Final Report
Grant #: NNX16AG31G
Title: QUANTIFYING AND ATTRIBUTING VARIABILITY AND TRENDS IN AEROSOL PARTICLE CHARACTERISTICS OF RELEVANCE TO CLIMATE FORCING
PI: Professor Sara C. Pryor, Cornell University
Project officer: Dr. Lucia S. Tsaoussi
Grant start date: 15 May 2016.

1 Project abstract (taken from proposal)

Expected significance: Aerosol particles are major players within Earth's climate system, and in large concentrations, can severely impact human health. However, their concentrations and properties are highly variable spatially (horizontal and vertical) as well as temporally, making it difficult to quantify their regional climate forcing and human health effects. In research component (RC) 1 we will quantify their spatial and temporal scales of coherence over the contiguous US and thus build regional climate change indicators and improve understanding of the predictability, length scales and covariance structures of aerosol properties at regional and smaller scales. In RC 2-4 we will focus on improved understanding of the causes of, and controls on, extreme aerosol events (i.e. unusually high aerosol loading in a specific place, at a given time), during which climate and health impacts are magnified. We will quantify the horizontal and temporal extent of different extreme event types, how these properties vary as a function of aerosol type, source strength, and meteorological conditions, and develop regional indicators to document how these events have changed through time (RC 2 and 3). Then we will conduct high-resolution numerical modeling to determine controls on extreme event intensity and scale, and how such events may change as a result of future climate change or emission control measures (RC 4).
Relevance to the NCA: Our project will advance assessment-relevant science by improving our ability to quantify regional aerosol climate forcing and human health impacts across the contiguous US. It is responsive to research needs identified in the 3rd NCA and will provide improved and traceable input to forthcoming NCA activities.
Approach: Our research methodology integrates information from the NASA 4-D reanalysis of aerosol properties (MERRA-2 to be released in Summer 2015), with aerosol particle properties from NASA satellite-borne instrumentation (e.g. MODIS, MISR and CALIPSO), and ground observations from AERONET. In addition we will conduct sensitivity analyses and diagnostic numerical modeling using WRF-Chem. Using geospatial statistical tools we will first quantify the scales on which aerosol properties of relevance to climate are coherent in different regions of the US. We will then identify the occurrence of extreme events in different regions, and will characterize the intensity and causes of those events. Using an event type classification we will document changes in event frequency and/or characteristics over the historical record. We will develop and implement key climate change indicators focused on aerosols. Finally, numerical simulations will be used to explore responses in extreme aerosol events to future changes in gas and particle emissions.

2 Project team

2.1 Cornell University: Department of Earth and Atmospheric Sciences

- Dr. Sara C. Pryor (Professor and project PI).
- Dr. Yafang Guo (Post Doc). 22 August 2019 to 21 August 2020. Now a Post Doc at the University of Arizona in the Department of Hydrology and Atmospheric Science.
- Dr. Qinjian Jin (Post Doc). 1 July 2018 to 30 June 2019. Now on the faculty in the Department of Geography and Atmospheric Science at University of Kansas.

- Dr. Diana Bernstein (Post Doc). 15 November 2016 to 14 November 2017. Diana subsequently took a Post Doctoral Associate position in the Department of Biological and Environmental Engineering at Cornell University, current location unknown.
- Mr. Ryan C Sullivan (PhD candidate). Ryan graduated with his PhD in August 2017, and is now an Assistant Atmospheric Scientist at Argonne National Laboratory.
- Mr. Lance Nino (Undergraduate student). Lance conducted two independent research study linking satellite Aerosol Optical Depth (AOD) to near-surface $PM_{2.5}$ concentrations, he was subsequently awarded a NASA REU and is in graduate school at Colorado State University.

2.2 Indiana University: Pervasive Technology Institute

- Mr. Robert Henschel (Director for Science Community Tools and project co-PI)
- Mr. Abhinav Thota (Team member, Scientific Applications and Performance)
- Mr. David M Reagen (Team member, Advanced Visualizations Laboratory)

2.3 NASA

- Dr. Robert C. Levy (Research Physical Scientist, and project co-PI)
- Dr. Arlindo M. da Silva (Research Meteorologist and project collaborator)

2.4 Other

Dr. Paola Crippa (Assistant Professor, University of Notre Dame, external collaborator)

3 Project tasks, milestones and products

Table 1. Project tasks, milestones and products.

Task/Milestone	Key Research Publications / Products
1. Analysis of spatial/temporal coherence of aerosol properties in MERRA-2	(Jin and Pryor 2020)
2. Develop and test regional summaries of scales of coherence of aerosol properties that can be used as climate change indicators	(Sullivan et al. 2017)
Milestone: Inherent scales of coherence of aerosol properties (AOD, AE, SSA) quantified and aerosol-based climate change indicators developed	Achieved
3. Determine local & global p90 AOD in MERRA-2 & find periods & regions > p90 (global & local)	Preliminary: (Pryor et al. 2017) Results in: (Guo et al. 2021a) and (Sullivan et al. 2017)
4: Event classification & node identification	
5: Compute distance metrics for each extreme to each event node (within type variability) and quantify frequency by season/year	
6: Meteorological context for each event type	
7: Quantify degree of coherence between surface $PM_{2.5}$ and extreme AOD events	(Jin et al. 2020)
8: Visualizations of extreme events	https://www.youtube.com/watch?v=Xk7jc1Vmxsg
Milestone: Extreme events characterized and occurrence documented. Extreme aerosol event climate change indicators developed.	Achieved
9: Analyze observations for extreme events from Level-2 remote sensing observations	(Guo et al. 2021a; Jin and Pryor 2020; Pryor et al. 2017)
10: Analyze sub-grid scale variability	
11: Integrate data into visualizations	https://www.youtube.com/watch?v=Xk7jc1Vmxsg

Milestone: Extreme events described at high resolution, verification of MERRA-based climate change indicators	Achieved
12: WRF-Chem simulation of each event node	Preliminary: (Pryor et al. 2017) Results in: (Guo et al. 2021a)
13: Sensitivity analyses of simulations	(Crippa et al. 2016; Crippa et al. 2017b; Crippa et al. 2019; Sullivan et al. 2018; Guo et al. 2021b)
14: Integration of simulations in visualizations	https://www.youtube.com/watch?v=Xk7jc1Vmxsg
Milestone: Attribution of drivers of extreme events and sensitivity to emission controls and climate change	Achieved

4 Precis of major activities and research outcomes

Example key knowledge advances obtained through this project are as follows:

4.1 Development of the first aerosol climate change indicators.

The importance of developing robust climate change indicators to track and communicate causes and effects of climate change was strongly emphasized in NCA3. A climate change indicator is defined by the EPA (*https://www.epa.gov/climate-indicators*) as follows: *'An indicator represents the state or trend of certain environmental or societal conditions over a given area and a specified period of time….they provide important evidence of "what climate change looks like."'* Aerosol particles are widely recognized as critical drivers of regional climate and regional climate change. Our project provided a robust framework to develop a climate change indicator for aerosols and applied it to the MERRA-2 reanalysis to provide a first objective index of trends in aerosol properties of relevance to regional radiative balance and thus climate. The aerosol optical properties tracked include estimates of total columnar burden (aerosol optical depth, AOD), dominant size mode (Ångström exponent, AE), and relative magnitude of radiation scattering versus absorption (single scattering albedo, SSA), along with metrics of the structure of the spatial field of these properties. Most of the 8 NCA regions exhibit negative temporal trends in mean and extreme AOD, and SSA (2000-2015), consistent with lower aerosol burdens and transition towards a relatively absorbing aerosol, driven primarily by declining sulfur dioxide emissions. Conversely, more remote regions (e.g. the Northwest) are characterized by increasing mean and extreme AOD that is attributed to increased local wildfire emissions and long-range (transcontinental) transport. Regional and national reductions in anthropogenic emissions of aerosol precursors are also leading to declining spatial autocorrelation in the aerosol fields and increased importance of local anthropogenic emissions in dictating aerosol burdens. A further interesting insight from this work is that synoptic scale meteorological conditions associated with high aerosol burdens are intensifying (becoming more warm and humid), and thus changes in synoptic meteorology may be offsetting aerosol burden reductions associated with emissions legislation.

Key reference: Sullivan R.C., Levy R., da Silva A. and Pryor S.C. (2017): Developing and diagnosing climate change indicators of regional aerosol optical properties. Nature: Scientific Reports **7** art # 18093, doi: 10.1038/s41598-017-18402-x

4.2 Coupling (and decoupling) of columnar AOD and near-surface $PM_{2.5}$.

Previous research has proposed use of satellite-retrieved aerosol optical depth (AOD) to generate geospatial assessments of near-surface $PM_{2.5}$ mass concentrations and potentially to provide air quality forecasts. We examined the degree of coupling of these parameters over the eastern USA and demonstrate that; eta (η, the ratio of $PM_{2.5}$ to AOD), the correlation coefficient (ρ) between daily values of $PM_{2.5}$ to AOD, and hit rate (θ, defined as co-occurrence of high $PM_{2.5}$ and AOD) all exhibit declines over 2000-2017 over the majority of the 301 EPA stations considered. Changes in anthropogenic emissions are disproportionately impacting near-surface air quality and is having lesser impact on total columnar aerosol burdens and thus climate forcing. These findings have profound implications for the potential to use remotely-sensed AOD to generate geospatial estimates of near-surface $PM_{2.5}$ and for making climate projections.

Key reference: Jin Q., Crippa P. and Pryor S.C. (2020): Spatial characteristics and temporal evolution of the relationship between $PM_{2.5}$ and aerosol optical depth over the eastern USA during 2003-2017. *Atmospheric Environment* **239** 117718 doi: 10.1016/j.atmosenv.2020.117718

4.3 Quantifying fidelity and simulation uncertainty for aerosol properties

A key focus of our research is improved quantification of the fidelity of WRF-Chem simulations for key aerosol properties. Critical insights from our work pertain to (i) the response of cloud albedo and structure to new particle formation (NPF, nucleation) and (ii) improved systematic assessment of fidelity and uncertainty in AOD as a function of simulation configuration.

Within this project we conducted very high resolution (4 km grid resolution) simulations with WRF-Chem including a new particle formation mechanism and performed validation relative to detailed measured particle size distributions. Using these validated simulations we found that over the Midwestern USA NPF is not associated with enhancement of regional cloud albedo. Our simulations indicate that NPF reduces ambient sulfuric acid concentrations sufficiently to inhibit growth of preexisting particles to CCN sizes, reduces CCN-sized particle concentrations, and reduces cloud albedo. The reduction in cloud albedo on NPF days results in a domain average positive top of atmosphere cloud radiative forcing, and thus warming of 10 W m^{-2} and up to ~50 W m^{-2} in individual grid cells relative to a simulation in which NPF is excluded.

Within this project we also performed a series of moderate resolution (12 or 24 km grid resolution) simulations with WRF-Chem to explore model fidelity as a function of model configuration and emission estimates. Use of the Modal Aerosol Dynamics Model for Europe/Secondary Organic Aerosol Model scheme leads to highest agreement with Moderate Resolution Imaging Spectroradiometer clear-sky AOD observations particularly when the 2005 NEI is used (for either PBL scheme that we considered). These members of our model ensemble exhibit small negative mean fractional bias over the eastern USA (<2%) and relatively high spatial correlation in summertime mean monthly AOD (>0.5). The aerosol scheme and NEI dominate the ensemble spread in AOD. Near-surface $PM_{2.5}$ is also dependent on PBL scheme and is best reproduced in runs adopting a sectional approach and emissions for 2011. Thus, the WRF-Chem configuration associated with highest agreement with AOD observations is not the same as for $PM_{2.5}$, possibly reflecting the importance of columnar water vapor in dictating AOD or other unexplored uncertainties linking surface mass concentrations to column optical properties.

Key references:

Sullivan R.C., Crippa P., Matsui H., Leung, L.R., Zhao C., Thota A., and Pryor S.C. (2018): New particle formation leads to cloud dimming. *npj Climate and Atmospheric Science* 1:9 doi:10.1038/s41612-018-0019-7

Crippa P., Sullivan R.C., Thota A., and Pryor S.C. (2019): Sensitivity of simulated aerosol properties over eastern North America to WRF-Chem parameterizations. *Journal of Geophysical Research: Atmospheres* **124** 3365-3383.

4.4 Describing extreme aerosol optical depth events and their sensitivity to emission changes

Improved characterization of the spatiotemporal extent, intensity and causes of extreme aerosol optical depth events is critical to quantifying their regional climate forcing and the link to near-surface air quality. We identified and characterized 6 regional-scale extreme AOD events that occurred during 2003-2007 over the eastern United States using MERRA-2 and MODIS. These events were then simulated using WRF-Chem applied at 12 km resolution. Statistical analyses show output from WRF-Chem during these events is generally negatively biased in terms of the mean AOD and $PM_{2.5}$, but WRF-Chem exhibits skill in capturing the peak AOD. WRF-Chem also exhibits fidelity in reproducing the spatiotemporal characteristics of the extreme AOD events in intensity, location of centroid, propagation, duration, and their spatial extension. The events were re-simulated to study the impact of anthropogenic emission changes between 2005 and 2015. An ensemble of simulations was generated where individual and all combined emissions of SO_2, NO_x and NH_3 are perturbed from the 2005 levels to 2015 values. These simulations are used to quantify fractional changes in the spatial and temporal characteristics of mean and peak AOD and near-surface particulate matter ($PM_{2.5}$), as well as changes in radiative forcing. Simulated AOD exhibits a spatially averaged decrease of 39-63% during the six extreme events in response to the combined perturbed emissions. Consistent with our research summarized above in section 2.2, the impact on near-surface $PM_{2.5}$ concentrations is larger, with average decreases of approximately 41% to 69%. Peak AOD is reduced to below 1 in the perturbed simulations from its initial values of 1.73-3.02 in the control runs driven by 2005 emissions. Radiative fluxes at the ground and top-of-the-atmosphere exhibit considerably smaller fractional changes across events, although changes in radiative fluxes during these extreme events are found to be larger than previously reported changes in seasonal mean values over the period 2005 to 2015.

Key references:

Guo Y., Cripa P., Thota A. and Pryor S.C. (2021b): Extreme aerosol events over eastern North America. Part 2: Responses to changing emissions. *Journal of Geophysical Research: Atmospheres* (2020JD033759).

Guo Y., Crippa P., Thota A. and Pryor S.C. (2021a): Extreme aerosol events over eastern North America. Part 1: Characterizing and simulating historical events. *Journal of Geophysical Research: Atmospheres* (2020JD033758).

5 Project management and integration

The project team worked efficiently together as evidenced by our joint publications.

6 Links to other NASA activities

Our project contributed to evaluation of the MERRA-2 product and novel applications of MODIS aerosol data for climate relevant activities and model performance analyses.

7 Research products

†Author was a Post Doc or # was a graduate student when the research was conducted.

7.1 Journal publications

- †Guo Y., Cripa P., Thota A. and Pryor S.C. (2021b): Extreme aerosol events over eastern North America. Part 2: Responses to changing emissions. *Journal of Geophysical Research: Atmospheres* (2020JD033759). *Note publication of this article is delayed due to an unexpected backlog with the AGU production vendor.*
- †Guo Y., Crippa P., Thota A. and Pryor S.C. (2021a): Extreme aerosol events over eastern North America. Part 1: Characterizing and simulating historical events. *Journal of Geophysical Research: Atmospheres* (2020JD033758). *Note publication of this article is delayed due to an unexpected backlog with the AGU production vendor.*
- †Jin Q., Crippa P. and Pryor S.C. (2020): Spatial characteristics and temporal evolution of the relationship between $PM_{2.5}$ and aerosol optical depth over the eastern USA during 2003-2017. *Atmospheric Environment* **239** 117718 doi: 10.1016/j.atmosenv.2020.117718.
- †Jin Q. and Pryor S.C. (2020): Long-term trends of extreme aerosol optical depth events and radiation in North America using multiple satellite retrievals, CERES and MERRA2. *Journal of Geophysical Research: Atmospheres* **125** e2019JD031137 https://doi.org/10.1029/2019JD031137.
- Crippa P., #Sullivan R.C., Thota A., and Pryor S.C. (2019): Sensitivity of simulated aerosol properties over eastern North America to WRF-Chem parameterizations. *Journal of Geophysical Research: Atmospheres* **124** 3365-3383.
- #Sullivan R.C., Crippa P., Matsui H., Leung, L.R., Zhao C., Thota A., and Pryor S.C. (2018): New particle formation leads to cloud dimming. *npj Climate and Atmospheric Science* 1:9 doi:10.1038/s41612-018-0019-7
- #Sullivan R.C., Levy R., da Silva A. and Pryor S.C. (2017): Developing and diagnosing climate change indicators of regional aerosol optical properties. *Nature: Scientific Reports* **7** art # 18093, doi: 10.1038/s41598-017-18402-x

- Crippa P., Castruccio S. and Pryor S.C. (2017a): Forecasting ultrafine particle concentrations from satellite and in situ observations. *Journal of Geophysical Research: Atmospheres* 122 1828-1837.
- Crippa P., #Sullivan R.C., Thota A., and Pryor S.C. (2017b): The impact of resolution on meteorological, chemical and aerosol properties in regional simulations with WRF-Chem. *Atmospheric Chemistry and Physics* **17** 1511-1528.
- #Sullivan R.C., Crippa P., Hallar A.G., Larisse L., Whitburn S., Van Damme M., Leaitch W.R., Walker J., Khlystov A., and Pryor S.C. (2016): Using satellite-based measurements to explore spatiotemporal scales and variability of drivers of new particle formation. *Journal of Geophysical Research: Atmospheres* **121** 12217–12235.

Additional journal publication – research conducted in part for proof of principle for the proposal but published prior start of award:

- Crippa P., #Sullivan R.C., Thota A., and Pryor S.C. (2016): Evaluating the skill of high resolution WRF-Chem simulations in describing drivers of aerosol direct climate forcing on the regional scale. *Atmospheric Chemistry and Physics* **16** 397-416.

7.2 Other publications

- Pryor S.C., Sullivan R.C., Bernstein D.N., Thota A., and Crippa P. (2017): Detection and attribution of trends in aerosol populations and extreme aerosol events over North America. 3rd PEEX Science Conference, Moscow, Russia, September 2017. *6 page printed paper published in Report Series in Aerosol Science, University of Helsinki, p403-409 in Proceedings of the 3rd Pan-Eurasian Experiment (PEEX) Conference and the 7th PEEX Meeting, Editors: Hanna K. Lappalainen, Päivi Haapanala, Alla Borisova, Sergey Chalov, Nikolay Kasimov, Sergej Zilitinkevich, and Markku Kulmala.*

7.3 Conference presentations

- Guo Y., Crippa P., Thota A. and Pryor S.C. (2021): Sensitivity of extreme aerosol events to anthropogenic emission changes. *101st American Meteorological Society Annual Meeting (13th Symposium of Aerosol-Cloud-Climate Interactions)*, Virtual Meeting, January 2021.
- Sullivan R.C., Crippa P., Matsui H., Leung R., Zhao C., Thota A., and Pryor S.C. (2018): Modeling the impact of new particle formation on regional cloud radiative forcing. *98th American Meteorological Society Annual Meeting (10th Symposium on Aerosol-Cloud-Climate Interactions),* Austin, TX, January 2018.
- Bernstein D., Sullivan R.C., Thota A., Crippa P. and Pryor S.C. (2017): Diagnosing causes of extreme aerosol optical depth events. *AGU Annual conference*, New Orleans, LA, December 2017.
- Pryor S.C., Sullivan R.C., Bernstein D.N., Thota A., and Crippa P. (2017): Detection and attribution of trends in aerosol populations and extreme aerosol events over North America. *3rd PEEX Science Conference*, Moscow, Russia, September 2017.
- Sullivan R.C., Levy R., Da Silva A., Pryor S.C. (2017): Developing and diagnosing climate change indictors of regional aerosol optical properties. *EGU General Assembly 2017*, Vienna, Austria, April 2017.

7.4 Website

http://www.geo.cornell.edu/eas/PeoplePlaces/Faculty/spryor/NASA_NCA_project/index.html