Supplementary Materials

A Comparative Study of Two-way and Offline Coupled WRF v3.4 and CMAQ v5.0.2 over the Contiguous U.S.: Performance Evaluation and Impacts of Chemistry-Meteorology Feedbacks on Air Quality

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Table S1. The model configurations of the two-way WRF-CMAQ simulation.

Attributes	Model Configurations
Model	WRF v3.4-CMAQ v5.0.2
Simulation period	2008-2012
Domain	CONUS
Horizontal grid spacing	36-km (148 × 112 grid cells)
Vertical grid	34 layers from surface to 100 hPa
Physical options	
Shortwave radiation	Rapid and accurate Radiative Transfer Model for GCM (RRTMG)
Longwave radiation	RRTMG
PBL	ACM2
Land surface	Pleim-Xiu
Microphysics	Morrison two-moment
Cumulus	Kain-Fritsch
Aerosol activation	Abdul-Razzak and Ghan
Chemical options	
Gas-phase chemistry	CB05 with updated chlorine chemistry
Aerosol module	AERO6
Photolysis	CMAQ inline
Aqueous-phase chemistry	AQ chemistry module (AQCHEM)
Meteorological and chemical IC and BC	Downscaled from the modified Community Earth System Model/Community Atmosphere Model (CESM/CAM5) v1.2.2; Meteorological ICs/BCs bias-corrected with National Center for Environmental Protection's Final (FNL) Operational Global Analysis data
Anthropogenic emission	NEI 2008 updated to 2010, and NEI 2011
Biogenic emission	BEIS3
Dust emission	CMAQ inline
Sea-salt emission	CMAQ inline

Table S2 summarizes the observational databases and the variables evaluated in this work. For evaluation of chemical concentrations and meteorological variables, the surface networks include the National Climatic Data Center (NCDC) Quality Controlled Local Climatological Data (QCLCD), Clean Air Status and Trends Network (CASTNET), the Aerometric Information Retrieval System (AIRS) – Air Quality System (AQS), the Interagency Monitoring of Protected Visual Environments (IMPROVE), the Chemical Speciation Network (CSN), the Southeastern Aerosol Research and Characterization (SEARCH), and the National Atmospheric Deposition Network (NADP). Several aerosol-cloud-radiation variables are also evaluated against satellite retrievals including the Clouds and the Earth's Radiant Energy System (CERES) and the Moderate Resolution Imaging Spectroradiometer (MODIS).

NCDC QCLCD data contains data over 700 U.S. locations from July 1996 to December 2004, and over 1600 locations from 2005 onwards (http://www.ncdc.noaa.gov/data-access/land-based-station-data/land-based-datasets/quality-controlled-local-climatological-data-qclcd). CASTNET observations have been collected in a range of rural environments, from desert to agricultural locations, and from flat to complex terrains (http://java.epa.gov/castnet/epa_jsp/sites.jsp). It contains measurement data for meteorological variables and chemical concentrations. AIRS-AQS is the U.S. EPA's repository for ambient air quality data from over 5000 active monitors (http://www.epa.gov/ttn/airs/airsaqs/). While IMPROVE observations have been collected in protected visual environments, i.e., in National Parks and Wilderness Areas (http://vista.cira.colostate.edu/improve/), CSN sites are located in a range of locations from urban to rural areas (http://www.epa.gov/ttnamti1/specgen.html). Both networks contain data for $PM_{2.5}$ and major $PM_{2.5}$ species. NADP contains precipitation data from rain gauges.

The MODIS satellite retrievals for AOD (Remer et al., 2005), CF, COT, and CWP come from the level 3 MODIS gridded atmosphere monthly global joint product (MOD08_M3) collected from the Terra platform (http://modis-atmos.gsfc.nasa.gov/MOD08_M3/). The CDNC data used in this study are derived from MODIS by Bennartz (2007).

Table S2. Observational datasets and variables evaluated in this study.

Gases and PM Species			
Observational database	**Variables evaluated**	**Sampling Frequency**	**Number of Sites**
CASTNET	Max 1-hr and 8-hr O_3	Daily for O_3	~90
AIRS–AQS	O_3	Hourly	~1150
IMPROVE	$PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, EC, OC	24-hour data. Data availability once every 3 days	~160
CSN	$PM_{2.5}$, SO_4^{2-}, NO_3^-, NH_4^+, EC, TC	24-hour data. Data availability once every 3 days	~200
Meteorology			
Observational Database	*Variables evaluated*	*Temporal Resolution*	*Spatial Resolution*
NCDC QCLCD	T2, RH, WS10,WD10	Hourly	~700 before 2005 ~1600 after 2005

NADP	Precipitation	Weekly	255
Radiation and other Aerosol/Cloud variables			
Observational Database/ Satellite	*Variables evaluated*	*Temporal Resolution*	*Number of sites/ Spatial Resolution*
CERES	SWDOWN	Monthly	$1º \times 1º$
MODIS	AOD, CF, COT, CWP, QVAPOR, CCN	Monthly	$1º \times 1º$
MODIS derived based on Bennartz (2007)	CDNC	Monthly	$1º \times 1º$
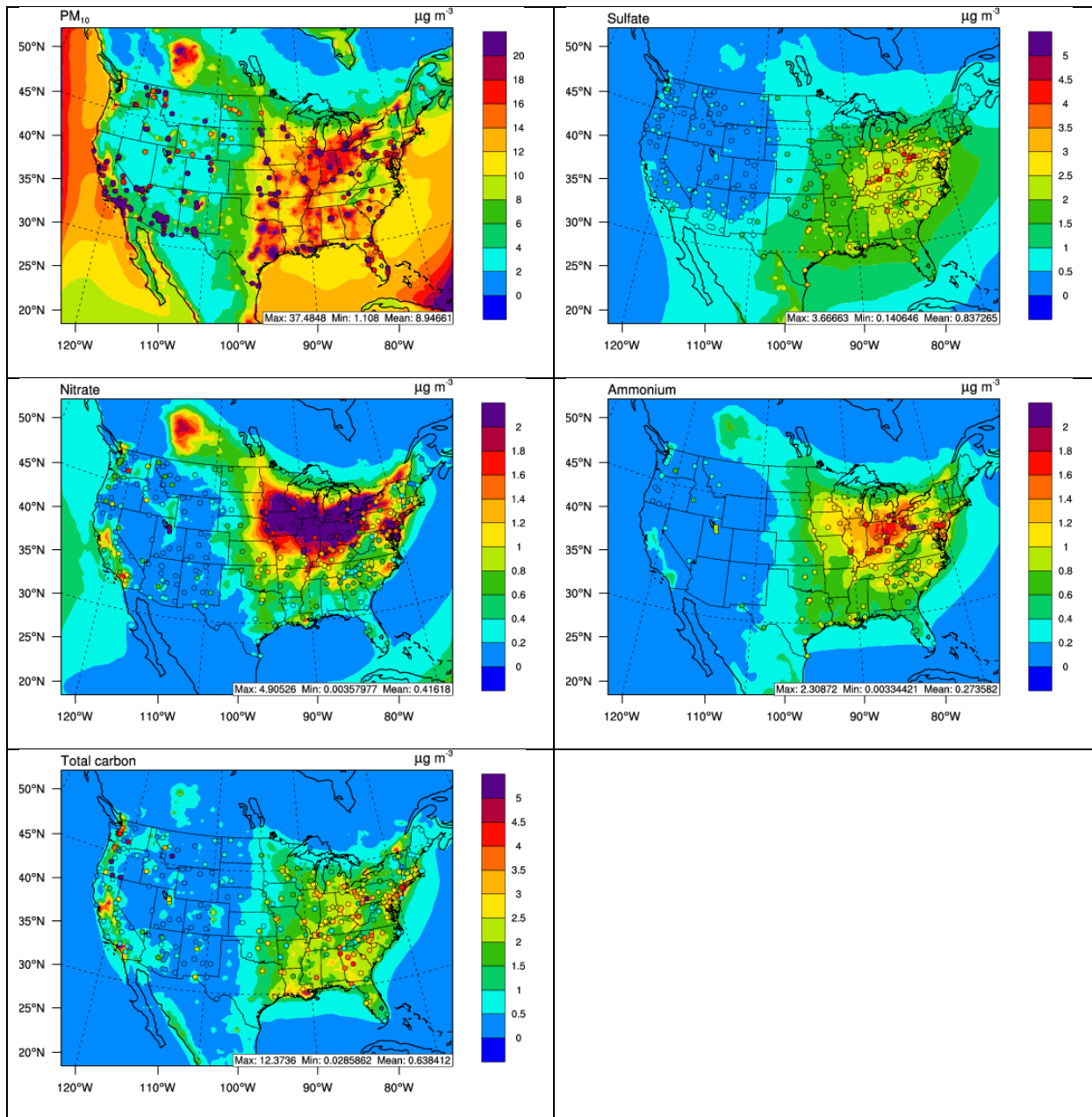
Figure S1. Spatial distributions of 5-year averaged daily PM$_{10}$ and PM$_{2.5}$ constituents overlaid with observations.