in the microphysics currently implemented in WRF-Chem.

Pritchard et al. (2011) evaluates the character of mesoscale organized convection in the Central United States – our target analysis region for wildfire plume interactions with rainfall. The results suggest the superparameterization technique used in the MMF can successfully capture intermittent organized convective events. Hence the MMF approach has the potential to simulate washing the atmosphere of aerosols with a characteristic space-time pattern that is not accessible to conventionally parameterized global models. In the lee of major mountain chains worldwide, diurnal physics of organized propagating convection project onto seasonal and climate timescales of the hydrologic cycle, but this phenomenon is not represented in conventional global climate models (GCMs). Analysis of an experimental version of the Super-Parameterized- (SP-) Community Atmosphere Model (CAM) demonstrates that propagating orogenic nocturnal convection in the Central US warm season is however representable in GCMs that use the embedded explicit convection model approach (i.e. Multiscale Modeling Frameworks; MMFs). SP-CAM admits propagating organized convective systems in the lee of the Rockies during synoptic conditions similar to those that generate mesoscale convective systems in nature. The simulated convective systems exhibit spatial scales, phase speeds, and propagation speeds comparable to radar observations, and the genesis mechanism in the model agrees qualitatively with established conceptual models. Convective heating and condensate structures are examined on both resolved scales in SP-CAM, and coherently propagating cloud ``meta-structures'' are shown to transcend individual cloud resolving model arrays. In reconciling how this new mode of diurnal convective variability is admitted in SP-CAM despite the severe idealizations in the cloud resolving model configuration, an updated discussion is presented of what physics may transcend the re-engineered scale interface in MMFs. We suggest that the improved diurnal propagation physics in SP-CAM are mediated by large-scale first-baroclinic gravity wave interactions with a prognostic organization lifecycle, emphasizing the physical importance of preserving ``memory'' at the inner resolved scale.

Kooperman et al. (2012) evaluates the representation of aerosol indirect effects in CAM5 and the MMF. This work relies on the nudging method used to observationally constrain CAM5 and MMF simulations, which drive WRF-Chem with observed meteorological conditions in our unified global-regional aerosol-climate modeling system. Natural modes of variability on many timescales influence aerosol particle distributions and cloud properties, such that isolating statistically significant differences in cloud radiative forcing due to anthropogenic aerosol perturbations (indirect effects) typically requires integrating over long simulations. For state-of-the-art global climate models (GCMs), especially those in which embedded cloud-resolving models replace conventional statistical parameterizations (i.e. multi-scale modeling frameworks, MMFs), the required long integrations can be prohibitively expensive. Here an alternative approach is explored, which implements Newtonian relaxation (nudging) to constrain simulations with both pre-industrial and present-day aerosol emissions toward identical meteorological conditions, thus reducing differences in natural variability and dampening feedback responses in order to isolate radiative forcing. Ten-year GCM simulations with nudging provide a more stable estimate of the global-annual mean net aerosol indirect radiative forcing than do conventional free-running simulations. The estimates have mean values and 95% confidence intervals of -1.19 ± 0.02 W/m^2 and -1.37 ± 0.13 W/m^2 for nudged and free-running simulations, respectively. Nudging also substantially increases the fraction of the world’s area in which a statistically significant aerosol indirect effect can be detected (66% and 28% of the Earth's surface for nudged and free-running simulations, respectively). One-year MMF simulations with and without nudging provide global-annual mean net aerosol indirect radiative forcing estimates of -0.81 W/m^2 and -0.82 W/m^2, respectively. These results compare well with previous estimates from three-year free-running MMF simulations (-0.83 W/m^2), which showed the aerosol-cloud relationship to be in better agreement with observations and high-resolution models than in the results obtained with conventional cloud parameterizations.

Zhao et al. (2013) An aerosol-enabled globally driven regional modeling system has been developed by coupling the National Center for Atmospheric Research's (NCAR) Community Atmosphere Model version 5 (CAM5) with the Weather Research and Forecasting model with chemistry (WRF-Chem). In this new modeling system, CAM5, a state-of-the-science global climate model (GCM) that interactively simulates aerosol emissions, transport, removal, and cloud-interactions, is downscaled to provide initial and boundary conditions for regional WRF-Chem simulations, as shown in Figure 1. CAM5 not only provides the meteorological forcing for WRF-Chem, but also simulates aerosol particle and gas-phase constituents important to cloud microphysics. In this modeling framework, aerosol particle emissions originating outside the WRF-Chem domain can be advected across its lateral boundaries, accounting for a potentially important non-local aerosol source that is neglected in almost all regional studies. This advance in framework specification opens the door to many new experiments that were previously not possible with individual global or regional models. As a test case, the potential impacts of forest fire aerosol particles on non-local regional precipitation and radiation were investigated in WRF-Chem simulations over the Northeastern United States in the summer of 2004. During this period, forest fires in Alaska and Western Canada lofted aerosol particles into the mid-atmosphere, which were advected across the United States by upper level winds. Daily forest fire emissions were implemented in CAM5 and a suite of explicit convection WRF-Chem simulations (with 3-km horizontal resolution) was conducted to explore the sensitivities of clouds and meteorological feedbacks to black carbon and total aerosol concentrations. WRF-Chem simulations that included non-local forest-fire aerosol had domain-mean aerosol optical depths that were nearly three times higher than those without, which reduced peak downwelling shortwave radiation at the surface by ~25 W m^{-2}. Higher aerosol concentrations led to a ~10% reduction in surface precipitation from a convective system in the region, which resulted from an ~8% decrease in domain mean CAPE and a decrease in precipitation efficiency associated with increased (decreased) cloud droplet number concentration (cloud droplet sizes). Although variations in domain-mean cloud liquid water path were negligible (about a 1% decrease), there is evidence that the fire aerosol plume had an impact on cloud vertical distribution. In simulations with interactive aerosol scavenging, approximately 23% of $PM_{2.5}$ was removed by the convection system. When this removal mechanism was turned off, higher aerosol loading resulted in a small increase (~3%) in surface precipitation, which may be attributed to the melting of ice hydrometeors as a result of heating by absorbing aerosols.

<u>Continuing Studies</u>

Pritchard et al. (AGU, 2011) assessed the simulated realism of an acute wildfire plume event in the external component of our modeling system by comparing model output against *in situ* aircraft and satellite data. A large-scale wildfire pollution event was successfully produced with the observed dimensions and positioning, validating our strategy for numerically implementing custom wildfire emissions under real-world synoptic constraints. Sensitivities of the plume structure to super-parameterization were characterized. Evaluating new aerosol physics in climate models is confounded by uncertainties in emission source magnitude, its vertical distribution, and inadequate sampling of long range plume concentrations. A common modeling practice is to tune emission sources and/or vertical distributions on a case-by-case basis to match intermittent downstream observational constraints on concentration and optical depth. However, this practice suffers from a systematic error, in that cloud parameterizations distort the intermittency and intensity of rainfall, potentially misrepresenting the precipitational scavenging sink between emissions source and downstream plume burden constraints. To explore this issue, sensitivity experiments bracketing the effect of cloud super-parameterization (i.e. embedded explicit convection) were carried out to examine the response of an acute 2004 Arctic plume transport event to changes in the representation of scavenging. These preliminary findings are illustrated in Fig. 3. This work was inspired by the findings of this DOE project, but is beyond the scope of the proposed work. Mike Pritchard, as a new faculty member at UC Irvine, plans to seek future funding to complete this work.

Other Funding and Activities that Contributed to this Project

We have been able to achieve an important degree of synergy between this DOE project and a major NSF initiative in global climate modeling. This leverage has benefited both projects and has also greatly enriched the educational experience of two graduate students. The NSF project is the Center for Multiscale Modeling of Atmospheric Processes (CMMAP), which is described at www.cmmap.org and which provided us with supercomputer resources and an advanced global climate model, the CMMAP Multiscale Modeling Framework (MMF). CMMAP is a large NSF Science and Technology Center with a ten-year lifetime. It focuses on improving the representation of cloud processes in climate models.

Richard Somerville is the primary advisor of the two Scripps graduate students, Michael Pritchard and Gabriel Kooperman. CMMAP has provided the bulk of the financial support for both students. Our DOE project provided opportunities for them to: (1) visit PNNL and meet with Ghan, and in the case of Kooperman establish a collaboration on nudging; (2) learn about regional modeling by working with Zhan Zhao (who was funded by this DOE project); and (3) learn about aerosols by working with Russell. None of these opportunities would have existed without DOE support. Lynn Russell is a member of the Ph. D. dissertation committees for both Pritchard and Kooperman, and it is primarily due to her expertise and advice that their Ph. D. dissertation research has been broadened to include aerosol-cloud-climate interactions. Pritchard defended his Ph. D. dissertation in August 2011 and has recently accepted a faculty position at the University of California, Irvine. Our DOE project deserves credit for playing a key role in their research and the publications resulting from it, and in their

educational experience as graduate students. Dr. Sam Iacobellis of Scripps has received partial salary support from this project, for which he provided expertise and observational data from the DOE ARM archive. Iacobellis has 25 years of experience in making use of ARM data for the development and evaluation of improvements in cloud-radiation parameterizations.

<u>Figures</u>

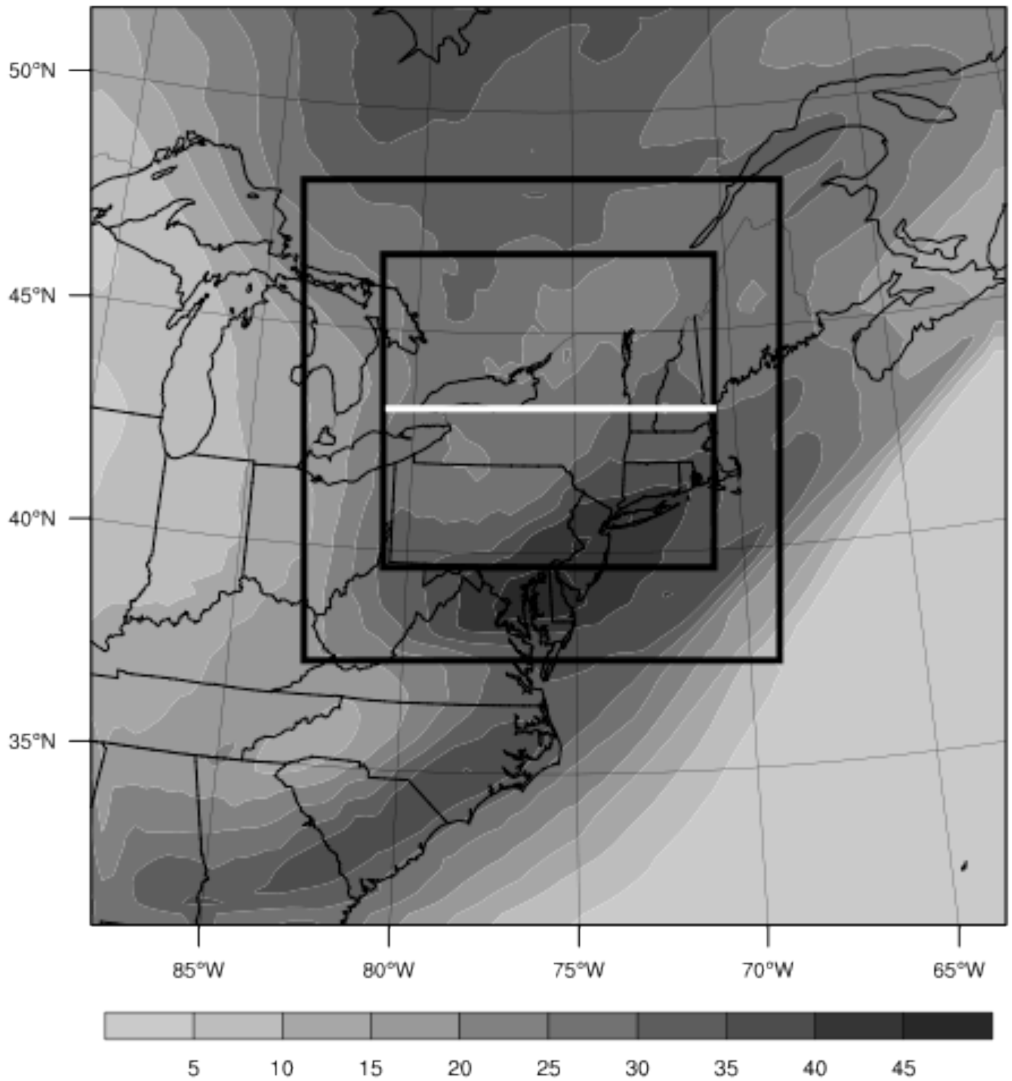

Figure 1: WRF-Chem nested domain and the difference of $PM_{2.5}$ total column burden (mg m^{-2}) from FIRE simulation relative to NoFIRE temporally averaged for 21 and 22 July 2004.

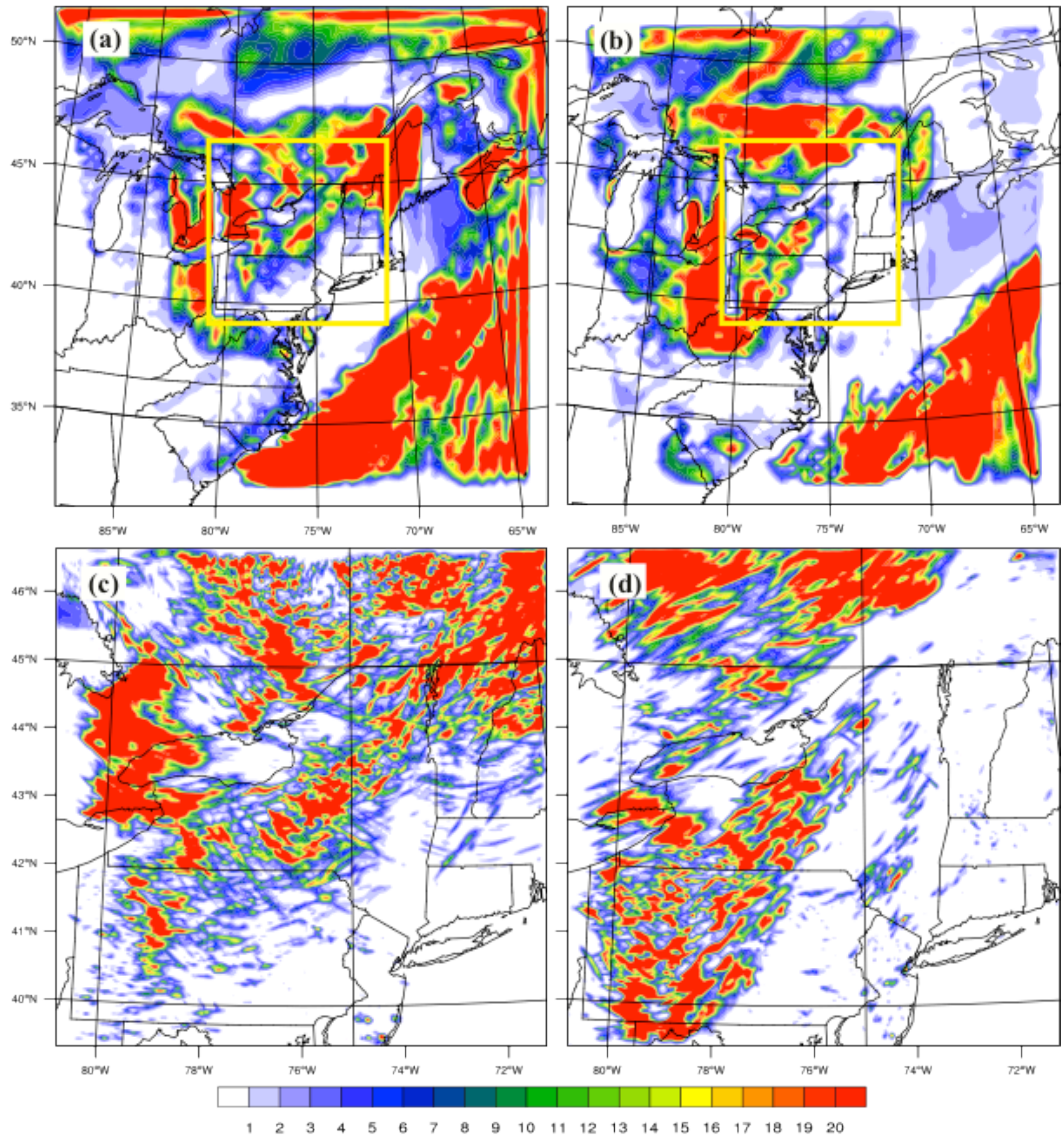

Figure 2: Total precipitation (mm) during the 2-day simulation period for the outermost WRF-Chem domain from (a) CAM_driven and (b) NARR_driven, and for the innermost WRF-Chem domain from (c) CAM_driven and (d) NARR_driven. Yellow boxes in (a) and (b) indicate the region represented in (c) and (d). Adapted from Zhao et al. (2013).

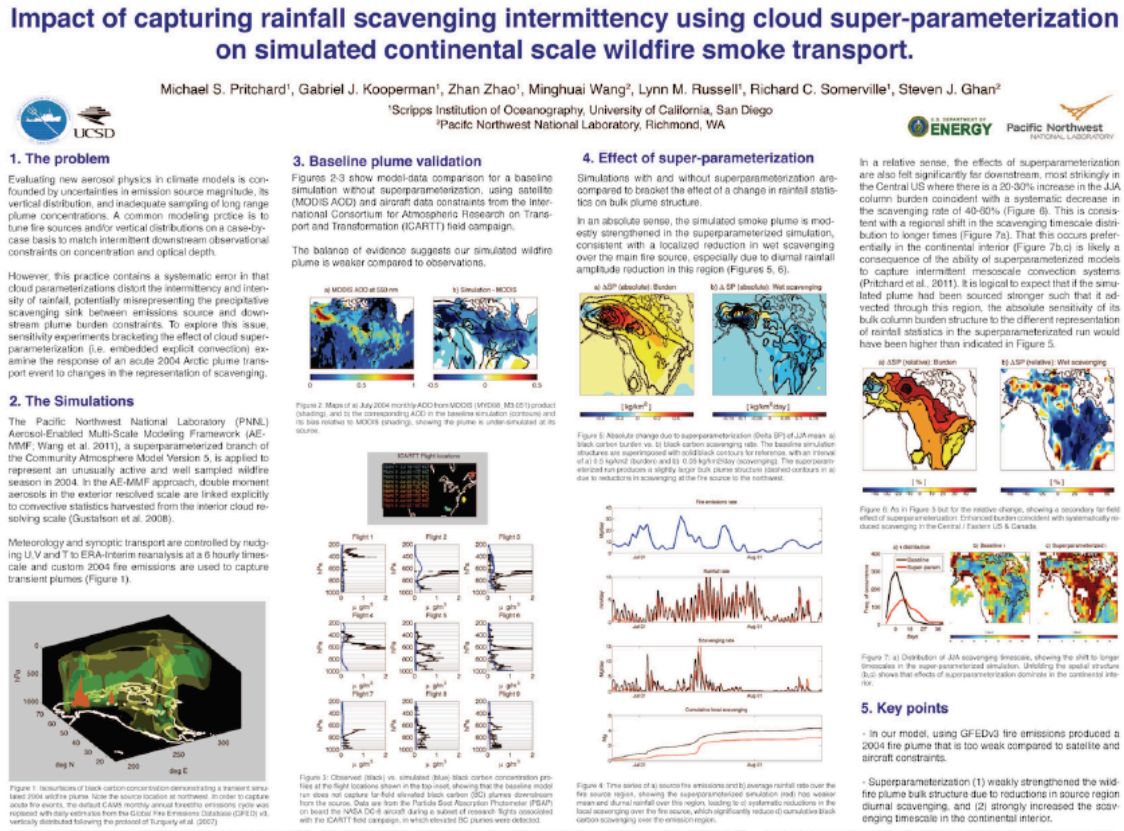

Figure 3: Poster presented at AGU in December 2011 by Mike Pritchard (Prichard et al., AGU 2011).

6. Products developed under the award: Publications, conference papers, or other public releases of results.

Publications:

Zhao, Z., M. S. Pritchard, and L. M. Russell (2012), Effects on precipitation, clouds, and temperature from long-range transport of idealized aerosol plumes in WRF-Chem simulations, *Journal of Geophysical Research-Atmospheres,* **117**, D05206, doi:10.1029/2011JD016744.

Pritchard, M. S., M. W. Moncrieff and R. C. J. Somerville (2011). Orogenic propagating precipitation systems over the US in a global climate model with embedded explicit convection. *Journal of the Atmospheric Sciences* **68** (8), 1821-1840.

Kooperman, G. J., M. S. Pritchard, S. J. Ghan, M. Wang, R. C. J. Somerville, and L. M. Russell (2012). Constraining the influence of natural variability to improve estimates of global aerosol indirect effects in a nudged version of the Community Atmosphere Model 5, *Journal of Geophysical Research-Atmospheres,* in press.

Zhao, Z., G. J. Kooperman, M. S. Pritchard, R. C. J. Somerville, and L. M. Russell (2013), Investigating impacts of forest fires in Alaska and Western Canada on regional weather over the Northeastern United States using CAM5 global simulations to constrain transport to a WRF-Chem regional domain, *Journal of Geophysical Research-Atmospheres,* in revision.

Conference papers

Pritchard, M. S., G. J. Kooperman, Z. Zhao, M. Wang, L. M. Russell, R. C. J. Somerville, and S. J. Ghan (2011). Impact of capturing rainfall scavenging intermittency using cloud super-parameterization on simulated continental scale wildfire smoke transport, *2011 Fall Meeting of the American Geophysical Union*, San Francisco, CA, December 2011.

7. For projects involving computer modeling:

a. Model description, key assumptions, version, source and intended use

Our CAPSPARCS project aims to study the importance of non-local, large-scale aerosol plumes on regional meteorology. Since no single numerical model was available to tackle this problem in its entirety we combined several mature subcomponent models from the community into an integrated system. These included two global models: the Community Atmosphere Model version 5 (CAM5) and a super-parameterized version of CAM5 called a Multiscale Modeling Framework (MMF); and one regional model: the Weather Research and Forecasting Model with Chemistry version 3.1 (WRF-Chem).

All of the above models are uncontroversial, widely validated research tools with proven track records in the literature. A full scientific description of CAM5 is given by *Neale et al.* [2010]. The MMF is a branch of CAM5 that uses embedded explicit convection (*Wang et al.* 2011a). WRF is a regional numerical weather prediction model developed through a multi-institutional endeavor headed at NCAR and is used for both forecasting and climate research applications. WRF-Chem is a modified version of the WRF model, which includes online chemistry, capable of simulating aerosol-cloud-radiation direct and indirect interactions at cloud-resolving scales. The chemistry component of the WRF model is described by *Grell et al.* [2005] and *Fast et al.* [2006].

To combine the above subcomponents into a unified system we leveraged conventional dynamical downscaling techniques (Zhao et al. [2011]) with a custom chemical downscaling scheme described in further detail below. In this new modeling framework, aerosol particle emissions originating outside the WRF-Chem domain can be advected across its lateral boundaries, accounting for a potentially important non-local aerosol source that is neglected in almost all regional studies.

b. Performance criteria for the model related to the intended use

While each of the subcomponents in our system have been validated throughout their development (WRF-Chem: *Grell et al.* [2005][1], *Fast et al.* [2006]; CAM5: *Liu et al.* [2012]; *Ghan et al.* [2012]; MMF5 - *Wang et al.* [2011a, 2011b, 2012]), our integrated system requires independent verification. The key model performance criteria for this project include evaluation against atmospheric observations and mechanisms, specifically involving aerosol-cloud interactions. To appropriately compare the model to satellite and field data a scheme was developed and

[1] An ongoing list of WRF-Chem studies comparing model results to observations can be found at http://ruc.noaa.gov/wrf/WG11/References/WRF-Chem.references_July2012.htm;

implemented to observationally constrain the exterior scale [*Kooperman et al.,* 2012].

c. Test results to demonstrate the model performance criteria were met

The performance of the synoptic constraining algorithm incorporated into CAM5 was validated extensively, and its sensitivities characterized, in *Kooperman et al*. (2012). History matching against *in situ* aircraft data, and satellite data was further verified in *Pritchard et al.* (2011). The sensitivity of the interior model component to variations in aerosol laterally boundary condition positioning was characterized and published in *Zhao et al.* (2012).

The primary references for these test results are:

Kooperman, G. J., M. S. Pritchard, S. J. Ghan, M. Wang, R. C. J. Somerville, and L. M. Russell (2012), Constraining the influence of natural variability to improve estimates of global aerosol indirect effects in a nudged version of the Community Atmosphere Model 5, *Journal of Geophysical Research-Atmospheres*, in press.

Pritchard, M. S., G. J. Kooperman, Z. Zhao, M. Wang, L. M. Russell, R. C. J. Somerville, and S. J. Ghan (2011). Impact of capturing rainfall scavenging intermittency using cloud super-parameterization on simulated continental scale wildfire smoke transport, *2011 Annual Meeting of the American Geophysical Union*, San Francisco, CA, December 2011.

Zhao, Z., M. S. Pritchard, and L. M. Russell (2012), Effects on precipitation, clouds, and temperature from long-range transport of idealized aerosol plumes in WRF-Chem simulations, *J. Geophys. Res.*, 117, D05206.

A third paper is currently in preparation evaluating the performance of the combined (CAM5 and WRF-Chem) global-regional aerosol-climate modeling system:

Zhao, Z., G. J. Kooperman, M. S. Pritchard, L. M. Russell, and R. C. J. Somerville (2013), Investigating impacts of forest fires in Alaska and Western Canada on regional weather over the Northeastern United States using CAM5 global simulations to constrain transport to a WRF-Chem regional domain, in preparation.

d. Theory behind the model, expressed in non-mathematical terms

Regional climate modeling studies suffer from the need to prescribe lateral boundary conditions and are therefore limited by the quality of those boundary conditions, which, for aerosol particles and chemistry, are poorly observed in space and time. While in-situ surface aerosol emissions emanating within the regional domain might be reasonably constrained to observations, the transport of distantly-produced pollutants across the lateral boundaries of the domain remains crudely represented by idealized profiles or offline chemical transport models. Our solution to this problem is to downscale global CAM5 and MMF simulations to provide meteorological and chemical boundary conditions to WRF-Chem, an aerosol enabled regional climate model. In combination these models form a global-regional aerosol-climate modeling system that not only resolves high-resolution details in a particular region, but crucially does so while also remaining faithful to long-range non-local global aerosol-climate interactions.

e. Mathematics to be used, including formulas and calculation methods

For mathematical details regarding individual model subcomponents see references *Neale et al.* [2010] for CAM, *Wang et al.* [2011a] for the MMF, and *Grell et al.* [2005] for WRF-Chem.

Our coupling interface was developed so that mutually consistent meteorological and chemical lateral boundary conditions for WRF-Chem could be obtained from CAM5 and MMF simulation results. The development of the meteorological part of the interface was based on the method described in *Zhao et al.* [2012]. Three-dimensional GCM meteorological fields treated in this interface provided WRF-Chem lateral boundaries including temperature, wind, geopotential height, humidity, and mixing ratios of hydrometers (cloud, ice, rain, snow, and graupel). Two-dimensional GCM meteorological variables used as lateral boundaries for WRF-Chem included surface pressure, sea-level pressure, skin temperature, terrain height, 2-m temperature, surface temperature, sea surface temperature, and ice fractions. In addition, within this interface GCM land surface fields provided soil temperature, soil moisture, land sea mask, 2-m moisture, and water equivalent snow depth for WRF-Chem. GCM 2-dimensional variables were horizontally interpolated to WRF-Chem spatial resolution along lateral boundaries of the outermost WRF-Chem domain, while both horizontal and vertical interpolation were applied to obtain WRF-Chem lateral boundaries from CAM5/MMF for three-dimensional variables.

WRF-Chem's aerosol treatment was configured for consistency with CAM5/MMF, i.e. the Modal Aerosol Dynamics Model for Europe Secondary Organic Aerosol Model (MADE/SORGAM) aerosol module was selected, which also uses three modes (Aitken, accumulation, and coarse mode) to represent aerosol size distributions. GCM aerosols treated in the interface included masses of black carbon, dust, sea salt, sulfate, and primary and secondary organic matter, as well as total aerosol number concentrations. Sea salt aerosols were partitioned to derive sodium and chloride for WRF-Chem, assuming that 31% (55%) of sea salt mass was sodium (chloride) based on *Lewis and Schwartz* [2004]. Instead of predicting both ammonium and sulfate aerosols as in WRF-Chem, the aerosol module in CAM5/MMF predicts the mass mixing ratio of sulfate aerosols in the form of ammonium bisulfate (NH4HSO4) [*Neale et al.,* 2010]. Therefore, the mixing ratio of ammonium is calculated in the interface based on the mass mixing ratio of sulfate aerosols and molar mass composition of ammonium bisulfate. Gas-phase species are expressed in the format of mass mixing ratio (in kg kg^{-1}) and volume fraction (in ppmv) in CAM5/MMF and WRF-Chem, respectively. The following formula is applied to convert gas-phase species from kg kg^{-1} to ppmv:

$$\phi = \frac{R \times (273.15 + T_c) \times 10^6}{\rho \times M \times 12.187} \qquad (1)$$

where φ is volume fraction in ppmv, R is mass mixing ratio in kg kg^{-1}, T_c is temperature in °C, ρ is air density in kg m^{-3}, and M is molecular weight of the gas-phase species.

f. Whether or not the theory and mathematical algorithms were peer reviewed, and, if so, include a summary of theoretical strengths and weaknesses

All three subcomponent models have been extensively peer reviewed. An ongoing list of select publications for each model can be found at:

CAM (http://www.cesm.ucar.edu/publications/),
MMF (http://www.cmmap.org/research/pubs-mmf.html),
and WRF (http://ruc.noaa.gov/wrf/WG11/References/WRF-Chem.references_July2012.htm).

g. Hardware requirements

All three models were integrated using distributed resources available through the National Science Foundation's XSEDE network. We ran the code on a Cray XT5 at the University of Tennessee's. In all experiments CAM5 was configured at 1.9˚x2.5˚ horizontal resolution with 30 hybrid vertical levels. The MMF was configured with the outer GCM settings the same as CAM5 and the inner CRM arranged in two dimensions with 32 vertical columns spaced at 4 km horizontal resolution. WRF-Chem was configured with three nested domains with resolutions of 27 km, 9 km and 3 km for domains 1 to 3, respectively. The finest resolution (3km) interior domain encompassed the Northeastern United States and used a 20 second time step. Under this configuration, and after optimization on our system architecture, the computational expense per one day simulation was 3 CPU-hours for CAM5 run at 96 processor scale, 600 CPU-hours for the MMF at 1152 processor scale, and 1250 CPU-hours for WRF-Chem at 32 processor scale. All three models have excellent check pointing and restart-capability such that long or expensive integrations could be reliably separated into shorter segments.

h. Documentation (e.g., user guide, model code)

The CAM user guide and model code are available on the NCAR website at http://www.cesm.ucar.edu. The MMF uses CAM as its host GCM and the System for Atmospheric Modeling (SAM) as its internal CRM. Documentation for SAM can be found at http://rossby.msrc.sunysb.edu/~marat/SAM.html. The MMF code is managed by the CMMAP community (www.cmmap.org). The WRF user guide and model code can be found at http://www.wrf-model.org.

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