

**Final Summary of Research
University Of Wisconsin-Madison**

(To be completed by RSP or the Department)

Project Title: **Activities with the Real-time Air Quality Modeling System (RAQMS) for Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) Data Analysis**

Award Number: **NNX09AE20G**

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For the Period of: **3/1/2009 through 2/28/2012**

Principal Investigator: **Todd Schaack**

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SSEC Number (Internal) **3930
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(To be completed by the Principal Investigator)

Inventions Report:

- No Inventions resulted from this award
 Yes

Inventory Report:

- No federally owned equipment is in the custody of the PI
 Yes

Publications: (Please list)

Dupont, R., B. Pierce, J. Worden, J. Hair, M. Fenn, P. Hamer, M. Natarajan, T. Schaack, A. Lenzen, E. Apel, J. Dibb, G. Diskin, G. Huey, A. Weinheimer, and D. Knapp, 2010: Reconstructing ozone chemistry from Asian wild fires using models, satellite and aircraft measurements during the ARCTAS campaign. Atmos. Chem. Phys. Discuss., 10, 26751–26812, 2010, www.atmos-chem-phys-discuss.net/10/26751/2010/doi:10.5194/acpd-10-26751-2010.

Natarajan, Murali; Pierce, R. Bradley; Schaack, Todd K.; Lenzen, Allen J.; Al-Saadi, Jassim A.; Soja, Amber J.; Charlock, Thomas P.; Rose, Fred G.; Winker, David M.; Worden, John R.: Radiative forcing due to enhancements in tropospheric ozone and carbonaceous aerosols caused by Asian fires during spring 2008 J. Geophys. Res., Vol. 117, No. D6, D06307
<http://dx.doi.org/10.1029/2011JD016584> 27 March 2012.

Summary of Technical Effort: (Usually several paragraphs. Please feel free to attach additional pages if you wish.)

Background

This project focused on utilization of the Real-time Air Quality Modeling System (RAQMS) (Pierce et al., 2003, 2007) at the University of Wisconsin-Madison Space Science and Engineering Center (SSEC) in support of post mission Arctic Research of the Composition of the Troposphere from Aircraft and Satellites (ARCTAS) science studies conducted in collaboration with NASA Langley Research Center and NASA Jet Propulsion Laboratory scientists. This research was aimed at understanding the production and loss mechanisms of Arctic ozone using ozone and carbon monoxide observations from the Tropospheric Emission Spectrometer (TES) in conjunction with RAQMS chemical and aerosol analyses studies and Lagrangian photochemical analysis tools. The scientific objectives of the research focused on the integration of satellite, ground based, and airborne observations through chemical data assimilation techniques and model/measurement synthesis studies to evaluate the impacts of long-range transport of anthropogenic pollution, emissions from boreal forest fires, and stratosphere-troposphere exchange on the chemical and aerosol composition of the Arctic. RAQMS chemical and aerosol analyses contributed to these studies by providing estimates of

chemical and aerosol transformation processes along trajectories initialized from regions where MODIS data indicates fires. Wildfire trajectories that intersect TES orbits were used to estimate the influence of these fires on ozone and CO observed by TES.

Accomplishments:

1. RAQMS post-mission ozone and aerosol re-analyses were completed.

RAQMS post-mission ozone re-analyses included assimilation of stratospheric profiles from the Microwave Limb Sounder (MLS) and cloud-cleared total ozone column measurements from the Ozone Monitoring Instrument (OMI) onboard Aura. The ozone re-analyses included improved representation of background error growth estimates which impacts upper troposphere/lower stratosphere ozone. Post-mission aerosol re-analyses included assimilation of aerosol optical depth (AOD) retrievals from the Moderate Resolution Imaging Spectroradiometer (MODIS) instruments onboard Terra and Aqua. Errors in the MODIS observation operator associated with the use of fixed geometric thicknesses in the conversion of aerosol mass to optical depth were corrected.

2. The fidelity of the RAQMS re-analyses was evaluated based on comparisons with CALIPSO and DC8 observations during Spring ARCTAS campaign.

Comparisons between CALIPSO and RAQMS aerosol extinction and aerosol optical depths were conducted in collaboration with CALIPSO scientists at the NASA Langley Research Center. Results of these studies showed that the RAQMS aerosol re-analyses tended to overestimate AOD relative to cloud free CALIPSO observations with biases (CALIPSO-RAQMS) of -0.11.

3. RAQMS wildfire ensemble trajectory analyses were conducted and used to identify TES sampling of Thailand, Kazakhstan, and Baykal wildfire plumes during April 2008.

Daily ensemble wild fire trajectory simulations were conducted which sampled the RAQMS chemical analyses to characterize the photochemical evolution of wildfire plumes during April and June 2008. The resulting Lagrangian photochemical analyses were used to identify the origins of high aerosol backscatter observed by the DIAL instrument during the April 19, 2008 DC-8 transit flight. The results showed that the majority of the aerosol backscatter observed by DIAL on the April 19, 2008 DC-8 flight originated from wildfires in Thailand and Kazakhstan. However, the Kazakhstan wildfire plume was advected over the Baykal wildfires during transit and included emissions from the Baykal wildfires.

4) Ozone (O₃) and carbon monoxide (CO) satellite measurements from the TES, simulations from the RAQMS and aircraft data from the NASA DC8 aircraft were used to characterize the chemical and dynamical evolution of Asian wildfire plumes during the spring ARCTAS campaign 2008 (Dupont et al., 2010).

A series of RAQMS simulations (without aerosol or ozone assimilation) were conducted where April 2008 wildfire emissions were restricted to within either the Kazakhstan, Siberian, or Thailand regions. Differences between the baseline RAQMS simulation and simulations with the restricted wildfire emissions were then used to infer which fires were primarily responsible for the aerosol and ozone enhancements observed by the DC8. Comparisons between satellite O₃ and CO measurements and RAQMS model results show consistency when the TES averaging kernel and constraint vector are applied to the model. However, RAQMS CO simulations suggest that TES observations do not capture the full range of CO variability in the plume due to low sensitivity. The analysis shows that the Kazakhstan plume is responsible for increases of O₃ and CO mixing ratios up to 6.4 ppbv and 38 ppbv in the lower troposphere, and the Thailand plume is responsible for increases of O₃ and CO mixing ratios up to 11 ppbv and 71 ppbv in the upper troposphere.

5) The radiative forcing due to enhancements in tropospheric ozone and carbonaceous aerosols was estimated

We collaborated with Murali Natarajan (NASA/LARC) using an off-line radiative transfer model along with the RAQMS chemical and aerosol analyses to calculate the direct radiative forcing due to the Asian wildfires that occurred in Thailand, Kazakhstan, and Siberia regions during spring 2008 (Natarajan et al., 2012). Results from these simulations, averaged over April 2008, indicate that tropospheric ozone column increased by more than 10 Dobson units (DU) near the Thailand region, and by lesser amounts in the other regions due to the fires. An off-line radiative transfer model was used to evaluate the direct radiative forcing due to the fire-induced changes in atmospheric composition. For clear sky, the monthly averaged radiative forcing at the top of the atmosphere (TOA) was mostly negative with peak values less than 12 W/m² occurring near the fire regions. At high latitudes, the radiative forcing was positive due to the presence of absorbing aerosols over regions of high surface albedo. Regions of positive forcing at TOA were more pronounced under total sky conditions. The monthly averaged radiative forcing at the surface was mostly negative, and peak values of less than 30 W/m² occurred near the fire regions. Persistently large negative forcing at the surface could alter the surface energy budget and potentially weaken the hydrological cycle.

6) RAQMS ARCTAS chemical analyses were used as part of a multi-scale modeling study examining the influence of background ozone concentrations on Southern California air quality during the ARCTAS-CARB period (Huang, M et al., 2010).

References:

Dupont, R., B. Pierce, J. Worden, J. Hair, M. Fenn, P. Hamer, M. Natarajan, T. Schaack, A. Lenzen, E. Apel, J. Dibb, G. Diskin, G. Huey, A. Weinheimer, and D. Knapp, 2010: Reconstructing ozone chemistry from Asian wild fires using models, satellite and aircraft measurements during the ARCTAS campaign, *Atmos. Chem. Phys. Discuss.*, 10, 26751–26812, 2010 doi:10.5194/acpd-10-26751-2010.

Huang, M, G. R. Carmichael, B. Adhikary, S. N. Spak, S. Kulkarni, Y. Cheng, C. Wei, Y. Tang, D. D. Parrish, S. J. Oltmans, A. D'Allura, A. Kaduwela, C. Cai, A. J. Weinheimer, M. Wong, R. B. Pierce, J. A. Al-Saadi, D. G. Streets, Q. Zhang, "Impacts of transported background Ozone on California air quality during 1 the ARCTAS-CARB period --A multi-scale modeling study", *Atmos. Chem. Phys.*, 10, 6947-6968, 2010

Natarajan, Murali; Pierce, R. Bradley; Schaack, Todd K.; Lenzen, Allen J.; Al-Saadi, Jassim A.; Soja, Amber J.; Charlock, Thomas P.; Rose, Fred G.; Winker, David M.; Worden, John R.: Radiative forcing due to enhancements in tropospheric ozone and carbonaceous aerosols caused by Asian fires during spring 2008 *J. Geophys. Res.*, Vol. 117, No. D6, D06307
<http://dx.doi.org/10.1029/2011JD016584> 27 March 2012.

Pierce, R. B., et al. 2003: Regional Air Quality Modeling System (RAQMS) predictions of the tropospheric ozone budget over east Asia. *J. Geophys. Res.*, 108(D21), 8825, doi:10.1029/2002JD003176.

Pierce, R. B., et al. 2007: Chemical data assimilation estimates of continental U.S. ozone and nitrogen budgets during the Intercontinental Chemical Transport Experiment–North America. *J. Geophys. Res.*, 112, D12S21, doi:10.1029/2006JD007722.