



# Supplement of

# Improvement and further development in CESM/CAM5: gas-phase chemistry and inorganic aerosol treatments

J. He and Y. Zhang

Correspondence to: Y. Zhang (yang\_zhang@ncsu.edu)

#### 1. Datasets for model evaluation

A number of observational datasets from surface networks and satellites are used for model evaluation. They are summarized along with the variables to be evaluated in Table S1.

#### 2. The Student's t-Test

To determine if the changes in model predictions due to changes in model configurations are statistically significant, the student's t-test analysis is performed between six pairs of 2001 simulations with different model configurations. Table S2 summarizes those results. The results show that the changes in most cloud/radiative variables due to changes in model configurations are statistically significant.

### 3. Performance Statistics for JJA from the 2001-2005 Simulations

As shown in Table S3, MAM\_NEW\_5YA improves performance of radiative variables such as LWD, SWD, and OLR with reduced absolute values of NMB, NME, and RMSE during June, July, and August (JJA) of 2001-2005, although it slightly degrades the performance of SWCF. MAM\_NEW\_5YA also improves cloud variables such as CF, COT, CWP, CCN5, and CDNC, with absolute reduction of NMBs of 0.9-18.0%. As shown in Table S4, with all new and modified treatments in MAM\_NEW\_5YA, SO<sub>4</sub><sup>2-</sup>, BC, OC, TC, and PM<sub>25</sub> are improved over CONUS, SO<sub>4</sub><sup>2-</sup>, PM<sub>25</sub>, and PM<sub>10</sub> are improved over Europe, and SO<sub>2</sub> and PM<sub>10</sub> are improved over East Asia, and TOR is improved over globe. Compared with full 5-year (2001-2005) average (ANU), JJA gives better model predictions for radiation (e.g., LWD, SWD, and OLR) and cloud (e.g., COT, CWP, column CCN5, and CDNC).

## 4. Impacts of Gas-Aerosol Partitioning

The chemical regimes is the controlling factor for gas-aerosol equilibrium partitioning, which is determined based on the ratio of  $SO_4^{2-}$  molar concentrations to total molar concentrations of cations and their respective gases (referred to as TCAT/TSO4) (Zhang et al., 2000). Three regimes are defined based on the values of TCAT/TSO4: (1) if TCAT/TSO4 < 2, the system contains excess sulfate and is in a sulfate-rich regime; (2) if TCAT/TSO4 = 2, the system contains just sufficient sulfate to neutralize the cation species and is in sulfate-neutral regime; (3) if TCAT/TSO4 > 2, the system contain insufficient sulfate to neutralize the cation species and is in sulfate-poor regime. Over land, the major cation is  $NH_4^+$ , and there are also crustal species ( $K^+$ ,  $Ca^{2+}$ , and  $Mg^{2+}$ ) associated with dust emissions, whereas over ocean, the major cation is Na<sup>+</sup>, which is a non-volatile species. Therefore, the gas-aerosol equilibrium partitioning behaves differently over land and over ocean. Figure S1 shows the absolute differences of H<sub>2</sub>SO<sub>4</sub>, fine particulate sulfate (SO4f), NH<sub>3</sub>, fine particulate ammonium (NH4f), HNO<sub>3</sub>, fine particulate nitrate (NO3f), HCl, and fine particulate chloride (CLf) for winter (December, January, and February (DJF)) 2001 between MAM\_CON and MAM\_CON/ISO. Figure S2 shows the distributions of TCAT/TSO4 in MAM CON and MAM CON/ISO, and their absolute differences for summer and winter, 2001. In summer, as shown in Figure 6 in the paper, compared to MAM\_CON, TCAT/TSO4 in MAM\_CON/ISO either increases up by 80.1 (mostly over ocean) or decreases up by 51.8 (over both land and ocean), leading to a net increase of 0.7. In MAM\_CON, most regions are in sulfate-poor regime, whereas Greenland, southeast U.S., North Africa, a small portion of Asia and North Atlantic Ocean, and some areas in North Pole are in sulfate-rich regime in summer. However, due to the simplified thermodynamics treatment in MAM\_CON, NH<sub>3</sub> is underpredicted and NH<sub>4</sub><sup>+</sup> is overpredicted (see Table 3 in the paper). With the inclusion of ISORROPIA II, most sulfate-poor regions over land and over part

of Pacific Ocean and most Atlantic Ocean become less sulfate-poor. The sulfate-poor regime can drive HNO<sub>3</sub>/HCl to produce NO<sub>3</sub><sup>-</sup>/Cl<sup>-</sup> by neutralizing excess NH<sub>4</sub><sup>+</sup>. If the amount of NO<sub>3</sub><sup>-</sup>/Cl<sup>-</sup> is insufficient to neutralize  $NH_4^+$ , sulfate-poor regime can drive  $NH_4^+$  to the gas phase to produce NH<sub>3</sub>. Therefore, the increase of NH<sub>3</sub> and decrease of NH<sub>4</sub><sup>+</sup> in MAM CON/ISO are mainly due to insufficient  $NO_3^{-7}/Cl^{-1}$  to neutralize  $NH_4^{+1}$  under sulfate-poor regime. Insufficient  $NO_3^{-7}/Cl^{-1}$  results from the thermodynamic partitioning under higher temperature conditions that favors the production of HNO<sub>3</sub> and HCl from NO<sub>3</sub><sup>-</sup>/Cl<sup>-</sup> to produce HNO<sub>3</sub> and HCl under higher temperature conditions. The slight increase of NO<sub>3</sub><sup>-</sup> over Pacific Ocean and South Atlantic Ocean is due to much higher  $Na^+$  concentrations yet insufficient  $SO_4^{2-}$  in those regions compared with those over the land areas. Unlike a sulfate-poor regime, a sulfate-rich regime (e.g., small portion of North Atlantic Ocean, South China Sea, and Greenland), requires more cations such as NH<sub>4</sub><sup>+</sup> and Na<sup>+</sup> to neutralize excess  $SO_4^{2^2}$  in the system and the thermodynamics favors the partitioning of volatile species such as NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup> in the gas phase as HNO<sub>3</sub> and HCl. Therefore, despite the increased temperatures, the decrease of  $NH_4^+$  due to its evaporation back to the gas-phase is not as significant as that of  $NO_3^-$  and  $Cl^-$ , because  $NH_4^+$  needs to stay in the system to neutralize SO<sub>4</sub><sup>2-</sup>. In winter, as shown in Figure S1, compared with MAM\_CON, the mixing ratios of H<sub>2</sub>SO<sub>4</sub> in MAM\_CON/ISO either increase by up to 4.3 ppt, or decrease by up to 1.0 ppt, leading to a net increase with the global mean of 0.001 ppt. NH<sub>3</sub> increases over most regions except Europe, eastern China, and some regions in North Pole. HNO3 decreases over most oceanic areas, Northeastern China, and East Europe, whereas increases over South Asia, North Pole, southern U.S., Africa, and most land areas in southern hemisphere. HCl increases over most areas except the northeastern portion of Asia and eastern Europe.

Compared with MAM\_CON, MAM\_CON/ISO predicts higher HNO<sub>3</sub> and HCl over some land areas. As shown in Figure S2, in MAM\_CON, most regions are in sulfate-poor regime, whereas Greenland, North Pole, North Africa, some portions of Asia and western Pacific Ocean are in sulfate-rich regime. For example, northeastern China is in sulfate-poor regime, driving HNO<sub>3</sub> and HCl partitioning to the aerosol phase to neutralize excess NH<sub>4</sub><sup>+</sup>. This results in an increase in NO3f and Clf, changing sulfate-poor regime to less sulfate-poor. North Pacific Ocean and southern oceanic areas are also in sulfate-poor regime, and the increase of NO3f is due to the partitioning HNO<sub>3</sub> to the aerosol phase to neutralize Na<sup>+</sup>, whose concentration is relatively higher compared to that over land areas. Therefore, more anions such as NO<sub>3</sub><sup>-</sup> are needed to neutralize the system. However, the decrease Cl<sup>-</sup> over these regions is due to the equilibrium state of HCl under different atmospheric conditions. The western Pacific Ocean is in sulfate-rich regime, driving NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup> partition to the gas phase, which results in a decrease in NO3f and Clf, and an increase in HNO<sub>3</sub> and HCl over this region. With the inclusion of ISORROPIA II, the western Pacific Ocean changes from sulfate-rich regime to less sulfate-rich regime.

Figure S3 shows the absolute differences of major inorganic gas and aerosol species between metastable (MAM\_NEWA) and stable (MAM\_NEWB) conditions. Compared with MAM\_NEWA, the global average changes predicted by MAM\_NEWB are overall small (within 5%) for most gaseous and aerosol species. For example, the global average changes are 0.01  $\mu$ g m<sup>-3</sup> (by 4.2%) for SO<sub>4</sub><sup>2-</sup>, 0.005  $\mu$ g m<sup>-3</sup>(by 12.8%) for NH<sub>4</sub><sup>+</sup>, 0.006  $\mu$ g m<sup>-3</sup>(by -0.01%) for NO<sub>3</sub><sup>-</sup>, and -4×10<sup>-4</sup>  $\mu$ g m<sup>-3</sup> (by 2.0%) for Cl<sup>-</sup>. The increase of SO<sub>4</sub><sup>2-</sup> results in an increase in NH<sub>4</sub><sup>+</sup> (e.g., East Asia and Northeast U.S.). The differences between stable and metastable conditions may be more significant under low RH conditions (RH < 50% for nitrate, Fountoukis et al., 2009). However, based on the simulated global annual mean RH values, most regions have RH values > 60-70% (exceptions are over desert/arid regions such as Australia, the northern Africa, Arabian Desert, northwestern China, and western U.S.). These results indicate that the assumption of metastable conditions is not a significant sources of uncertainty for global model predictions of gaseous and aerosol species.

5. Impact of New and Modified Treatments on JJA Average Results from the 2001-2005 Simulations

Figure S4 shows the absolute differences of surface SO<sub>2</sub>, NH<sub>3</sub>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, TC, PM<sub>2.5</sub>, PM<sub>10</sub>, J, and PM<sub>num</sub> and Figure S5 shows the absolute differences of cloud and radiative variables between MAM\_SIM\_5Y and MAM\_NEW\_5YA for JJA average of 2001-2005. Compared with MAM\_SIM\_5Y, MAM\_NEW\_5YA predicts lower SO<sub>2</sub> and NH<sub>3</sub> over East Asia with higher  $SO_4^{2-}$  and  $NH_4^+$  in this region. More  $SO_2$  is oxidized to form  $SO_4^{2-}$ , leading to enhanced acidity, which drives more  $NH_3$  partitioning into  $NH_4^+$  to neutralize the system in this region. SO<sub>4</sub><sup>2-</sup> decreases over CONUS whereas NH<sub>4</sub><sup>+</sup> increases, driving more HNO<sub>3</sub> and HCl partitioning into  $NO_3^-$  and  $Cl^-$  to neutralize  $NH_4^+$ . Therefore, the concentrations of  $PM_{2.5}$  and  $PM_{10}$  increase over CONUS. The overprediction of  $NH_4^+$  over Europe and CONUS is mainly due to additional anions (NO<sub>3</sub><sup>-</sup> and Cl<sup>-</sup>) in the system, leading to perturbations in the thermodynamic equilibrium. Similar to Figure 7 in the paper, J and  $PM_{num}$  increase near the surface, resulting in an increase in AOD and cloud variable predictions such as column CCN5, CDNC, and COT (see Figure S5). As shown in Figure S5, SWD decreases by 3.2 W m<sup>-2</sup> in global mean, which is due to the increased cloud predictions (e.g., column CCN5, CDNC, and COT). Due to aerosol direct and indirect effects, SWCF increases by 2.6 W m<sup>-2</sup> in global mean. Compared with ANU, the absolute change of most radiative variables are smaller in JJA. The

absolute changes of  $PM_{10}$  are larger in ANU than in JJA, which is mainly due to the dust events during other months (e.g., March-May over East Asia).

Species/Variables	Dataset		
Downwelling longwave radiation (LWD)	BSRN		
Downwelling shortwave radiation (SWD)	BSRN		
Outgoing longwave radiation (OLR)	NOAA/CDC		
Cloud fraction (CF)	MODIS		
Cloud optical thickness (COT)	MODIS		
Cloud water path (CWP)	MODIS		
Precipitating water vapor (PWV)	MODIS		
Aerosol optical depth (AOD)	MODIS		
Column cloud condensation nuclei (ocean) at $S = 0.5\%$ (CCN5)	MODIS		
Cloud droplet number concentration (CDNC)	BE07		
Shortwave cloud radiative forcing (SWCF)	CERES		
Carbon monoxide (CO)	East Asia: NIES of Japan, TAQMN		
	CONUS: CASTNET		
Ozone $(O_3)$	Europe: Airbase, BDQA, EMEP		
	East Asia: TAQMN		
	CONUS: CASTNET		
	Europe: Airbase, BDQA, EMEP		
Sulfur dioxide (SO <sub>2</sub> )	East Asia: MEP of China, NIES of Japan,		
	TAQMN		
	CONUS: CASTNET		
bNitric acid (HNO <sub>3</sub> )	Europe: EMEP		
Ammonia (NH <sub>3</sub> )	Europe: Airbase, EMEP		
Nitrogen disvide (NO.)	Europe: Airbase, BDQA, EMEP		
Nitrogen dioxide (NO <sub>2</sub> )	East Asia: NIES of Japan, TAQMN		
Sulfate $(SO_4^{2^-})$	CONUS: CASTNET, IMPROVE, STN		
	Europe: Airbase, EMEP		
Ammonium $(NH_4^+)$	CONUS: CASTNET, IMPROVE, STN		
Annomum (N11 <sub>4</sub> )	Europe: Airbase, EMEP		
Nitrate (NO <sub>3</sub> <sup>-</sup> )	CONUS: CASTNET, IMPROVE, STN		
$(100_3)$	Europe: Airbase, EMEP		
Chloride (Cl <sup>-</sup> )	CONUS: IMPROVE		
	Europe: Airbase, EMEP		
Organic carbon (OC), Black carbon (BC),	CONUS: IMPROVE, STN		
Total carbon (TC)	CONUS: INFROVE, STN		
Particulate matter with diameter less than 2.5 $\mu$ m	CONUS: IMPROVE, STN		
(PM <sub>2.5</sub> )	Europe: BDQA, EMEP		
Particulate matter with diameter less than 10 µm	Europe: Airbase, BDQA, EMEP		
$(PM_{10})$	East Asia: MEP of China, NIES of Japan,		
	TAQMN		
Column CO	Globe: MOPITT		
Column NO <sub>2</sub>	Globe: GOME		
Tropospheric ozone residual (TOR)	Globe: TOMS/SBUV		
New particle formation rate (J)	Globe: Kulmala et al. (2004); Yu et al. (2008)		

Table S1. Datasets for model evaluation

BSRN: Baseline Surface Radiation Network; NOAA/CDC: National Oceanic and Atmospheric Administration Climate Diagnostics Center; MODIS: Moderate Resolution Imaging Spectroradiometer; BE07: Bennartz, 2007; CERES: Clouds and Earth's Radiant Energy System; TOMS/SBUV: the Total Ozone Mapping Spectrometer/the Solar Backscatter UltraViolet; MOPITT: the Measurements Of Pollution In The Troposphere; GOME: Global Ozone Monitoring Experiment; CASTNET: Clean Air Status and Trends Network; IMPROVE: Interagency Monitoring of Protected Visual Environments; STN: Speciation Trends Network; EMEP: European Monitoring and Evaluation Program; BDQA: Base de Données sur la Qualité de l'Air; AirBase: European air quality database; MEP of China: Ministry of Environmental Protection of China; TAQMN: Taiwan Air Quality Monitoring Network; NIES of Japan: National Institute for Environmental Studies of Japan.

			<b>D' 1'</b>						
		Paired-simulation							
Species/Variables	MAM_SIM/	MAM_CB05_GE/	MAM_CON/	MAM_CON/	MAM_SIM/	MAM_NEWA/			
	MAM_CB05_GE	MAM_CON	MAM_CON/IMN	MAM_CON/ISO	MAM_NEWA	MAM_NEW/EMIS			
LWD	0.7	0.6	0.2	0.8	0.5	0.7			
SWD	8.1×10 <sup>-3</sup>	0	0.03	5.1×10 <sup>-12</sup>	1.3×10 <sup>-12</sup>	0.3			
OLR	4×10 <sup>-4</sup>	0	0.9	1.6×10 <sup>-10</sup>	4.6×10 <sup>-13</sup>	0.8			
SWCF	2×10 <sup>-4</sup>	$1.2 \times 10^{-12}$	0.4	0	5.7×10 <sup>-12</sup>	0.2			
CF	8.7×10 <sup>-5</sup>	$1.2 \times 10^{-12}$	0.05	5.3×10 <sup>-12</sup>	0	0.4			
СОТ	2.3×10 <sup>-3</sup>	0	3.9×10 <sup>-3</sup>	$1.5 \times 10^{-12}$	2.3×10 <sup>-12</sup>	0.3			
CWP	3.7×10 <sup>-3</sup>	0	0.06	5.4×10 <sup>-12</sup>	6.8×10 <sup>-13</sup>	0.2			
PWV	0.4	0.08	0.5	0.1	0.2	0.9			
AOD	7.9×10 <sup>-5</sup>	0	3.1×10 <sup>-6</sup>	3.8×10 <sup>-13</sup>	1.8×10 <sup>-11</sup>	0.03			
Column CCN5 (ocean)	5.3×10 <sup>-12</sup>	0	3.3×10 <sup>-12</sup>	0	0	0			
CDNC	2.7×10 <sup>-10</sup>	2.6×10 <sup>-12</sup>	9.8×10 <sup>-9</sup>	0	6.4×10 <sup>-13</sup>	0.5			

Table S2. Probability of differences in Radiative/Cloud Predictions between Paired-Simulation

\*Probability value is expressed in fraction, with a minimum value of 0 and a maximum value of 1. A value less than 0.05 (i.e., 5%) indicates that the differences between the simulation pairs are statistically significant at the 95% confidence level.

Species/Variables	Dataset	Obs	Simulations		
			MAM_SIM_5Y	MAM_NEW _5YA	MAM_NEW _5YB
LWD (W $m^{-2}$ )	CERES	320.3	317.3/-3.0/-0.9/3.0/13.0 <sup>a</sup>	318.0/-2.3/-0.7/3.0/12.8	318.1/-2.2/-0.7/2.9/12.6
SWD (W $m^{-2}$ )	CERES	192.4	197.9/5.5/2.9/9.7/25.1	194.1/1.7/0.9/9.5/24.3	196.2/3.8/2.0/9.5/23.6
OLR (W $m^{-2}$ )	NOAA-CDC	220.6	227.0/6.4/2.9/4.4/11.8	224.9/4.3/2.0/4.0/11.1	224.9/4.3/2.0/4.0/11.1
SWCF (W m <sup>-2</sup> )	CERES	-41.3	-40.1/1.2/-2.8/-25.9/16.4	-42.8/-1.5/3.6/-26.4/16.6	-41.2/0.1/-0.3/-26.3/16.2
CF (%)	MODIS	69.9	65.3/-4.6/-6.5/13.7/12.5	66.0/-3.9/-5.6/13.0/12.2	65.5/-4.4/-6.3/13.6/12.3
COT	MODIS	17.1	8.5/-8.6/-50.6/65.1/14.9	9.4/-7.7/-45.0/60.7/14.5	9.0/-8.1/-47.5/60.0/14.1
$CWP (g m^{-2})$	MODIS	87.9	41.7/-46.2/-52.6/53.3/54.5	47.2/-40.7/-46.3/47.4/50.7	46.6/-41.3/-47.0/47.8/51.4
PWV (cm)	MODIS	2.1	2.1/0.05/2.4/12.8/36.0	2.1/0.07/3.2/12.1/34.2	2.1/0.01/0.6/13.2/37.0
AOD	MODIS	0.2	0.2/-0.06/-34.1/54.5/0.2	0.2/-0.05/-29.0/51.3/0.2	0.2/-0.04/-25.0/48.7/0.2
Column CCN5 (ocean) (cm <sup>-2</sup> )	MODIS	2.3×10 <sup>8</sup>	6.1×10 <sup>7</sup> /-1.7×10 <sup>8</sup> / -74.2/74.7/2.6×10 <sup>8</sup>	$\begin{array}{c}9.1{\times}10^7{/}{}1.4{\times}10^8{/}\\59.8{/}61.9{/}2.3{\times}10^8\end{array}$	9.1×10 <sup>7</sup> /-1.4×10 <sup>8</sup> / -60.2/62.0/2.3×10 <sup>8</sup>
CDNC (cm <sup>-3</sup> )	BE07	117.4	48.5/-68.9/-58.8/61.1/87.7	69.5/-47.9/-40.8/49.7/76.2	67.7/-49.7/-42.3/50.8/77.1

Table S3. Statistical Performance of Radiative/Cloud Predictions during JJA, 2001-2005

<sup>a</sup> The values are expressed as Sim/MB/NMB/NME/RMSE, where Sim is modeled value; MB is mean bias; NMB is normalized mean bias (%); NME is normalized mean error (%); and RMSE is root mean square error.

	Simulations					
Variable <sup>a</sup>	Domain	Obs.	MAM_SIM_5Y	MAM_NEW _5YA	MAM_NEW _5YB	
СО	East Asia	535.4	-	119.4/-416.1/-77.7/77.7/441.1 <sup>b</sup>	114.8/-420.6/-78.6/78.6/444.7	
SO <sub>2</sub>	CONUS	2.0	8.3/6.3/309.8/312.1/9.6	8.2/6.2/309.0/311.4/9.6	8.0/6.0/297.5/299.9/9.3	
	Europe	5.3	6.0/0.7/12.4/85.2/7.1	6.0/0.7/13.3/87.3/7.3	6.3/1.0/19.2/90.1/7.5	
	East Asia	3.7	2.3/-1.4/-37.2/57.6/2.5	3.2/-0.5/-12.6/59.2/3.5	3.2/-0.5/-14.9/58.0/3.6	
NH <sub>3</sub>	Europe	6.0	2.7/-3.2/-54.4/84.0/18.0	2.2/-3.8/-62.9/82.6/18.0	2.4/-3.5/-59.1/82.7/18.0	
NO <sub>2</sub>	Europe	18.5	-	4.2/-14.3/-77.4/78.2/17.8	4.4/-14.1/-76.4/77.3/17.6	
	East Asia	13.2	-	2.1/-11.1/-84.2/84.2/12.1	2.1/-11.1-84.5/84.5/12.1	
03	CONUS	39.4	-	47.4/8.0/20.2/23.9/11.7	46.9/7.5/19.2/22.9/11.2	
	Europe	64.9	-	93.8/28.9/44.5/44.6/32.0	96.2/31.3/48.2/48.3/34.4	
	East Asia	21.7	-	34.2/12.5/57.5/57.5/13.7	33.9/12.2/56.4/56.4/13.3	
HNO <sub>3</sub>	CONUS	1.6	-	1.8/0.2/10.5/40.9/0.9	1.6/0.04/2.5/38.7/0.8	
	Europe	1.0		1.8/0.8/87.4/118.0/1.3	1.9/0.9/94.8/125.8/1.4	
SO4 <sup>2-</sup>	CONUS	3.6	3.2/-0.5/-12.4/22.1/1.1	3.2/-0.4/-10.3/19.9/1.0	3.1/-0.5/-12.7/19.1/0.9	
	Europe	2.4	2.9/0.5/20.5/46.7/1.6	2.8/0.4/17.4/42.2/1.5	2.9/0.5/22.0/44.7/1.5	
$\mathbf{NH_4}^+$	CONUS	1.3	1.3/-0.02/-1.3/20.2/0.4	1.4/0.1/9.8/31.8/0.6	1.4/0.1/11.0/28.1/0.5	
	Europe	0.8	1.0/0.2/30.3/46.8/0.6	1.6/0.8/95.0/99.7/1.1	1.6/0.8/96.0/100.6/1.1	
	CONUS	0.5	-	0.7/0.2/37.8/84.5/0.6	0.8/0.3/57.1/97.9/0.7	
NO <sub>3</sub> <sup>-</sup>	Europe	1.3	-	1.5/0.3/22.0/55.1/1.0	1.5/0.3/26.5/52.2/1.0	
Cľ	CONUS	0.1	-	0.1/-4.0×10 <sup>-3</sup> /-5.3/98.5/0.2	0.1/-4.0×10 <sup>-3</sup> /-5.4/101.2/0.2	
CI	Europe	0.1		1.1/1.0/650.0/650.0/1.4	1.1/1.0/666.8/666.8/1.4	
BC	CONUS	0.4	0.4/-0.02/-5.0/42.3/0.2	0.4/-0.02/-4.8/42.3/0.2	0.4/-0.02/-5.7/41.3/0.2	
OC	CONUS	1.7	1.1/-0.6/-34.8/50.5/1.1	1.5/-0.2/-13.6/50.6/1.1	1.4/-0.3/-15.6/45.9/1.0	
ТС	CONUS	3.5	1.5/-1.9/-55.8/60.0/2.5	2.1/-1.4/-40.6/49.7/2.2	2.0/-1.5/-42.8/50.3/2.2	
PM <sub>2.5</sub>	CONUS	10.6	8.4/-2.2/-20.5/35.5/4.9	10.0/-0.6/-5.6/27.0/4.1	9.9/-0.7/-6.3/19.4/2.6	
	Europe	13.1	8.7/-4.4/-33.6/34.8/5.4	10.1/-3.0/-23.0/30.4/5.0	11.9/-1.1/8.7/26.1/4.3	
PM <sub>10</sub>	Europe	25.9	16.4/-9.5/-36.6/41.4/12.5	18.3/-7.6/-29.4/34.5/11.7	21.6/-4.3/-16.7/26.6/9.7	
	East Asia	85.4	31.5/-54.0/-63.2/65.0/59.0	39.9/-45.6/-53.3/54.6/51.2	45.4/-40.0/-46.8/51.3/48.4	
Col.CO	Globe	1.3×10 <sup>18</sup>	-	1.2×10 <sup>18</sup> /-1.1×10 <sup>17</sup> /- 8.2/26.8/4.4×10 <sup>17</sup>	1.2×10 <sup>18</sup> /-1.3×10 <sup>17</sup> / -10.0/28.1/4.7×10 <sup>17</sup>	
Col.NO <sub>2</sub>	Globe	5.9×10 <sup>14</sup>	-	$\begin{array}{c}9.0{\times}10^{14}{/}3.1{\times}10^{14}{/}\\52.2{/}65.0{/}5.2{\times}10^{14}\end{array}$	$\frac{8.8{\times}10^{14}/2.9{\times}10^{14}}{48.9/62.4/5.1{\times}10^{14}}$	
TOR	Globe	32.1	29.8/-2.3/-7.4/18.9/6.9	31.1/-1.0/-3.0/14.2/5.9	30.4/-1.7/-5.4/15.4/6.2	

Table S4. Statistical Performance of Chemical Predictions during JJA, 2001-2005

<sup>a</sup>The units are CO, ppm (over East Asia); SO<sub>2</sub>, ppb (over East Asia) and  $\mu$ g m<sup>-3</sup> (over CONUS); O<sub>3</sub>, ppb (over CONUS) and  $\mu$ g m<sup>-3</sup> (over Europe); column CO and NO<sub>2</sub>, molecules cm<sup>-2</sup>; TOR, DU. All other concentrations are in  $\mu$ g m<sup>-3</sup>. <sup>b</sup>The values are expressed as Sim/MB/NME/RMSE, where Sim is modeled value, MB is mean bias; NMB is

normalized mean bias (%); NME is normalized mean error (%); RMSE is root mean squared error.

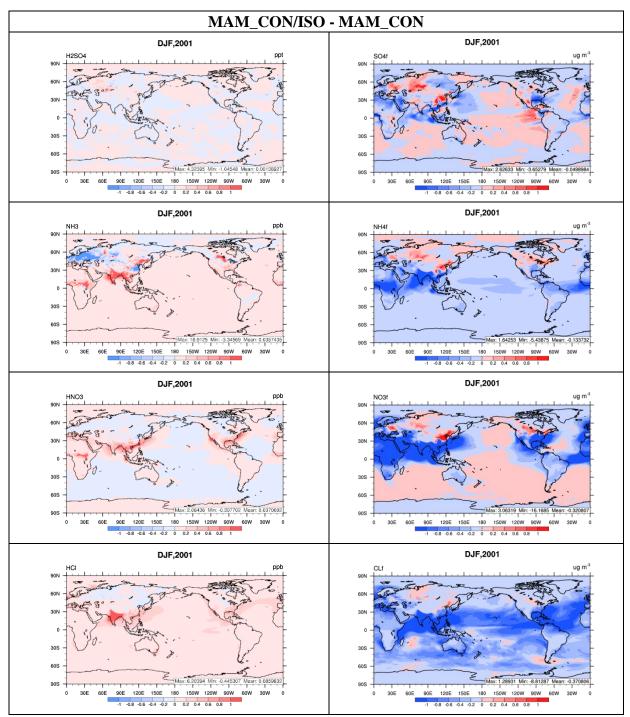


Figure S1. Absolute differences of major PM species and their gas precursors between MAM\_CON/ISO and MAM\_CON for winter, 2001.

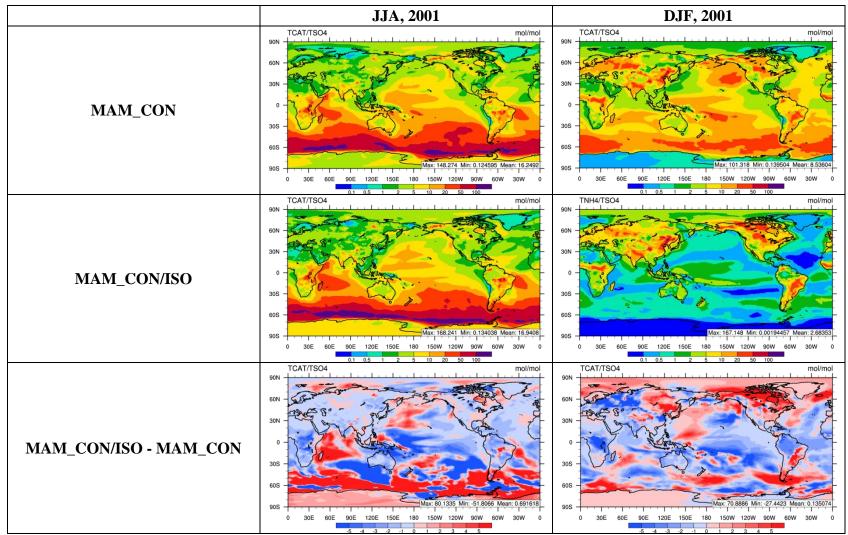


Figure S2. Surface distribution of TCAT/TSO4 in MAM\_CON and MAM\_CON/ISO and absolute differences of TCAT/TSO4 between MAM\_CON/ISO and MAM\_CON for summer and winter, 2001

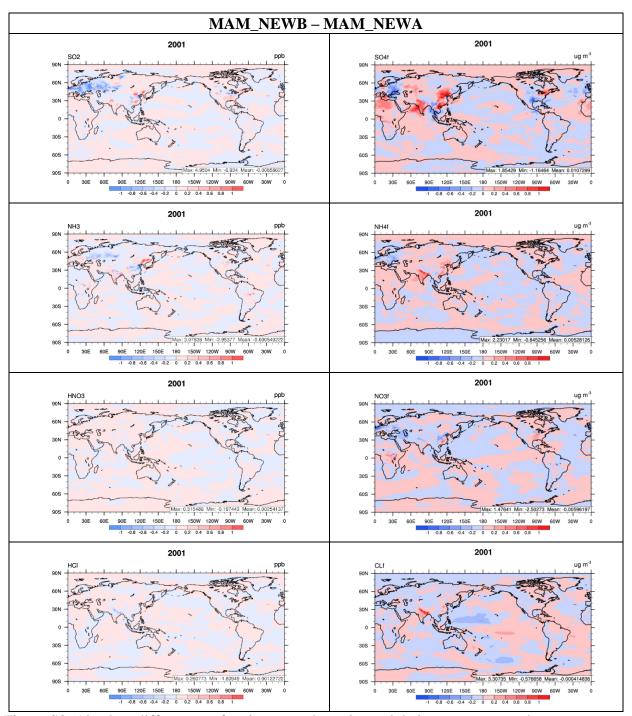


Figure S3. Absolute differences of major aerosol species and their gas precursors between metastable and stable conditions.

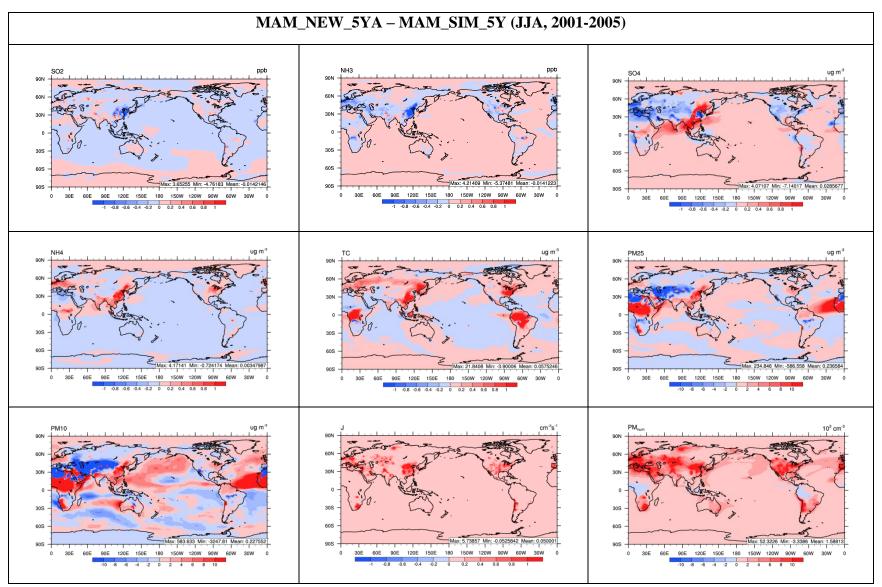


Figure S4. Absolute differences of major aerosol species and their gas precursors, new particle formation rate (J), and aerosol number between MAM\_NEW\_5YA and MAM\_SIM\_5Y for June, July, and August (JJA), 2001-2005.

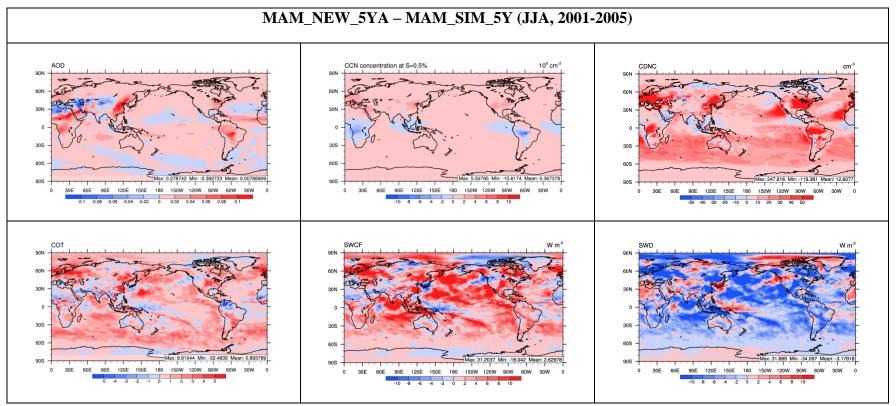


Figure S5. Absolute differences of major cloud and radiative variables between MAM\_NEW\_5YA and MAM\_SIM\_5Y for June, July, and August (JJA), 2001-2005.

References:

- Fountoukis, C., Nenes1, A., Sullivan, A., Weber, R., Farmer, D., and Cohen, R. C., 2009, Thermodynamic characterization of Mexico City aerosol during MILAGRO 2006, Atmos. Chem. Phys., 9, 2141-2156, 2009.
- Kulmala, M., Vehkamaki, H., Petaja, T., Dal Maso, M., Lauri, A., Kerminen, V.-M., Birmili, W., and McMurry, P.: Formation and growth rates of ultrafine atmospheric particles: A review of observations, J. Aerosol Sci., 35, 143-176, 2004.
- Yu, F., Wang, Z., Luo, G., and Turco, R. P.: Ion-mediated nucleation as an important global source of tropospheric aerosols, Atmos. Chem. Phys., 8, 2537-2554, 2008.
- Zhang, Y., Seigneur, C., Seinfeld, J. H., Jacobson, M., Clegg, S. L., Binkowski, F. S.: A comparative review of inorganic aerosol thermodynamic equilibrium modules: similarities, differences, and their likely causes, Atmos. Environ., 34, 117-137, 2000.