Executive editor

In my role as Executive editor of GMD, I would like to bring to your attention our Editorial version 1.1: http://www.geosci-model-dev.net/8/3487/2015/gmd-8-3487-2015.html.
This highlights some requirements of papers published in GMD, which is also available on the GMD website in the 'Manuscript Types' section:
http://www.geoscientific-model-development.net/submission/manuscript_types.html
In particular, please note that for your paper, the following requirement has not been met in the Discussions paper: "The main paper must give the model name and version number (or other unique identifier) in the title."
Please add the version numbers of WRF and CMAQ in the title upon your revised submission to GMD. Anyhow, the dash in WRF-CMAQ seem to be missing in the title, which is inconsitent with the rest of the article.

Response: The version numbers of WRF and CMAQ have been added to the paper title. We also changed "online coupled" to "two-way coupled" in the title for accuracy (Wong et al., 2012).

Anonymous Referee #1

This study concerns the application and evaluation of a regional climate-chemistry modeling system. This is certainly an interesting topic and represents an advancement in climate-chemistry modeling. The evaluation of the system for both climate model driven simulations and re-analysis driven simulations are reasonably thorough and successful.

Response: We thank Referee #1 for the positive comments. Please see below our point-by-point replies to other comments.

My primary criticism of this paper is the lack of detailed description of the GCM model, the downscaling, regional model configuration, and execution. Even though references are given for the CESM modeling and the chemical and aerosol processes are briefly described I would like to see further description of the CESM physics, spin-up, constraints, etc. I do not understand how this represents a climate scenario when it is for past years and is evaluated against observations. What does RCP4.5 for these years represent. Do these runs use observation based SSTs? If these runs were spun-up from pre-industrial times without any observed data constraints, there would be no reason to expect agreement with observations. If bias corrections are made to both the meteorology and chemistry, then how do these runs substantially differ from re-analysis driven runs? Please explain the rationale and expectations of these runs.

Response: The CESM-NCSU model development, application, and evaluation have been published in several journal papers (e.g., He and Zhang, 2014; He et al., 2015a, b; Gantt et al., 2014; Glotfelty et al., 2017a, b; Glotfelty and Zhang, 2017). Since CESM-NCSU has been well documented, it is a common practice for us to cite those references rather than repeat the CESM-NCSU model description in our paper. To address the reviewer's comment, we have added a brief description on the CESM-NCSU's configuration, initial conditions and the application mode in Section 2.2. We have also included a Table (Table S1) in the supplementary material to summarize the model configuration including physical schemes and chemical options used in CESM-NCSU applications under the RCP scenarios. More detailed descriptions can be found in He and Zhang (2014) and Glotfelty et al. (2017a, b).

The CESM-NCSU model has been applied for decadal global climate and air quality predictions to simulate the "current" climate (2001–2010) and the "future" climate (2046-2055) driven with the RCPs emissions for both the current and future decades (Glotfelty et al., 2017a and Glotfelty and Zhang, 2017). The CESM simulation for 2001–2010 is performed with fully-coupled CESM with CESM1.2.2 B_2000_STRATMAM7_CN configuration (rather than using prescribed SST), which represents a fully-coupled CESM configuration including prognostic simulation of the atmosphere, ocean, land, and sea ice from the various component models.

Global climate/chemistry models applied at a coarse spatial resolution may not well resolve mesoscale features over a regional domain of interest or well predict local air quality and thus are not suitable for high-resolution regional climate, air quality and health impact studies. Therefore, we have planned to downscale CESM runs with a regional model, which is the two-way coupled WRF-CMAQ for both a current period (2006-2010) and a future period (2046-2050) to study the impacts of projected changes in climate and anthropogenic emissions under the RCP4.5 scenario. In this paper, multi-year downscaling applications from CESM-NCSU simulations under RCP 4.5 were conducted to simulate regional climate and air quality in current year period (2006-2010) and evaluated against observations during this 5-yr period, which is the first part of the study. The results for future years will be presented in a future paper. The results from this Part I paper will establish a baseline for a future Part II paper. The WRF-CMAQ simulations are driven with CESM-NCSU downscaling data under RCP 4.5, and the projected emissions for 2046-2050 WRF-CMAQ simulations are based on MIX2008 and RCP4.5, so the downscaling simulations during 2006-2010 represent multi-year climatological baseline simulations under RCP4.5, and they will be further used to investigate future regional climate and air quality change in a future paper. To avoid confusion, we have revised the paper to clarify the above points in several places including abstract, Section 1, and conclusion.

This study presents the first application and evaluation of the two-way coupled WRF-CMAQ model for multi-year climatological simulations using the dynamical downscaling technique. Because GCMs generally suffer from systematic biases to a certain extent, bias correction to the GCM (i.e., CESM) initial and boundary conditions was applied in this study to improve the model performance in simulating regional climate. The bias-correction method corrected the mean climatological biases in temperature, water vapor, geopotential height, wind, and soil moisture variables using the NCEP reanalysis data following the approach of Xu and Yang (2012), and allowed the retention of the CESM-NUSU simulated climatic changes in the mean seasonal state, diurnal cycle, and variance of inter-annual variation. The bias-correction method used for the initial and boundary conditions derived from CESM-NCSU is described in Yahay et al. (2016). As described in Section 2.2, in this bias-correction approach, monthly climatological averages for ICs and BCs are first derived from both NCEP and CESM_NCSU cases. The differences between the ICs and BCs from the NCEP and CESM_NCSU climatological averages are then added onto the CESM_NCSU ICs and BCs to generate bias-corrected CESM_NCSU ICs/BCs. WRF-CMAQ simulations using bias-corrected meteorological ICs/BCs from CESM-NCSU are therefore different from the simulations using the NCEP reanalysis for meteorological ICs/BCs. The bias-correction method corrected the major biases in the meteorological variables that can cause serious issues for regional climate downscaling while retaining climate variability within the GCM for both current and future simulations. So we do not expect the climatological runs achieve the same performance as the re-analysis driven runs. Note that previous studies (Xu and Yang, 2012; Bruyère et al., 2014; Done et al., 2015) have shown that the improved dynamical downscaling method with GCM bias corrections greatly improves the downscaled climate. The bias-correction technique is also used in the NCAR CESM global bias-corrected CMIP5 output to support WRF/MPAS research (https://rda.ucar.edu/datasets/ds316.1/).

I'm also wondering about data assimilation in WRF. Our experience has been that long runs of WRF (one month or longer) need some sort of DA or frequent re-initialization. If not in this case, how were the meteorology statistics this good? Even downscaling from GCMs often use data assimilation from the GCM. Also, an important omission from the WRF physics description is the LSM.

Response: In order to simulate regional meteorology as accurately as possible and preserve the chemistry–meteorology feedbacks, re-initialization in WRF was used in the multi-year climatological application. The climatological simulations were reinitialized every 15 days in this work, which provides a compromise to allow the simulation of mesoscale features and aerosol feedbacks while periodically constraining the meteorological fields not significantly deviated from the GCM. Qian et al. (2003) found that frequent re-initialization with frequencies of 10 days to 1 month improved the accuracy in regional climate downscaling. Data assimilation in WRF was not used to allow chemistry–meteorology feedbacks within the system. We have clarified this in the Section 2.1 of the revised manuscript. The land surface model is the Pleim–Xiu land surface model. We have added them into Table 1.

Overall, I think that this study is worthy of reporting in GMD, especially the sensitivities of AQ and meteorology to aerosol direct radiative effects, and also the effects of dynamic BCs and biogenic and dust emissions. However, more explanation and description is needed particularly to help the reader understand the significance of the climate runs.

Response: We have added more explanation and description to help the readers understand the significance of the climate runs. Please refer to the above responses.

Specific comments:
P4lns21-22: This statement about "correcting the roughness length by increasing the friction velocity by 1.5 times when calculating wind speeds in the ACM2 PBL scheme to reduce the overpredictions of wind speeds" needs more explanation. First, if the roughness lengths need correcting why not change them and not the friction velocity. Second, what is the problem with roughness lengths? How are they specified and what are they? Our experience has not shown general overpredictions in windspeed. Windspeed and friction velocity are strongly affected by the LSM and surface layer scheme which are not even mentioned here. Also the LU scheme and data are important. The USGS 24cat data is way out of date especially for China where urbanization has been dramatic. Why not use MODIS LU?

Response: Large overpredictions in WS10 with NMBs of 48.7%-101.0% from WRF simulations have been reported in the literature (Penrod et al., 2014; Cai et al., 2016; Zhang et al., 2016a) because of unresolved subgrid-scale topographic features and uncertainties in parameterizations of turbulent fluxes in WRF (Hanna and Yang, 2001; Rontu, 2006; Mass and Ovens, 2011). The overpredictions in WS10 are likely caused by low surface drag due to the inappropriate representation of surface roughness because the detailed surface structure cannot be reproduced at a coarse grid resolution of 36-km. However, a rigorous surface roughness correction algorithm is not available in WRF v3.4 that is used in the two-way coupled WRF-CMAQ. To correct the WS10 bias, following Mass and Ovens (2010) and our previous studies (Zheng et al., 2015; Zhang et al., 2016b), a highly simplified indirect correction method is used in this study, namely, the surface drag is increased by 1.5 times (which is applied to the friction velocity) when calculating wind speeds in the ACM2 PBL scheme. The simple wind correction method effectively reduces the overpredictions of wind speeds. To address the reviewer's concern, we have indicated the highly simplified wind bias correction method as a limitation of this work in the conclusion section. A more rigorous method should be used for future work.
The USGS 24-category land use data is indeed way out of date for China where urbanization has been dramatic, which would also partly contribute to the overprediction in WS10. We have indicated this in the Section 3.1 of the revised manuscript.
We used Pleim–Xiu land surface model (PX-LSM, Xiu and Pleim, 2001) and Pleim–Xiu surface layer scheme. For best consistency between the WRF and CMAQ model, the PX-LSM and the ACM2 PBL scheme were used in the two-way coupled WRF-CMAQ model (https://www.airqualitymodeling.org/index.php/CMAQ_version_5.1_(November_2015_release)_Techn

ical_Documentation). We have added them into Table 1.

P5ln11-12: Why not use same vertical structure for WRF-CMAQ as CESM?

Response: The vertical coordinate in CESM is a hybrid sigma-pressure system, which is different from the WRF sigma coordinate. Thus we do not use the same vertical structure for WRF-CMAQ as CESM. We have clarified this in the Section 2.2 of the revised manuscript.

P5ln27: what is TOR?

Response: TOR represents tropospheric ozone residual. We have clarified this in the Section 2.2 of the revised manuscript.

Page 6: I don't understand what is the point of using RCP projections when modeling retrospectively. It seems that 2008 emission inventories are used for more detailed spatial-temporal allocation. Then why not just use these inventories? What is an RCP projection for past years? Please explain the logic here.

Response: RCP emissions are available for current and future decadal periods. The CESM-NCSU model has been recently applied for decadal global climate and air quality predictions to simulate the "current" climate scenario (2001–2010) and the "future" climate scenario (2046-2055) driven with the RCPs emissions for both current and future decades (Glotfelty et al., 2017a), therefore those "current" and "future" simulations represent multi-year climatological simulations under RCPs. The regional climatological simulations were driven with CESM-NCSU downscaling data under RCP 4.5. In order to achieve better performance for the regional WRF-CMAQ simulation, the MIX 2008 emission inventory is used for current years, and the emissions of some sectors that were not available from MIX 2008 were taken from RCP4.5. As we explained above, this paper only focuses on current year simulations which will be used as a baseline simulation for a future paper. We have clarified this in the Section 2.3 of the revised manuscript.

P8ln1-2: Should also report RMSE or MAE. Small biases don't tell whole story. Large over and under predictions could cancel out.

Response: As suggested, we have added the root mean square error (RMSE) in the statistics tables (Table 3, 4 and S3) in place of the normalized mean error (NME), and added the mean absolute gross error (MAGE) in Table 3. The model performed well for T2 and RH2, with MBs of -0.6 ℃ and 0.8%, correlation coefficients of 0.97 and 0.72, MAGEs of 2.4 ℃ and 9.7%, and RMSEs of 3.2 ℃ and 12.6%, respectively. WS10 was moderately overpredicted by 22.2%, with an MB of 0.6 m/s, an MAGE of 1.2 m/s and a RMSE of 1.6 m/s. Emery et al. (2001) suggested the benchmarks for satisfactory performance for T2 (MB within ±0.5 ℃, MAGE of ≤ 2.0 ℃) and WS10 (MB within ± 0.5 m/s, MAGE and RMSE of 2.0 m/s). In the climatological application, the MB and MAGE of T2 and the MB of WS10 are close to the benchmark, the MAGE and RMSE of WS10 are within the benchmark, and hence the performance is deemed acceptable. We have added this in the Section 3.1 of the revised manuscript.

P8ln15-16: saying that large errors could be attributable to KF and Morrison schemes is pretty *meaningless.*

Response: The convective precipitation dominated the overprediction of total precipitation in the southern oceanic area, which may be possibly due to overprediction of convective precipitation intensity by the Kain–Fritsch cumulus scheme. The non-convective precipitation dominated the overprediction of total precipitation in the northeastern oceanic area, which could be attributed to possible errors in the Morrison double-moment microphysics scheme. We have clarified this in the Section 3.1 of the revised manuscript.

P8ln28: Are the results shown in Fig5 averages for all 5 years?

Response: Yes, they are. We have clarified this in the Section 3.1 of the revised manuscript.

P9ln16: what are "upper BCs"? and where do they come from? And why are they particularly uncertain?

Response: Upper BCs represent upper layer boundary conditions (BCs) of O_3, which are derived from CESM. Because total column O_3 is mainly determined by O_3 concentrations in upper troposphere (Tang et al., 2009; Zhang et al., 2016a, c), the overpredictions of TOR (column O_3) can be largely attributed to the inappropriateness of the upper layer BCs of O_3. From the results of Tang et al. (2009), we could also find large uncertainties in upper layer BCs of O_3. We have clarified this in the Section 3.1 of the revised manuscript.

P9ln16-17: Another meaningless statement about uncertainties in about everything possibly causing errors in NO2 column. Can you provide more insightful analyses?

Response: Column NO_2 was moderately overpredicted by 18.3%. Potential uncertainties in NO_x emissions and the model treatment of deposition and chemistry processes may contribute to the model-observation difference. As discussed by Lin et al. (2010) and Han et al. (2015), there are several uncertainties in the modeled NO_x lifetime. Uncertainties in the NO_2 column retrievals from OMI (with a relative error of 25%, Boersma et al., 2011) and the averaging kernels (Han et al., 2015) could also help to explain the bias. We have added this in the Section 3.1 of the revised manuscript.

P9ln28-30: Please clarify this sentence.

Response: For the air quality application driven with NCEP-FNL data, the observation and simulation data pairs for surface meteorological variables against NCDC observational data were on an hourly basis. The high correlations for major meteorological variables in Table S3 indicated that the model showed good skills in hourly meteorological predictions, thus NCEP-FNL data were sufficient to support the air quality applications for hourly air quality predictions. We have clarified this in the Section 3.2 of the revised manuscript.

P10ln23-24: If a figure is important enough to be discussed in the text (S2) it should be in the main paper and not in the supplement. The reader should not need to see the supplement to follow the *discussion.*

Response: We think that Figure S2 is not so important to be discussed in the text. So we have removed Figure S2 and the corresponding discussion in the revised manuscript.

P11ln12-13: The names of the simulations are confusing. The "baseline" is NCEP_BASE_Imp but the sensitivity is NCEP_BASE which sounds more like it should be the base.

Response: The names of the simulations have been changed from CESM_BASE_Imp, CESM_BASE_Imp_Sens, NCEP_BASE_Imp and NCEP_BASE to CESM_BASE, CESM_BASE _Sens, NCEP_BASE and NCEP_BASE_WoImp.

P11ln25-26: How are the fixed BCs derived?

Response: The fixed BCs are provided by the operational CMAQ system. We have clarified this in the Sections 2.1 and 3.3 of the revised manuscript.

P12ln4: S4 should be in main paper.

Response: As suggested, we have moved Figure S4 to the main paper (i.e., Figure 11 in the revised manuscript).

P12ln15: "close" should be "closer"

Response: Revised as suggested.

P13ln11-12: Aerosol effects on photolysis in CMAQ do not depend on aerosol feedback in the WRF-CMAQ system. The more likely cause for ozone decline in the feedback run is increased NOx titration in cities due to reduced PBL mixing. Table 1: what LSM and surface layer scheme? Table 3 and 4: Better to have un-normalized error for T2, RH2, WS10, WD10

Response: The decrease in O_3 concentrations in the feedback run may be attributed to the increased NO_x titration resulted from increased atmospheric stability and reduced PBL height. We have clarified this in the Section 3.4 of the revised manuscript. Table 1: The land surface model is Pleim–Xiu land surface model (Xiu and Pleim, 2001). The surface layer scheme is Pleim–Xiu surface layer scheme. We have added them into Table 1. Table 3 and 4: We have added the mean absolute gross error (MAGE) in Table 3, and added the root mean square error (RMSE) in place of the normalized mean error (NME) in Tables 3 and 4.

Anonymous Referee #2

This study reports the evaluation against measurements of the output from a dynamical downscaling link between the global Community Earth System Model (CESM) and the WRF-CMAQ modelling system over the East Asia region for a number of meteorological and air quality composition variables. The climatological simulations were for RCP4.5 for 2006-10 and the air quality applications were for winter and summer months in 2013 (principal compositional variables of interest: PM2.5 and O3). The authors report satisfactory prediction of major meteorological variables, although see the first of the general comments below. The paper reports on a major piece of work, with what appear to be generally appropriate methods, and is within the scope for consideration of publication in GMD.

Response: We thank Referee #2 for the constructive comments. Please see below our point-by-point replies to other comments.

General comments

(1) The description of the downscaling (P5-6) indicates that aspects of it involves significant bias corrections, so to what extent is it valid to judge model performance by model-observation statistics? For example, it is stated on P8, lines 1-6, that the improved statistical performance of the modelling approach used in this study may be related to the bias-correction applied. If a bias correction is applied then presumably we expect better model-observation statistics, so have we learned anything fundamental about the model performance by these comparison statistics?

Response: While using bias-corrected ICs/BCs does improve WRF-CMAQ's model performance, it does not make model-observation comparison invalid. While meteorological reanalysis data were used to correct biases in meteorological ICs/BCs based on CESM-NCSU's results and satellite retrievals of O_3 were used to constrain their upper boundary conditions, observational data were used for model performance evaluation. Because GCMs generally suffer from systematic biases to a certain extent, bias correction to the GCM (i.e., CESM) boundary conditions was applied in this study to improve the model performance in simulating regional climate. By comparing to the traditional approach without GCM bias corrections, previous studies (Xu and Yang, 2012; Bruyère et al., 2014; Done et al., 2015) have shown that the improved dynamical downscaling method with GCM bias corrections greatly improves the downscaled climate. The bias-correction technique is also used in the NCAR CESM global bias-corrected CMIP5 output to support WRF/MPAS research (https://rda.ucar.edu/datasets/ds316.1/). Also note that the bias correction is applied to the ICs/BCs, rather than the model results. So, the model-observation comparison will provide insights into the model's capability in capturing observations.

(2) The model-observation statistics should include RMSE instead of, or in place of, the normalized mean error (NME). The former is the statistic usually used alongside the correlation coefficient and mean bias (or normalised mean bias) in the suite of statistics that captures the spectrum of model performance characteristics.

Response: As suggested, we have added the root mean square error (RMSE) in the statistics tables (Table 3, 4 and S3) in place of the normalized mean error (NME), and added the mean absolute gross error (MAGE) in Table 3. The model performed well for T2 and RH2, with MBs of -0.6 ℃ and 0.8%, correlation coefficients of 0.97 and 0.72, MAGEs of 2.4 ℃ and 9.7%, and RMSEs of 3.2 ℃ and 12.6%, respectively. WS10 was moderately overpredicted by 22.2%, with an MB of 0.6 m/s, an MAGE of 1.2 m/s and a RMSE of 1.6 m/s. We have added this in the Section 3.1 of the revised manuscript.

(3) In general, the discussion of model output against meteorological and compositional variations is (i) vague, i.e. non-quantitative (using phrasing like agreed well, satisfactory, etc.), and (ii) lacking explanatory insight, i.e. lists of potential reasons for discrepancy are given which could be written down as potential explanations without needing to do these comparisons. The authors should endeavour to provide more quantitative assessments of model performance, including how their mod-obs statistics compare with expectation and with other studies, and also to provide some more informed analysis of what is the driving explanation for mod-obs discrepancies for particular variables or circumstances.

Response: As suggested, we have provided more quantitative assessments of model performance in terms of MB, NMB, or RMSE in the abstract, result and conclusion sections of the revised manuscript.

The model biases or errors can be attributed to many factors. Pinpointing the exact causes is not a trivial effort, often involving large amounts of sensitivity simulations and in some cases, model further development and improvement that are not permitted with our very limited resources. Nevertheless, we have provided more insights into the model's performance statistics and how they are compared with other studies, wherever possible. For example, we have compared the performance of several meteorological variables with the benchmarks suggested by Emery et al. (2001) in the Section 3.1 of the manuscript. Emery et al. (2001) suggested the benchmarks for satisfactory performance for T2 (MB within ±0.5 ℃, MAGE of ≤ 2.0 ℃) and WS10 (MB within ± 0.5 m/s, MAGE and RMSE of 2.0 m/s). In the climatological application, the MB and MAGE of T2 and the MB of WS10 are close to the benchmark, the MAGE and RMSE of WS10 are within the benchmark, and hence the performance is deemed acceptable.

We have also compared the CMAQ performance of chemical predictions in this study with other studies, as shown in the Section 3.2 of the manuscript. The revised text is as follows:

The CMAQ performance of chemical predictions in this study was comparable to or even better than those of other air quality studies over East Asia (Wang et al., 2009; 2012; Liu et al., 2010; Zheng et al., 2015; Hu et al., 2016; Liu et al., 2016; Zhang et al., 2016a). This study predicted relatively well for most chemical species in most months. Compared with other regional modeling studies, WRF-CMAQv5.0.2 used in this study outperformed MM5/CMAQv4.6, which tend to underpredict the surface concentrations of major species with NMBs generally greater than -40% and overpredict surface O_3 concentrations in most months with NMBs generally higher than 20% over East Asia according to the evaluation results of Zhang et al. (2016a). A relatively good performance of CMAQv5.0.1 was also reported by Hu et al. (2016). Global models such as GEOS-Chem and CESM tend to underpredict $PM_{2.5}$ concentrations (by about -50% as reported by Jiang et al., 2013) and overpredict O_3 concentrations (by about 50% as reported by He and Zhang, 2014; Wang et al., 2013) in China/East Asia because of relatively coarse grid resolution and limitations in some model treatments (e.g., missing emissions of unspeciated primary $PM_{2.5}$, and discrepancies in surface layer height and vertical mixing).

Specific comments

P1, L27: The phrasing "The model showed good ability to predict PM2.5 . . ..and O3. . ." is non-quantitative and vague.

Response: The above sentence has been revised to include more quantitative assessment as follows:
The model showed good ability to predict $PM_{2.5}$ in winter (with a normalized mean bias (NMB) of 6.4% in 2013) and O_3 in summer (with an NMB of 18.2% in 2013) in terms of statistical performance and spatial distributions.
In addition, we have added this in the abstract of the revised manuscript.

P4, L20: Rephrase as "Several modifications in model. . ."

Response: Revised as suggested.

P7, L16: Although the acronym TOR is defined here, there needs to be some further explanation of what it means in practice, particularly in the context of its relevance to model performance evaluation.

Response: TOR represents tropospheric ozone residual or column abundance of O_3. We have clarified this in the Section 2.2 of the revised manuscript.

P12, L18: "were much closer to. . .

Response: Revised as suggested.

Reference

Boersma, K. F., Eskes, H. J., Dirksen, R. J., van der A, R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitão, J., Richter, A., Zhou, Y., and Brunner, D.: An improved tropospheric NO2 column retrieval algorithm for the Ozone Monitoring Instrument, Atmos. Meas. Tech., 4, 1905–1928, doi:10.5194/amt-4-1905-2011, 2011.

Bruyere, C. L., Done, J. M., Holland, G. J., and Fredrick, S.: Bias corrections of global models for regional climate simulations of high-impact weather, Clim. Dynam., 43, 1847–1856, doi:10.1007/s00382-013-2011-6, 2014.

Cai, C., Zhang, X., Wang, K., Zhang, Y., Wang, L., Zhang, Q., Duan, F., He, K., and Yu, S.: Incorporation of new particle formation and early growth treatments into WRF/Chem: Model improvement, evaluation, and impacts of anthropogenic aerosols over East Asia, Atmos. Environ., 124, 262–284, doi:10.1016/j.atmosenv.2015.05.046, 2016.

Done, J. M., Holland, G. J., Bruyere, C. L., Leung, L. R., and Suzuki-Parker, A.: Modeling high-impact weather and climate: lessons from a tropical cyclone perspective, Climatic Change, 129, 381–395, doi:10.1007/s10584-013-0954-6, 2015.

Emery, C., Tai, E., and Yarwood, G.: Enhanced meteorological modeling and performance evaluation for two texas episodes, Report to the Texas Natural Resources Conservation Commission, prepared by ENVIRON, International Corp., Novato, CA, available at: http://www.tceq.state.tx.us/assets/public/implementation/air/am/contracts/reports/mm/EnhancedMetModelingAndPerformanceEvaluation.pdf, 2001.

Gantt, B., He, J., Zhang, X., Zhang, Y., and Nenes, A.: Incorporation of advanced aerosol activation treatments into CESM/CAM5: model evaluation and impacts on aerosol indirect effects, Atmos. Chem. Phys., 14, 7485–7497, doi:10.5194/acp-14-7485-2014, 2014.

Glotfelty, T., He, J., and Zhang, Y.: The Impact of Future Climate Policy Scenarios on Air Quality and Aerosol/Cloud Interactions using an Advanced Version of CESM/CAM5: Part I. Model Evaluation for the Current Decadal Simulations, Atmos. Environ., 152, 222–239, doi:10.1016/j.atmosenv.2016.12.035, 2017a.

Glotfelty, T., He, J., and Zhang, Y.: Improving Organic Aerosol Treatments in CESM/CAM5: Development, Application, and Evaluation, Journal of Advances in Modeling Earth Systems, in review, 2017b.

Glotfelty, T., and Zhang, Y.: The Impact of Future Climate Policy Scenarios on Air Quality and Aerosol/Cloud Interactions using an Advanced Version of CESM/CAM5: Part II. Future Trend Analysis and Impacts of Projected Anthropogenic Emissions, Atmos. Environ., 152, 531-552, doi: 10.1016/j.atmosenv.2016.12.034, 2017.

Han, K. M., Lee, S., Chang, L. S., and Song, C. H.: A comparison study between CMAQ-simulated and OMI-retrieved NO2 columns over East Asia for evaluation of NOx emission fluxes of INTEX-B, CAPSS, and REAS inventories, Atmos. Chem. Phys., 15, 1913-1938, doi:10.5194/acp-15-1913-2015, 2015.

Hanna, S. R., and Yang, R. X.: Evaluations of mesoscale models' simulations of near-surface winds, temperature gradients, and mixing depths, J. Appl. Meteorol., 40, 1095–1104, doi:10.1175/1520-0450(2001)040<1095:EOMMSO>2.0.CO;2, 2001.

He, J., and Zhang, Y.: Improvement and further development in CESM/CAM5: gas-phase chemistry and inorganic aerosol treatments, Atmos. Chem. Phys., 14, 9171–9200, doi:10.5194/acp-14-9171-2014, 2014.

He, J., Zhang, Y., Glotfelty, T., He, R., Bennartz, R., Rausch, J., and Sartelet, K.: Decadal simulation and comprehensive evaluation of CESM/CAM5.1 with advanced chemistry, aerosol microphysics, and aerosol-cloud interactions, J. Adv. Model. Earth Sy., 7, 110–141, doi:10.1002/2014MS000360, 2015a.
He, J., Zhang, Y., Tilmes, S., Emmons, L., Lamarque, J. F., Glotfelty, T., Hodzic, A., and Vitt, F.: CESM/CAM5 improvement and application: comparison and evaluation of updated CB05_GE and MOZART-4 gas-phase mechanisms and associated impacts on global air quality and climate, Geosci. Model Dev., 8, 3999–4025, doi:10.5194/gmd-8-3999-2015, 2015b.
Hu, J., Chen, J., Ying, Q., and Zhang, H.: One-year simulation of ozone and particulate matter in China using WRF/CMAQ modeling system, Atmos. Chem. Phys., 16, 10333–10350, doi:10.5194/acp-16-10333-2016, 2016.
Jiang, H., Liao, H., Pye, H. O. T., Wu, S., Mickley, L. J., Seinfeld, J. H., and Zhang, X. Y.: Projected effect of 2000-2050 changes in climate and emissions on aerosol levels in China and associated transboundary transport, Atmos. Chem. Phys., 13, 7937–7960, doi:10.5194/acp-13-7937-2013, 2013.
Lin, J.-T., McElroy, M. B., and Boersma, K. F.: Constraint of anthropogenic NOx emissions in China from different sectors: a new methodology using multiple satellite retrievals, Atmos. Chem. Phys., 10, 63–78, doi:10.5194/acp-10-63-2010, 2010.
Liu, X., Zhang, Y., Cheng, S., Xing, J., Zhang, Q., Streets, D. G., Jang, C., Wang, W., and Hao, J.: Understanding of regional air pollution over China using CMAQ, part I performance evaluation and seasonal variation, Atmos. Environ., 44, 2415–2426, doi:10.1016/j.atmosenv.2010.03.035, 2010.
Liu, X., Zhang, Y., Zhang, Q., and He, M.: Application of online-coupled WRF/Chem-MADRID in East Asia: Model evaluation and climatic effects of anthropogenic aerosols, Atmos. Environ., 124, 321–336, doi:10.1016/j.atmosenv.2015.03.052, 2016.
Mass, C., and Ovens, D.: WRF model physics: progress, problems, and perhaps some solutions, in: The 11th WRF Users' Workshop, NCAR Center Green Campus, http://www.mmm.ucar.edu/wrf/users/workshops/WS2010/presentations/session%204/4-1_WRFworkshop2010Final.pdf (last access: June 2014), 21–25 June, 2010.
Mass, C., and Ovens, D.: Fixing WRF's High Speed Wind Bias: a New Subgrid Scale Drag Parameterization and the Role of Detailed Verification, Preprints, 24th Conference on Weather and Forecasting/20th Conference on Numerical Weather Prediction, American Meteorological Society, Seattle, W.A., 9B.6. Available online at: http://ams.confex.com/ams/91Annual/webprogram/Paper180011.html, 2011.
Penrod, A., Zhang, Y., Wang, K., Wu, S., and Leung, L. R.: Impacts of future climate and emission changes on US air quality, Atmos. Environ., 89, 533–547, doi:10.1016/j.atmosenv.2014.01.001, 2014.
Qian, J. H., Seth, A., and Zebiak, S.: Reinitialized versus continuous simulations for regional climate downscaling, Mon. Weather Rev., 131, 2857–2874, doi:10.1175/1520-0493(2003)131<2857:RVCSFR>2.0.CO;2, 2003.
Rontu, L.: A study on parametrization of orography-related momentum fluxes in a synoptic-scale NWP model, Tellus A, 58, 69–81, doi:10.1111/j.1600-0870.2006.00162.x, 2006.
Tang, Y., Lee, P., Tsidulko, M., Huang, H., McQueen, J. T., DiMego, G. J., Emmons, L. K., Pierce, R. B., Thompson, A. M., Lin, H., Kang, D., Tong, D., Yu, S., Mathur, R., Pleim, J. E., Otte, T. L., Pouliot, G., Young, J. O., Schere, K. L., Davidson, P. M., and Stajner, I.: The impact of chemical lateral boundary conditions on CMAQ predictions of tropospheric ozone over the continental United States, Environ. Fluid Mech., 9, 43–58, doi:10.1007/s10652-008-9092-5, 2009.
Wang, K., Zhang, Y., Jang, C., Phillips, S., and Wang, B.: Modeling intercontinental air pollution

transport over the trans-Pacific region in 2001 using the Community Multiscale Air Quality modeling system, J. Geophys. Res. Atmos., 114, D04307, doi:10.1029/2008JD010807, 2009.

Wang, K., Zhang, Y., Nenes, A., and Fountoukis, C.: Implementation of dust emission and chemistry into the Community Multiscale Air Quality modeling system and initial application to an Asian dust storm episode, Atmos. Chem. Phys., 12, 10209–10237, doi:10.5194/acp-12-10209-2012, 2012.

Wang, Y., Shen, L., Wu, S., Mickley, L., He, J., and Hao, J.: Sensitivity of surface ozone over China to 2000-2050 global changes of climate and emissions, Atmos. Environ., 75, 374–382, doi:10.1016/j.atmosenv.2013.04.045, 2013.

Wong, D. C., Pleim, J., Mathur, R., Binkowski, F., Otte, T., Gilliam, R., Pouliot, G., Xiu, A., Young, J. O., and Kang, D.: WRF-CMAQ two-way coupled system with aerosol feedback: software development and preliminary results, Geosci. Model Dev., 5, 299–312, doi:10.5194/gmd-5-299-2012, 2012.

Xiu, A., and Pleim, J. E.: Development of a land surface model. Part I: Application in a mesoscale meteorological model, J. Appl. Meteorol., 40, 192–209, doi:10.1175/1520-0450(2001)040<0192:doalsm>2.0.co;2, 2001.

Xu, Z., and Yang, Z.: An Improved Dynamical Downscaling Method with GCM Bias Corrections and Its Validation with 30 Years of Climate Simulations, J. Climate, 25, 6271–6286, doi:10.1175/JCLI-D-12-00005.1, 2012.

Yahya, K., Wang, K., Campbell, P., Glotfelty, T., He, J., and Zhang, Y.: Decadal evaluation of regional climate, air quality, and their interactions over the continental US and their interactions using WRF/Chem version 3.6.1, Geosci. Model Dev., 9, 671–695, doi:10.5194/gmd-9-671-2016, 2016.

Zhang, Y., Zhang, X., Wang, L., Zhang, Q., Duan, F., and He, K.: Application of WRF/Chem over East Asia: Part I. Model evaluation and intercomparison with MM5/CMAQ, Atmos. Environ., 124, 285–300, doi:10.1016/j.atmosenv.2015.07.022, 2016a.

Zhang, Y., Zhang, X., Wang, K., Zhang, Q., Duan, F., and He, K.: Application of WRF/Chem over East Asia: Part II. Model improvement and sensitivity simulations, Atmos. Environ., 124, 301–320, doi:10.1016/j.atmosenv.2015.07.023, 2016b.

Zhang, Y., Hong, C., Yahya, K., and Zhang, Q.: Multi-Year Application and Evaluation of WRF/Chem-MADRID for Real-Time Air Quality Forecasting over Southeastern United States, Atmos. Environ., 138, 162-182, doi:10.1016/j.atmosenv.2016.05.006, 2016c.

Zheng, B., Zhang, Q., Zhang, Y., He, K. B., Wang, K., Zheng, G. J., Duan, F. K., Ma, Y. L., and Kimoto, T.: Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China, Atmos. Chem. Phys., 15, 2031–2049, doi:10.5194/acp-15-2031-2015, 2015.

Multi-year Downscaling Application of Two-Way Coupled WRF3.4 and CMAQv5.0.2 over East Asia for Regional Climate and Air Quality Modeling: Model Evaluation and Aerosol Direct Effects

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Abstract. In this study, a regional coupled climate-chemistry modeling system using the dynamical downscaling technique was established by linking the global Community Earth System Model (CESM) and the regional two-way coupled Weather Research and Forecasting - Community Multiscale Air Quality (WRF-CMAQ) model for the purpose of comprehensive assessments of regional climate change and air quality and their interactions within one modeling framework. The modeling system was applied over East Asia for a multiyear climatological application during 2006-2010 driven with CESM downscaling data under Representative Concentration Pathway 4.5 (RCP 4.5) as well as a short-term air quality application in representative months in 2013 driven with a reanalysis dataset. A comprehensive model evaluation was conducted against observations from surface networks and satellite observations to assess the model's performance. This study presents the first application and evaluation of the two-way coupled WRF-CMAQ model for climatological simulations using the dynamical downscaling technique. The model was able to satisfactorily predict major meteorological variables. The improved statistical performance for the 2-m temperature (T2) in this study (with a mean bias of -0.6 ℃) compared with the Coupled Model Inter-comparison Project Phase 5 (CMIP5) multi-models might be related to the use of the regional model WRF and the bias-correction technique applied for CESM downscaling. The model showed good ability to predict $PM_{2.5}$ in winter (with a normalized mean bias (NMB) of 6.4% in 2013) and O_3 in summer (with an NMB of 18.2% in 2013) in terms of statistical performance and spatial distributions. Compared with global models that tend to underpredict $PM_{2.5}$ concentrations in China, WRF-CMAQ was able to capture the high $PM_{2.5}$ concentrations in urban areas. In general, the two-way coupled WRF-CMAQ model performed well for both climatological and air quality applications. The coupled modeling system with direct aerosol feedbacks predicted aerosol optical depth relatively well and significantly reduced the overprediction in downward shortwave radiation at the surface (SWDOWN) over polluted regions in China. The

performance of cloud variables was not as good as other meteorological variables, and underpredictions of cloud fraction resulted in overpredictions of SWDOWN and underpredictions of shortwave and longwave cloud forcing. The importance of climate-chemistry interactions was demonstrated via the impacts of aerosol direct effects on climate and air quality. The aerosol effects on climate and air quality in East Asia (e.g., SWDOWN and T2 decreased by 21.8 W m^{-2} and 0.45 ℃, respectively, and most pollutant concentrations increased by 4.8%~9.5% in January over China's major cities) were more significant than in other regions because of higher aerosol loadings that resulted from severe regional pollution, which indicates the need for applying online-coupled models over East Asia for regional climate and air quality modeling and to study the important climate-chemistry interactions. This work established a baseline for WRF-CMAQ simulations for a future period under the RCP4.5 climate scenario, which will be presented in a future paper.

1 Introduction

Climate change and air pollution are two critical environmental issues that humanity must face. There are complex interactions between air pollution and climate change (Fiore et al., 2012; von Schneidemesser et al., 2015; Fuzzi et al., 2015). Air pollutants (e.g., aerosols) have direct effects on radiative forcing by scattering or absorbing incoming radiation and also indirect effects via their role in cloud formation; the effects in turn affect climate systems. Climate change can affect meteorological fields (e.g., temperature, humidity, precipitation, wind speed, cloud cover, and boundary layer mixing) as well as natural emissions (e.g., biogenic volatile organic compounds (BVOCs) emissions, soil and lightning nitrogen oxides (NO_x) emissions, and dust emissions) and thereby affect air quality. Global climate and chemistry modeling simulations (Fiore et al., 2012; Stevenson et al., 2013; Kim et al., 2015) have been conducted to investigate global climate change and air quality under the Special Report on Emissions Scenarios (SRES) and the Representative Concentration Pathways (RCPs) scenarios developed for the Intergovernmental Panel on Climate Change (IPCC) Fourth Assessment Report (AR4) (IPCC, 2007) and Fifth Assessment Report (AR5) (IPCC, 2013). Global climate models used in the AR5 include more detailed representations of aerosol and cloud processes and their interactions than those used in the AR4. There is high confidence in the radiative forcing mechanisms due to aerosol–radiation interactions, although low confidence in the forcing mechanisms of aerosol–cloud interactions in the models remains (IPCC, 2013).

However, global climate/chemistry models applied at a coarse spatial resolution may not well resolve mesoscale features over a regional domain of interest or well predict local air quality and thus are not suitable for high-resolution regional climate, air quality and health impact studies. As a result of these deficiencies, the dynamical downscaling technique has been widely used in regional climate studies (Oh et al., 2014; Wang et al., 2015; Xu et al., 2015). Dynamical downscaling uses initial conditions (ICs) and boundary conditions (BCs) from global models to drive regional models for high-resolution simulations. Several regional air quality studies using dynamical downscaling approaches have been conducted to predict future air quality under a changing climate at a regional scale (Gao et al., 2013; Penrod et al., 2014; Sun et al., 2015). However, these studies tended to use offline models—chemical transport models (CTMs) driven by future climate archived from general circulation models (GCMs), lacking climate-chemistry interactions. Previous regional downscaling studies tended to focus on extreme climate events or the impacts of climate change on air quality rather than on the important chemistry and climate interactions.

Online-coupled regional climate-chemistry models have developed rapidly in recent years (Zhang et al., 2008; Baklanov et al., 2014). Recently, the online-coupled Weather Research Forecast (WRF) model with Chemistry (WRF/Chem) was evaluated for decadal application over the continental U.S. under RCP 4.5 and RCP 8.5 (Yahya et al., 2016, 2017a) and applied to decadal projections of future climate and air quality under both scenarios (Yahya et al., 2017b). The Community Multi-scale Air Quality (CMAQ) model has historically been an offline model developed by the U.S. Environmental Protection Agency (EPA) and widely used for air quality simulations over numerous countries and regions (Wang et al., 2009; 2012; Liu et al., 2010; Gao et al., 2013; Penrod et al., 2014; Sun et al., 2015; Zheng et al., 2015; Hu et al., 2016); it has recently been further developed to provide an online-coupled version with the Weather Research Forecast (WRF) model to simulate feedbacks between chemistry and meteorology (Wong et al., 2012). Several applications of the two-way coupled WRF-CMAQ model have been conducted to evaluate the performance of the coupled model system and to investigate aerosol direct effects (Wang et al., 2014; Gan et al., 2015; Hogrefe et al., 2015; Xing et al., 2016) and indirect effects (Yu et al., 2014) on climate and air quality. However, there is a lack of comprehensive evaluation of the two-way coupled WRF-CMAQ model over East Asia, where aerosol loadings are extremely high and have been found to have great impacts on regional climate and air quality (Wang et al., 2014; Liu et al., 2016). Additionally, the two-way coupled WRF-CMAQ model has not been applied and evaluated for multi-year climatological modeling.

In this study, following the work of Yahya et al. (2016, 2017a and b), a regional coupled climate-chemistry modeling system using the dynamical downscaling technique was established for the purpose of comprehensive assessments of regional climate change and air quality and their interactions within one modeling framework (see Figure 1). The two-way coupled WRF-CMAQ model, which takes into account the air quality and climate interactions, is driven by the Community Earth System Model (CESM) implemented with advanced chemistry and aerosol treatments by North Carolina State University (NCSU) (hereafter CESM-NCSU) (He and Zhang, 2014; He et al., 2015a, b; Gantt et al., 2014; Glotfelty et al., 2017a, b; Glotfelty and Zhang, 2017) for high-resolution regional simulation under a changing climate. Both meteorological dynamical downscaling and chemical composition downscaling from the CESM-NCSU were applied following the work of Yahya et al. (2016, 2017a and b). The dynamical downscaling methods fully take advantage of global climate-chemistry models that can well predict large-scale global changes and regional models that can better represent regional phenomena. The modeling system was applied over East Asia for a climatological application driven with CESM downscaling data for 5 years from 2006 to 2010 under RCP 4.5 as well as an air quality application in 2013 driven by the National Centers for Environmental Prediction (NCEP) Final Reanalyses (NCEP-FNL) dataset. Comprehensive model evaluation for meteorological, chemical and aerosol-cloud-radiation variables was conducted against surface observations and satellite observations to assess and improve the model's performance for regional climatological and air quality applications. This study presents the first application and evaluation of the two-way coupled WRF-CMAQ model for climatological-type simulations using the dynamical downscaling technique; it also demonstrates the importance of climate-chemistry interactions via the impacts of aerosol direct effects on climate and air quality. The main goals of this work are to evaluate the WRF-CMAQ's capability in reproducing the observations and to establish a baseline simulation during a current year period, which will be compared with a simulation during a future year period in order to assess the impacts of changes in climate and emissions on future air quality over East Asia in future work.

This paper is organized as follows. Section 2 describes the model configurations and simulation design, dynamical downscaling methods and evaluation protocols. Section 3 presents the results of comprehensive model evaluations for climatological and air quality applications, the improvements of model performance within the modeling system, and aerosol direct effects on regional climate and air quality. Section 4 summarizes the major conclusions and limitations of this study.

2 Model setup and evaluation protocol

2.1 Model configurations and simulation design

The two-way coupled WRF-CMAQ (WRF3.4 and CMAQv5.0.2) model is used for regional climate and air quality simulations. More details of the two-way coupled WRF-CMAQ are described by Wong et al. (2012). The current release of the WRF-CMAQ model supports the Rapid and accurate Radiative Transfer Model for General Circulation Models (RRTMG) radiation scheme for shortwave aerosol direct effects and uses a core-shell model to perform the aerosol optics calculation. It does not include aerosol indirect effects that result from interactions between aerosols and cloud microphysics. The detailed model configurations for the climatological application in this study are shown in Table 1. The WRF model configuration included the Morrison double-moment scheme (Morrison et al., 2009), version 2 of the Kain-Fritsch cumulus scheme (Kain, 2004), the Asymmetric Convective Model version 2 (ACM2) planetary boundary layer (PBL) scheme (Pleim, 2007), the Pleim–Xiu land surface model (Xiu and Pleim, 2001), and the RRTMG shortwave and longwave radiation scheme (Iacono et al., 2008). The CMAQ model was configured using the Carbon Bond 2005 (CB05) chemical mechanism (Yarwood et al., 2005; Whitten et al., 2010) and the sixth generation CMAQ aerosol module (AERO6) (Appel et al., 2013). The regional domain using a horizontal resolution of 36 km covered most of China and parts of East Asia. The two-way coupled WRF-CMAQ used the same vertical resolution for WRF and CMAQ, i.e., 23 sigma layers from the surface to 100 hPa.

Several modifications in model inputs and treatments were made in this study to improve the model performance. These included (1) correcting the surface roughness by increasing the friction velocity by 1.5 times when calculating wind speeds in the ACM2 PBL scheme to reduce the overpredictions of wind speeds (Mass and Ovens, 2010; Zheng, et al., 2015); (2) using the inline Biogenic Emissions Inventory System (BEIS3) model (Vukovich and Pierce, 2002; Schwede et al., 2005) over East Asia; (3) revising the default dust module developed by Tong et al. (2017) with updated friction velocity thresholds to generate more dust emissions following the work of Dong et al. (2015); (4) using bias-corrected chemical boundary conditions (BCs)/initial conditions (ICs) from CESM rather than using the fixed BCs/ICs provided by the operational CMAQ system.

Table 2 shows the four simulations conducted in this study. Climatological application (CESM_BASE) was driven by the climatological dataset (CESM-NCSU) over a 5-yr period (2006-2010) and aimed to assess the model performance on a climatological average timescale. Air quality application (NCEP_BASE) was driven by a reanalysis dataset (NCEP-FNL, NCEP Final Reanalysis) and aimed to assess the model performance for short-term air quality application. The air quality application was conducted for three representative months (January, April, and July) in 2013 because more surface air quality monitoring data were available for the evaluation of chemical predictions (refer to Section 2.4). NCEP_BASE_WoImp simulation without the improvements indicated above was designed for comparison to support the improvements made in NCEP_BASE. Sensitivity simulation without aerosol feedback (CESM_BASE_Sens) was designed to assess the aerosol direct effects on regional climate and air quality.

In order to simulate regional meteorology as accurately as possible and preserve the chemistry–meteorology feedbacks, re-initialization in WRF was used in the multi-year climatological application. The climatological simulations were reinitialized every 15 days in this work, which provides a compromise to allow the simulation of mesoscale features and aerosol feedbacks while periodically constraining the meteorological fields not significantly deviated from the GCM. Qian et al. (2003) found that frequent re-initialization with frequencies of 10 days to 1 month improved the accuracy in regional climate downscaling.

2.2 Dynamical downscaling from CESM-NCSU

The CESM-NCSU model with advanced chemistry and aerosol treatments has been applied for decadal global climate and air quality predictions to simulate the “current” climate scenario (2001–2010) and the “future” climate scenario (2046-2055) driven with the RCPs emissions (projected from base year 2000) (Glotfelty et al., 2017a and Glotfelty and Zhang, 2017). The CESM simulation for 2001–2010 is performed with fully coupled CESM with CESM1.2.2 B_2000_STRATMAM7_CN configuration, which represents a fully-coupled CESM configuration including prognostic simulation of the atmosphere, ocean, land, and sea ice from the various component models. The initial conditions for CAM5.1 are derived from a 10 year (1990–2000) CAM5.1 standalone simulation with the MOZART chemistry provided by NCAR. The initial conditions for ice and ocean models are from CESM default settings. The initial conditions for the land model are based on the output from the NCAR CESM/CAM4 B_1850–2000_CN simulation. Table S1 in the supplementary material summarizes the model configurations including physical schemes and chemical options used in CESM-NCSU simulations. More detailed descriptions can be found in He and Zhang (2014) and Glotfelty et al. (2017a, b). In this work, both meteorological dynamical downscaling and chemical composition downscaling from the CESM-NCSU were applied to provide meteorological and chemical ICs/BCs for regional WRF-CMAQ simulations.

Major processes for chemical composition downscaling included species mapping and horizontal and vertical interpolations. ICs were only needed for the first time step, whereas BCs were provided every 6 hours. The horizontal and vertical resolutions of CESM were 0.9 ° (latitude) × 1.25 ° (longitude) and 30 layers in hybrid sigma-pressure coordinates, respectively. Those of WRF-CMAQ were 36-km in Lambert projection coordinates and 23 layers in sigma coordinates, respectively. The horizontal interpolations to WRF-CMAQ grids were first applied by calculating distance weighted mean from four neighboring CESM grids, and the vertical interpolations to WRF-CMAQ layers were then applied by calculating pressure weighted mean from two nearest CESM layers. CESM/CAM5 and CMAQ both use the Carbon Bond 2005 (CB05) (Yarwood et al., 2005) chemical mechanism; therefore, most gas species can be directly mapped. CESM/CAM5 uses the 7-mode prognostic Modal Aerosol Model (MAM7) (Liu et al., 2012) with volatility-basis-set (VBS) (Glotfelty et al., 2017b), whereas CMAQ uses the 3-mode AERO6 aerosol module. The mapping table between CESM/CAM5 and CMAQ aerosol species is shown in Table S2. Secondary organic aerosol (SOA) species in CESM/CAM5 were divided according to different volatility levels. However, the CMAQ model includes specific SOA semi-volatile and nonvolatile species. The anthropogenic and biogenic SOA species in CESM/CAM5 were first lumped into total semi-volatile SOA and total nonvolatile SOA. The ratios among the SOA species derived from the default BCs/ICs were then used to allocate each SOA species in CMAQ based on the combined SOA, as suggested by Carlton et al. (2010). Bias-corrections were applied to the chemical ICs/BCs for species such as O_3 to reduce high biases against satellite retrievals (Zhang et al., 2016c). As indicated by Zhang et al. (2016c), using satellite-constrained boundary conditions for O_3 showed substantial improvement in model performance of tropospheric ozone residual (TOR, or column O_3). In this study, the boundary conditions for O_3 were constrained with satellite observations following the similar work of Zhang et al. (2016c). Scale factors of 0.8 to 0.95 were applied to adjust the original O_3 boundary conditions derived from CESM-NCSU. CESM-NCSU tends to overpredict natural dust emissions over East Asia, and modelled dust concentrations from CESM-NCSU were thus divided by 3 to reduce the high biases in dust simulations (see the Supplement).

Because GCMs generally suffer from systematic biases to a certain extent, to improve the meteorological downscaling results, meteorological ICs/BCs derived from CESM-NCSU were bias-corrected using the method developed by Yahya et al. (2016) following the work of Xu and Yang (2012), Done et al. (2015), and Bruyère et al. (2014) based on the NCEP-FNL dataset. Monthly varying mean climatological biases in ICs/BCs between CESM-NCSU and NCEP-FNL were calculated and then subtracted from the original CESM-NCSU ICs/BCs to generate bias-corrected meteorological ICs/BCs. Major variables corrected in this study included air temperature, relative humidity, zonal wind, meridional wind, geopotential height, and soil moisture because Bruyère et al. (2014) found that correcting all boundary data provides the greatest improvement. The bias-correction method assumed that the biases remain the same in the future and allowed the retention of the CESM-NUSU simulated climatic changes in the mean seasonal state, diurnal cycle, and variance of inter-annual variation. The bias-correction method corrected the major biases in the meteorological variables that can cause serious issues for regional climate downscaling while retaining climate variability within the GCM for both current and future simulations.

2.3 Emissions

The CESM-NCSU simulations were driven with the RCPs emissions for both current and future decades (Glotfelty et al., 2017a). In this study, RCP 4.5 (Thomson et al., 2011) was selected as a representative scenario because it is a relatively medium scenario and aggressive emission reductions of major air pollutants in this scenario might be more suitable for China's future air quality control needs. The RCP dataset v2.0 provides global emission projections for CO (carbon monoxide), CH_4 (methane), SO_2 (sulfur dioxide), NO_x (nitrogen oxides), NMVOC (non-methane volatile organic compounds), NH_3 (ammonia), BC (black carbon) and OC (organic carbon) as monthly averages every 10 years starting from base year 2000 at a spatial resolution of 0.5 ° (http://tntcat.iiasa.ac.at:8787/RcpDb/dsd?Action=htmlpage&); emission sources include energy, industry, solvent use, transport, domestic combustion, agriculture, open burning of agricultural waste, waste treatment, biomass burning, shipping, and aviation sectors. In the RCP 4.5 emissions, only biomass burning, shipping, and aviation emission change monthly, and emission altitudes are given only for aviation. In order to achieve better performance for the regional WRF-CMAQ simulation, the MIX Asian emission inventory (a mosaic Asian anthropogenic emission inventory for the Model Inter-Comparison Study for Asia (MICS-Asia) and the (Hemispheric Transport of Air Pollution) HTAP projects, http://www.meicmodel.org; Li et al., 2017) for 2008 was used for the current year period (2006-2010), which has better spatial-temporal allocation profiles and particulate matter (PM) and VOC speciation profiles to generate model-ready emissions for regional scale air quality modeling over East Asia. The MIX inventory provides better monthly profiles compared to RCP 4.5 emissions and finer gridded emissions at a spatial resolution of 0.25 °, which is close to the resolution of 36 km used in WRF-CMAQ. Emissions of biomass burning, shipping and aviation sector were directly used from the RCP 4.5 emissions as they were not included in the MIX inventory.

Emissions from natural sources, including biogenic VOCs emissions, soil and lightning NO_x emissions, and dust emissions, were calculated inline within the two-way coupled WRF-CMAQ. The windblown dust emission scheme used in the CMAQ was developed by Tong et al. (2017). For biogenic emissions over East Asia, the Biogenic Emissions Inventory System (BEIS3) version 3.14 (Vukovich and Pierce, 2002; Schwede et al., 2005) was used in the coupled system rather than the widely used Model of Emissions of Gases and Aerosols from Nature version 2 (MEGAN2) (Guenther et al., 2012) because MEGAN2 has not been integrated into the CMAQ model. Soil NO_x emissions were also calculated by the inline BEIS3 module. Lightning NO_x emissions were inline calculated by estimating the number of lightning flashes based on the simulated convective precipitation (Allen et al., 2012).

2.4 Evaluation protocols

The model performance was evaluated against surface observations and satellite observations. Surface observations included hourly meteorological data from the National Climate Data Center (NCDC) and the real-time (i.e., hourly) concentrations of air pollutants from the China National Environmental Monitoring Center (CNEMC). The nationwide routine monitoring of $PM_{2.5}$ in China was not initiated until 2013; CNEMC began to release hourly concentrations of CO, SO_2, NO_2, O_3, $PM_{2.5}$, and PM_{10} in 74 major cities in China since January 2013 (http://www.cnemc.cn/), which is a much better dataset for air quality evaluation than the daily Air Pollution Index (API) dataset used in previous studies (Zhao et al., 2013; Zhang et al., 2016a). Satellite observations included data from the Global Precipitation Climatology Project (GPCP), the Clouds and the Earth's Radiant Energy System (CERES), the Moderate Resolution Imaging Spectroradiometer (MODIS), the Measurements of Pollution in the Troposphere (MOPITT), the Ozone Monitoring Instrument (OMI), and the SCanning Imaging Absorption SpectroMeter for Atmospheric ChartographY (SCIAMACHY). The variables that were evaluated in this study included temperature at 2 m (T2), relative humidity at 2 m (RH2), wind speed at 10 m (WS10), wind direction at 10 m (WDR10), precipitation (Precip), downward shortwave radiation at the surface (SWDOWN), downward longwave radiation at the surface (LWDOWN), net shortwave radiation (GSW), outgoing longwave radiation at the top of the atmosphere (OLR), shortwave cloud forcing (SWCF), longwave cloud forcing (LWCF), cloud fraction (CF); gas-phase species (CO, SO_2, NO_2, O_3), $PM_{2.5}$, PM_{10}; column CO, NO_2, SO_2, and HCHO; tropospheric ozone residual (TOR), and aerosol optical depth (AOD).

Two types of model evaluations were conducted in this study: evaluation for the climatological application to assess the model performance on a climatological average timescale over a 5-yr period (2006-2010) and evaluation for the short-term air quality application (2013) to assess the model performance on a monthly time scale. The observational data and simulated data were paired on an hourly basis for air quality evaluation in 2013, whereas they were paired on a 5-year average monthly basis for climatological-type evaluation when conducting statistical analyses. Moreover, evaluation for the air quality application in 2013 focused more on surface chemical variables because more observational data were available. For the climatological application, only satellite observations of column abundance were used to assess the chemical prediction because of the shortage in surface air quality observations during 2006-2010. The performance statistical analyses were performed following Zhang et al. (2006, 2009a, b). The statistical parameters included correlation coefficient (R), mean bias (MB), normalized mean biases (NMB), mean absolute gross error (MAGE), and root mean square error (RMSE). The statistical evaluation was in general performed for the entire regional domain. However, evaluation for surface chemical variables focused more on China where hourly air quality monitoring data are available.

3 Results and discussion

3.1 Model performance for climatological application (2006-2010)

Table 3 summarizes the performance statistics for the climatological application during the period 2006-2010. The model performed well for T2 and RH2, with MBs of -0.6 ℃ and 0.8%, correlation coefficients of 0.97 and 0.72, MAGEs of 2.4 ℃ and 9.7%, and RMSEs of 3.2 ℃ and 12.6%, respectively. From the evaluation results from Xu and Xu (2012), the Coupled Model Inter-comparison Project Phase 5 (CMIP5) multi-models tended to underpredict T2 over China with MBs ranging from –1.0 ℃ to –2.0 ℃ for the period 1961–2005. The improved statistical performance for T2 in this study compared with CMIP5 models may be related to the use of the regional model WRF and the bias-correction technique applied for CESM downscaling. This indicates that WRF-CMAQ driven by bias-corrected CESM-NCSU ICs/BCs performs well on a climatological average timescale. WS10 was moderately overpredicted by 22.2%, with an MB of 0.6 m/s, an MAGE of 1.2 m/s and a RMSE of 1.6 m/s. Large overpredictions in WS10 with NMBs of 48.7%-101.0% from WRF simulations have been reported in the literature (Penrod et al., 2014; Cai et al., 2016; Zhang et al., 2016a) because of unresolved subgrid-scale topographic features and uncertainties in parameterizations of turbulent fluxes in WRF (Hanna and Yang, 2001; Rontu, 2006; Mass and Ovens, 2011). The overpredictions in WS10 are likely caused by low surface drag due to the inappropriate representation of surface roughness because the detailed surface structure cannot be reproduced at a coarse grid resolution of 36-km. The high wind biases were reduced in this study because of the use of the simple wind correction method of Mass and Ovens (2010). The USGS 24-category land use data is out of date for China where urbanization has been dramatic, which would also partly contribute to the overprediction in WS10. Precipitation was well-predicted against GPCP with an NMB of -0.9% and moderately overpredicted by 27.4% against NCDC, and the model could generally capture the observed spatial distribution (see Figure 2). The convective precipitation dominated the overprediction of total precipitation in the southern oceanic area, which may be possibly due to overprediction of convective precipitation intensity by the Kain–Fritsch cumulus scheme. The non-convective precipitation dominated the overprediction of total precipitation in the northeastern oceanic area, which could be attributed to possible errors in the Morrison double-moment microphysics scheme. Emery et al. (2001) suggested the benchmarks for satisfactory performance for T2 (MB within ±0.5 °C, MAGE of ≤ 2.0 °C) and WS10 (MB within ± 0.5 m/s, MAGE and RMSE of 2.0 m/s). In the climatological application, the MB and MAGE of T2 and the MB of WS10 are close to the benchmark, the MAGE and RMSE of WS10 are within the benchmark, and hence the performance is deemed acceptable.

As shown in Figure 3, MBs for 5-year average T2, RH2, WS10 and precipitation were generally small over eastern China. Relatively large biases were found over the coast, especially in Japan and North and South Korea, similar to previous WRF simulations (Chen et al., 2015; Zhang et al., 2016a), which indicates certain limitations in the WRF model over complex terrain and air-sea interactions. Figure 4 compares the 5-year average monthly simulated T2, RH2, WS10 and precipitation against observations. The model could generally capture the monthly variations of T2, although there were cold biases of –1.0 ℃ in spring and summer months. The model also well reproduced the observed monthly variations of RH2; the minimum RH2 was observed in April (~ 63%) and the maximum RH2 was observed in July (~ 76%). For WS10, the model predicted a minimum in summer, which was similar to observations. However, the overprediction of WS10 in winter (with an MB of 0.9 m/s) was slightly higher than in other seasons (with an MB of 0.5 m/s). The model accurately predicted the observed precipitation maximum occurring in summer (~ 5 mm/day) and the minimum in winter (~ 1 mm/day). Overall, the climate predictions of WRF-CMAQ represent a good approximation of the current atmosphere in terms of spatial distributions and seasonal variations, and can thus provide acceptable meteorological fields for air quality simulations.

Figure 5 compares the 5-year averaged simulated spatial distributions of cloud, aerosol and radiation variables against satellite observations. CF was underpredicted with an NMB of -30.5%, similar to previous WRF simulations over East Asia

(Liu et al., 2016). Large underpredictions of CF were found over the oceanic area in the southern part of the domain. Such underpredictions may be because of the model's limitations in simulating cloud microphysics and the lack of aerosol-cloud interactions. LWDOWN was predicted very well with an NMB of -2.8%, whereas SWDOWN was overpredicted with an NMB of 14.1%. The overpredictions of SWDOWN were likely because of underpredictions of cloud radiative forcing resulted from underpredictions of CF as well as underpredictions of aerosol direct radiative forcing resulted from underpredictions of AOD. WRF version 3.4 has neglected sub-grid cloud feedbacks to radiation, which could contribute in part to the overpredictions in SWDOWN (Alapaty et al., 2012). Overpredictions in SWDOWN generally corresponded to underpredictions in CF. Fewer clouds led to underpredictions in SWCF (with an MB of 13.6 W m^{-2}), which allowed more SWDOWN to reach the ground. The overpredictions in OLR were associated with the underpredictions in LWCF (with an MB of -12.7 W m^{-2}). The model underpredicted AOD with an NMB of -36.3%; however, it could capture the high value over eastern China. Similar underpredictions of AOD were found over North America using offline-coupled WRF and CMAQ (Wang et al., 2012; Penrod et al., 2014) and the two-way coupled WRF-CMAQ (Hogrefe et al., 2015).

Figure 6 compares the 5-year averaged simulated spatial distributions of column mass abundance of chemical variables against satellite retrievals. In general, the model could reproduce the spatial distributions of column mass abundance of chemical variables; correlation coefficients were generally higher than 0.8. Column CO, NO_2, HCHO, and O_3 were well predicted in terms of domain mean performance statistics, with NMBs of -11.7%, 18.3%, -4.0%, and 16.4%, respectively. Large underpredictions in column CO occurred over eastern China as well as North and South Korea and Japan, likely because of uncertainties in anthropogenic emissions as well as biomass burning (Streets et al., 2003). Simulated TOR could capture the observed low values in the south and in Tibet and the high values in the north. The overprediction of TOR in the north can be attributed to uncertainties in upper layer BCs of O_3 which dominate O_3 concentrations in upper troposphere as well as total column O_3 (Zhang et al., 2016a, c). Column NO_2 was moderately overpredicted by 18.3%. Potential uncertainties in NO_x emissions and the model treatment of deposition and chemistry processes may contribute to the model-observation difference. As discussed by Lin et al. (2010) and Han et al. (2015), there are several uncertainties in the modeled NO_x lifetime. Uncertainties in the NO_2 column retrievals from OMI (with a relative error of 25%, Boersma et al., 2011) and the averaging kernels (Han et al., 2015) could also help to explain the bias. Although column SO_2 was slightly overpredicted, with an NMB of 7.5% over the entire domain, larger overprediction occurred over eastern China. The overall error annual mean SO_2 columns retrieved from satellites could be as large as 45%–80% in polluted regions (Lee et al., 2009), which might impact the evaluation results of column SO_2. Large uncertainties in SO_2 emissions (Hong et al., 2017) would also contribute to the biases in column SO_2. Column HCHO over eastern China was well predicted in terms of both the magnitude and the spatial pattern. Possible reasons for the overpredictions of column HCHO in Southeast Asia and northeastern India include uncertainties in biogenic and anthropogenic VOCs emissions, and satellite retrievals.

3.2 Model performance for short-term air quality application in 2013

Table S3 summarizes performance statistics of meteorological variables for the air quality application in January, April, and July, 2013. The model performance for major meteorological variables in the air quality application was similar to that for the climatological application. Note that for the air quality application driven with NCEP-FNL data, the observation and simulation data pairs for surface meteorological variables against NCDC observational data were on an hourly basis. The high correlations for major meteorological variables in Table S3 indicated that the model showed good skills in hourly meteorological predictions, thus NCEP-FNL data were sufficient to support air quality applications for hourly air quality predictions.

Table 4 summarizes performance statistics of chemical variables for the air quality application in January, April and July, 2013. The model performed very well for surface concentrations of SO_2, NO_2, $PM_{2.5}$ in January and April, with NMBs generally within ±10%, and moderately overpredicted O_3 and $PM_{2.5}$ in July. Surface CO concentrations were underpredicted in all months with NMBs ranging from -48% to -33%. The simulated column abundances of CO were also underpredicted with NMBs ranging from -14% to -2%, indicating that CO emissions were likely underestimated. Overpredictions in WS10 shown in Table S3 also contributed to the underpredictions in surface CO concentrations. Surface SO_2 and NO_2 concentrations were largely overpredicted in summer with NMBs higher than 40%, especially at nighttime, which could be attributed to biases in meteorological predictions (e.g., turbulent mixing) (Pleim, et al., 2015), uncertainties in emissions (e.g., monthly profiles and vertical distributions) (Zhang et al., 2016a) and biases in model treatments (e.g., SO_2 wet deposition). $PM_{2.5}$ concentrations were slightly overpredicted in winter with an NMB of 6.4%. O_3 was moderately overpredicted in summer with an NMB of 18.2%, which could be partly because of overpredictions in SWDOWN and partly because of overpredicted NO_2. Figure 7 shows the spatial distribution of $PM_{2.5}$ and O_3 in January, April, and July, 2013. High $PM_{2.5}$ concentrations were predicted in winter over the North China Plain (NCP) and the Sichuan Basin (SCB) where anthropogenic emissions are high, consistent with the observational data. $PM_{2.5}$ concentrations over western China were predicted to be higher in April because of spring dust emissions that could contribute not only coarse PM but also fine PM. In summer, O_3 concentrations were predicted to be higher over northern China but lower over southern China because the East Asian summer monsoon (EASM) brings clean air masses from the oceans in the south (He et al., 2008; Wang et al., 2011). The simulated spatial distribution of O_3 was consistent with observational data. Figure 8 shows the time series of hourly concentrations of $PM_{2.5}$ in January and O_3 in July over three heavily-polluted regions in China: the Beijing-Tianjin-Hebei (BTH) region, the Yangtze River Delta (YRD) and the Pearl River Delta (PRD). The model well reproduced the observed hourly variations of $PM_{2.5}$ as well as the observed diurnal and daily variations of O_3 over three key regions. The simulated diurnal variability of $PM_{2.5}$ in BTH and YRD is somewhat larger than observations. The overprediction in surface $PM_{2.5}$ concentration at nighttime might be partly attributed to errors in the parameterization of PBL turbulent mixing (Pleim, et al., 2015). The discrepancies between the simulated and observed time series of $PM_{2.5}$ may be attributable to several possible causes, including inaccuracies in meteorological predictions (e.g., turbulent mixing, precipitation, and WS10) and uncertainties in some model treatments (e.g., secondary organic aerosol formation and dry and wet deposition). The model slightly overpredicted the peak concentrations of O_3, which may be partly because of overpredictions in SWDOWN.

The CMAQ performance of chemical predictions in this study was comparable to or even better than those of other air quality studies over East Asia (Wang et al., 2009; 2012; Liu et al., 2010; Zheng et al., 2015; Hu et al., 2016; Liu et al., 2016; Zhang et al., 2016a). This study predicted relatively well for most chemical species in most months. Compared with other regional modeling studies, WRF-CMAQv5.0.2 used in this study outperformed MM5/CMAQv4.6, which tend to underpredict the surface concentrations of major species with NMBs generally greater than -40% and overpredict surface O_3 concentrations in most months with NMBs generally higher than 20% over East Asia according to the evaluation results of Zhang et al. (2016a). A relatively good performance of CMAQv5.0.1 was also reported by Hu et al. (2016). Global models such as GEOS-Chem and CESM tend to underpredict $PM_{2.5}$ concentrations (by about -50% as reported by Jiang et al., 2013) and overpredict O_3 concentrations (by about 50% as reported by He and Zhang, 2014; Wang et al., 2013) in China/East Asia because of relatively coarse grid resolution and limitations in some model treatments (e.g., missing emissions of unspeciated primary $PM_{2.5}$, and discrepancies in surface layer height and vertical mixing). The comparison of the model performances for $PM_{2.5}$ and O_3 predictions of WRF-CMAQ and CESM is shown in Figure 9. Compared with global models, WRF-CMAQ was able to capture the high $PM_{2.5}$ concentrations in urban areas where most observational data were obtained. The model was also able to predict low O_3 concentrations and predicted well for O_3 over China with small NMBs of 15-30% in both winter and summer. CESM tended to underpredict $PM_{2.5}$ concentrations over China with NMBs ranging from -30% to -70% and overpredict O_3 concentrations with NMBs ranging from 50% to 100%. It should be noted that although the years of observational data and CESM simulations were not consistent (i.e., 2013 and 2006-2010, respectively), we do not think inter-annual changes in meteorological fields and emissions contributed to such large biases, as is indicated by the results of the two WRF-CMAQ simulations for the two periods (see Figure 9).

3.3 Improvements of model performance within the modeling system

To demonstrate the model improvements made in this study, sensitivity simulations were conducted. The comparison of the baseline simulation (i.e., NCEP_BASE) and the sensitivity simulation (i.e., NCEP_BASE_WoImp) against observational data is shown in Figure 10. The coupling model system predicted AOD relatively well. The two-way coupled WRF-CMAQ with aerosol direct feedbacks could generally replicate lower SWDOWN values over heavily-polluted regions (such as eastern and southern China). The overprediction of SWDOWN in January in the sensitivity simulation without aerosol feedbacks (with an NMB of 19.9%) was significantly reduced when the aerosol feedbacks were included (with an NMB of 11.1%). The remaining overprediction in SWDOWN in the NCEP_BASE simulation was because of underpredictions in AOD and CF, which indicates that including the aerosol feedback in the coupled system is important for better simulating the shortwave radiation fields in WRF, consistent with the findings of Yahya et al. (2016).

The chemical composition downscaling approach was applied in this study to provide dynamical chemical BCs for regional modeling. The main advantage of applying chemical composition downscaling is the representation of global changes in atmospheric composition in regional simulations, which is important to better simulate relatively long-lived species such as CO and O_3 under a globally changing atmospheric environment. Another advantage is the representation of spatial and temporal variations in BCs, which could also help improve the model performance (Tang et al., 2009). The comparison between BCs derived from CESM and fixed boundary profiles provided by the operational CMAQ system is shown in Fig. S2. CESM-derived BCs produced better spatial variability, such as higher O_3 concentrations from the northern boundary and lower O_3 concentrations from the southern boundary. When using BCs derived from CESM, the model performance of column variables (e.g., TOR) was improved in terms of spatial distribution and seasonal variations. The overprediction of TOR in January in the sensitivity simulation using fixed BCs (with an NMB of 48.9%) was significantly reduced when using CESM-derived BCs (with an NMB of 22.3%).

Inline emissions from natural sources were calculated within the coupled system. Although BEIS3 has been widely used in the U.S, the model performance over other regions such as East Asia should be evaluated. We conducted the sensitivity simulation using offline MEGAN2, which has been widely used over East Asia. The major differences in emissions were found for isoprene emissions (see Figure 11). The summertime isoprene emissions over China estimated using MEGAN2 were approximately 100% higher than those estimated using BEIS3. Similar large discrepancies in isoprene emissions were also found from previous studies over the U.S. (Lam et al., 2011; Hogrefe et al., 2011) because of the different methods used to estimate isoprene emissions (Lam et al., 2011). We evaluated CMAQ-simulated HCHO columns using the BEIS3 and MEGAN2 emissions against OMI satellite observations. HCHO columns have been used to evaluate biogenic VOC emission inventories (Han et al., 2013) because HCHO is an intermediate oxidation product of anthropogenic and biogenic VOCs. The evaluation results in Figure 10 show that using BEIS3 emissions in CMAQ could capture both the magnitude and the spatial pattern of HCHO columns from OMI, whereas using MEGAN2 emissions resulted in 30%~50% overpredictions of HCHO columns over northern China.

The default dust scheme in CMAQ developed by Tong et al. (2017) underpredicted dust emissions over East Asia (Fu et al., 2014; Dong et al., 2015). In this study, the dust module was revised with updated friction velocity thresholds to avoid double counting of the impacts of soil moisture (Dong et al., 2015). Compared with sensitivity simulation results using the default dust scheme, the revised model was able to simulate springtime dust emissions over northwest China, where dust storms often occur. As shown in Figure 10, the revised model predicted AOD values in April as high as 0.2~0.6 over northwest China, which were much closer to the satellite observations, while the original model generally predicted AOD values less than 0.05 over northwest China. The improved dust module was able to capture the spatial distribution and the temporal variations of dust emissions, although some biases still existed.

3.4 Aerosol direct effects on regional climate and air quality

To examine the aerosol direct effects on regional climate and air quality, we conducted a sensitivity simulation without aerosol feedback. The differences between the simulations with and without aerosol direct feedback (i.e., CESM_BASE: with feedback and CESM_BASE_Sens: without feedback) are shown in Figure 12. Aerosol direct radiative effects resulted in a reduction of shortwave radiation reaching the surface because of aerosol extinction (i.e., scattering and absorbing). The aerosol extinction led to a more stable planetary boundary layer (PBL) during the haze episode through enhancing the temperature inversion in two ways: diminished surface solar radiation led to a decrease of air temperature at the surface, and the absorption of light-absorbing particles such as black carbon (BC) caused an increase of air temperature in the upper PBL. As shown in Figure 12, the domain mean reductions in SWDOWN were -7.5 W m^{-2} in January and -7.0 W m^{-2} in July. The domain mean reductions in T2 were -0.09 ℃ in January and -0.08 ℃ in July. The effects of anthropogenic aerosols on SWDOWN and T2 were comparable to the results over East Asia from WRF/Chem-MADRID (Liu et al., 2016) and WRF/Chem (Zhang et al., 2016b). The reductions in SWDOWN in July were somewhat smaller than those of Liu et al. (2016) because aerosol indirect effects were not considered in this study. We also found that SWDOWN decreased in July in northwest China because of the natural dust aerosols. Slight increases in SWDOWN and T2 occurred in July in some areas, which could be attributed to semi-indirect effects of aerosols (Forkel et al., 2012). The aerosol feedbacks were significant over heavily-polluted regions such as eastern China and the Sichuan Basin. With the aerosol feedbacks, the monthly mean SWDOWN and planetary boundary layer height (PBLH) in January decreased by 21.8 W m^{-2} (14%) and 35.7 m (7.6%), respectively, in major cities of China, and air temperature at the surface decreased by 0.45 ℃.

The aerosol direct feedbacks affect not only climate but also air quality because of changing climate. We investigated the aerosol direct effects on air quality in different seasons. Enhanced PBL stability resulted from the aerosol direct effects enhanced the air pollution by suppressing the dispersion of air pollutants. Because of aerosol feedbacks, mean concentrations of major pollutants (except for O_3) over major cities of China increased by 4.8%~9.5%, and $PM_{2.5}$ concentrations increased by 6.6 μg m^{-3} in January. However, O_3 concentrations in January decreased by 5.1% because of aerosol feedbacks, which may be attributed to the increased NO_x titration resulted from increased atmospheric stability and reduced PBL height. Similar aerosol direct effects were also found in July. Because of aerosol feedbacks, mean concentrations of major pollutants (except for O_3) increased by 4.8%~7.1% over major cities in China, and $PM_{2.5}$ concentrations increased by 3.8 μg m^{-3} in July. The aerosol direct effects on $PM_{2.5}$ concentrations in July were smaller than those in January because of lower aerosol loadings in July. Compared with simulated aerosol effects over the continental U.S. and Europe (Zhang et al., 2010; Hogrefe et al., 2015), the magnitudes of aerosol effects on regional climate and air quality were much larger over East Asia because of higher aerosol loadings resulted from severe regional pollution.

4 Conclusions

A regional coupled climate-chemistry modeling system using the dynamical downscaling technique was established by linking the global CESM model and the regional two-way coupled WRF-CMAQ model for the purpose of comprehensive assessments of regional climate change and air quality and their interactions within one modeling framework. The modeling system took full advantage of global climate-chemistry models that can well predict large-scale global changes and regional models that can better represent regional phenomena. The modeling system was applied over East Asia for a multiyear climatological application during 2006-2010 under RCP 4.5 as well as a short-term air quality application for three months in 2013 driven by the NCEP-FNL reanalysis dataset. Comprehensive model evaluation was conducted against surface observations and satellite observations to assess the model's performance.

The two-way coupled WRF-CMAQ generally performed well for both the climatological and the short-term air quality applications. The model was able to predict major meteorological variables satisfactorily. The improved statistical performance for T2 in this study (with an MB of -0.6 ℃) compared with CMIP5 multi-models may be related to the use of the regional model WRF and the bias-correction technique applied for CESM downscaling. The model showed good ability to predict $PM_{2.5}$ in winter (with an NMB of 6.4% in 2013) and O_3 in summer (with an NMB of 18.2% in 2013) in terms of statistical performance and spatial distributions. Compared with global models that tend to underpredict $PM_{2.5}$ concentrations in China, WRF-CMAQ was able to capture the high $PM_{2.5}$ concentrations in urban areas. Model improvements made in this study were quantified by the sensitivity simulation. The coupled modeling system with direct aerosol feedbacks predicted AOD relatively well and significantly reduced the overprediction of SWDOWN (NMBs in January were reduced from 19.9% to 11.1%). The two-way coupled WRF-CMAQ with aerosol direct feedbacks could generally replicate lower SWDOWN values over heavily polluted regions (such as eastern and southern China). Applying chemical composition downscaling to introduce global background changes in atmospheric composition could also help improve the model performance of column variables (e.g., TOR). The overprediction of TOR in January when using fixed BCs (with an NMB of 48.9%) was significantly reduced when using CESM-derived BCs (with an NMB of 22.3%). The BEIS3 biogenic online emission module was applied in this study, and the model performance over East Asia was examined. Sensitivity simulations showed that using BEIS biogenic emissions resulted in improved performance for column HCHO, whereas using MEGAN2 emissions resulted in large overpredictions (30%~50%) of HCHO columns over northern China. The improved dust module was able to capture the spatial distribution and the temporal variations of dust emissions, although some biases remained. The revised model was able to capture the high AOD values (0.2~0.6) in April over northwest China where dust storms often occur in spring. We also demonstrated the impacts of aerosol direct effects on climate and air quality to address important climate-chemistry interactions. With aerosol direct feedbacks in January, the monthly mean SWDOWN and PBLH over major cities of China decreased by 21.8 W m^{-2} (14%) and 35.7 m (7.6%), respectively, air temperature at the surface decreased by 0.45 ℃, and mean concentrations of most pollutants (except for O_3) increased by 4.8%-9.5%. The aerosol effects on climate and air quality were more significant in East Asia than the U.S. and Europe because of higher aerosol loadings resulting from severe pollution in East Asia, which indicates the need to apply online-coupled models over East Asia for regional climate and air quality modeling and to study the important climate-chemistry interactions.

This work has established the baseline simulation for WRF-CMAQ application for a future time period in order to access the projected changes in climate and anthropogenic emissions on future air quality over East Asia under the RCP4.5 scenario. Although the modeling system generally had acceptable performance, this work suggested further model development and improvement that could improve the model performance. First, this work used a highly simplified method to correct wind bias, a more rigorous method that is available in WRF version 3.6 should be used. Second, larger biases were found for cloud fraction against satellite data and also for surface SO_2 concentrations during summer against surface observations. The performance of cloud variables was not as good as that of other meteorological variables, and underpredictions of cloud fraction resulted in overpredictions of SWDOWN and underpredictions of shortwave and longwave cloud forcing. The model biases possibly resulted from uncertainties in simulated meteorology (e.g., precipitation and WS10), emissions (e.g., vertical profiles and biogenic emissions), boundary conditions derived from the global CESM model, and limitations in some model treatments (e.g., cumulus scheme, secondary organic aerosol). Further model improvement should focus on these areas identified from this work. Finally, aerosol indirect effects on cloud properties are currently not included in the released version of the two-way coupled WRF-CMAQ model. An initial implementation and evaluation of aerosol indirect effects on resolved clouds over the U.S. has recently been completed (Yu et al., 2014), but its performance outside the U.S. needs to be further evaluated in subsequent studies.

Code availability

The two-way coupled WRF-CMAQ model is open-source and publicly available. The WRF version 3.4 codes can be downloaded at http://www2.mmm.ucar.edu/wrf/users/download/get_source.html. The CMAQ version 5.0.2 codes and the WRF-CMAQ two-way package can be downloaded at https://www.cmascenter.org/download.cfm. The build instructions and run instructions for the two-way coupled WRF-CMAQ model are available at http://www.airqualitymodeling.org/cmaqwiki/index.php?title=CMAQv5.0.2_Two-way_model_release_notes. We have modified the surface drag parameterization in WRF3.4 for correction of wind speed bias and the dust module in CMAQv5.0.2 to generate more dust emissions. The modified codes can be provided upon request.

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References

Alapaty, K., Herwehe, J. A., Otte, T. L., Nolte, C. G., Bullock, O. R., Mallard, M. S., Kain, J. S., and Dudhia, J.: Introducing subgrid-scale cloud feedbacks to radiation for regional meteorological and climate modeling, Geophys. Res. Lett., 39, L24809, doi:10.1029/2012GL054031, 2012.

Allen, D. J., Pickering, K. E., Pinder, R. W., Henderson, B. H., Appel, K. W., and Prados, A.: Impact of lightning-NO on eastern United States photochemistry during the summer of 2006 as determined using the CMAQ model, Atmos. Chem. Phys., 12, 1737–1758, doi:10.5194/acp-12-1737-2012, 2012.

Appel, K. W., Pouliot, G. A., Simon, H., Sarwar, G., Pye, H. O. T., Napelenok, S. L., Akhtar, F., and Roselle, S. J.: Evaluation of dust and trace metal estimates from the Community Multiscale Air Quality (CMAQ) model version 5.0, Geosci. Model Dev., 6, 883–899, doi:10.5194/gmd-6-883-2013, 2013.

Baklanov, A., Schluenzen, K., Suppan, P., Baldasano, J., Brunner, D., Aksoyoglu, S., Carmichael, G., Douros, J., Flemming, J., Forkel, R., Galmarini, S., Gauss, M., Grell, G., Hirtl, M., Joffre, S., Jorba, O., Kaas, E., Kaasik, M., Kallos, G., Kong, X., Korsholm, U., Kurganskiy, A., Kushta, J., Lohmann, U., Mahura, A., Manders-Groot, A., Maurizi, A., Moussiopoulos, N., Rao, S. T., Savage, N., Seigneur, C., Sokhi, R. S., Solazzo, E., Solomos, S., Sorensen, B., Tsegas, G., Vignati, E., Vogel, B., and Zhang, Y.: Online coupled regional meteorology chemistry models in Europe: current status and prospects, Atmos. Chem. Phys., 14, 317–398, doi:10.5194/acp-14-317-2014, 2014.

Boersma, K. F., Eskes, H. J., Dirksen, R. J., van der A, R. J., Veefkind, J. P., Stammes, P., Huijnen, V., Kleipool, Q. L., Sneep, M., Claas, J., Leitão, J., Richter, A., Zhou, Y., and Brunner, D.: An improved tropospheric NO_2 column retrieval algorithm for the Ozone Monitoring Instrument, Atmos. Meas. Tech., 4, 1905–1928, doi:10.5194/amt-4-1905-2011, 2011.

Bruyere, C. L., Done, J. M., Holland, G. J., and Fredrick, S.: Bias corrections of global models for regional climate simulations of high-impact weather, Clim. Dynam., 43, 1847–1856, doi:10.1007/s00382-013-2011-6, 2014.

Cai, C., Zhang, X., Wang, K., Zhang, Y., Wang, L., Zhang, Q., Duan, F., He, K., and Yu, S.: Incorporation of new particle formation and early growth treatments into WRF/Chem: Model improvement, evaluation, and impacts of anthropogenic aerosols over East Asia, Atmos. Environ., 124, 262–284, doi:10.1016/j.atmosenv.2015.05.046, 2016.

Carlton, A. G., Bhave, P. V., Napelenok, S. L., Edney, E. D., Sarwar, G., Pinder, R. W., Pouliot, G. A., and Houyoux, M.: Model Representation of Secondary Organic Aerosol in CMAQv4.7, Environ. Sci. Technol., 44, 8553–8560, doi:10.1021/es100636q, 2010.

Chen, Y., Zhang, Y., Fan, J., Leung, L. R., Zhang, Q., and He, K.: Application of an Online-Coupled Regional Climate Model, WRF-CAM5, over East Asia for Examination of Ice Nucleation Schemes: Part I. Comprehensive Model Evaluation and Trend Analysis for 2006 and 2011, Climate, 3, 627–667, doi:10.3390/cli3030627, 2015.

Done, J. M., Holland, G. J., Bruyere, C. L., Leung, L. R., and Suzuki-Parker, A.: Modeling high-impact weather and climate: lessons from a tropical cyclone perspective, Climatic Change, 129, 381–395, doi:10.1007/s10584-013-0954-6, 2015.

Dong, X., Fu, J. S., Huang, K., Tong, D., and Zhuang, G.: Model development of dust emission and heterogeneous chemistry within the Community Multiscale Air Quality modeling system and its application over East Asia, Atmos. Chem. Phys., 16, 8157–8180, doi:10.5194/acp-16-8157-2016, 2016.

Emery, C., Tai, E., and Yarwood, G.: Enhanced meteorological modeling and performance evaluation for two texas episodes, Report to the Texas Natural Resources Conservation Commission, prepared by ENVIRON, International Corp., Novato, CA, available at: http://www.tceq.state.tx.us/assets/public/implementation/air/am/contracts/reports/mm/EnhancedMetModelingAndPerformanceEvaluation.pdf, 2001.

Fiore, A. M., Naik, V., Spracklen, D. V., Steiner, A., Unger, N., Prather, M., Bergmann, D., Cameron-Smith, P. J., Cionni, I., Collins, W. J., Dalsoren, S., Eyring, V., Folberth, G. A., Ginoux, P., Horowitz, L. W., Josse, B., Lamarque, J., MacKenzie, I. A., Nagashima, T., O'Connor, F. M., Righi, M., Rumbold, S. T., Shindell, D. T., Skeie, R. B., Sudo, K., Szopa, S., Takemura, T., and Zeng, G.: Global air quality and climate, Chem. Soc. Rev., 41, 6663–6683, doi:10.1039/c2cs35095e, 2012.

Forkel, R., Werhahn, J., Hansen, A. B., McKeen, S., Peckham, S., Grell, G., and Suppan, P.: Effect of aerosol-radiation feedback on regional air quality - A case study with WRF/Chem, Atmos. Environ., 53, 202–211, doi:10.1016/j.atmosenv.2011.10.009, 2012.

Fu, X., Wang, S. X., Cheng, Z., Xing, J., Zhao, B., Wang, J. D., and Hao, J. M.: Source, transport and impacts of a heavy dust event in the Yangtze River Delta, China, in 2011, Atmos. Chem. Phys., 14, 1239–1254, doi:10.5194/acp-14-1239-2014, 2014.

Fuzzi, S., Baltensperger, U., Carslaw, K., Decesari, S., van der Gon, H. D., Facchini, M. C., Fowler, D., Koren, I., Langford, B., Lohmann, U., Nemitz, E., Pandis, S., Riipinen, I., Rudich, Y., Schaap, M., Slowik, J. G., Spracklen, D. V., Vignati, E., Wild, M., Williams, M., and Gilardoni, S.: Particulate matter, air quality and climate: lessons learned and future needs, Atmos. Chem. Phys., 15, 8217–8299, doi:10.5194/acp-15-8217-2015, 2015.

Gan, C. M., Binkowski, F., Pleim, J., Xing, J., Wong, D., Mathur, R., and Gilliam, R.: Assessment of the aerosol optics component of the coupled WRF-CMAQ model using CARES field campaign data and a single column model, Atmos. Environ., 115, 670–682, doi:10.1016/j.atmosenv.2014.11.028, 2015.

Gantt, B., He, J., Zhang, X., Zhang, Y., and Nenes, A.: Incorporation of advanced aerosol activation treatments into CESM/CAM5: model evaluation and impacts on aerosol indirect effects, Atmos. Chem. Phys., 14, 7485–7497, doi:10.5194/acp-14-7485-2014, 2014.

Gao, Y., Fu, J. S., Drake, J. B., Lamarque, J. F., and Liu, Y.: The impact of emission and climate change on ozone in the United States under representative concentration pathways (RCPs), Atmos. Chem. Phys., 13, 9607–9621, doi:10.5194/acp-13-9607-2013, 2013.

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K., and Wang, X.: The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1): an extended and updated framework for modeling biogenic emissions, Geosci. Model Dev., 5, 1471–1492, doi:10.5194/gmd-5-1471-2012, 2012.

Glotfelty, T., He, J., and Zhang, Y.: The Impact of Future Climate Policy Scenarios on Air Quality and Aerosol/Cloud Interactions using an Advanced Version of CESM/CAM5: Part I. Model Evaluation for the Current Decadal Simulations, Atmos. Environ., 152, 222–239, doi: 10.1016/j.atmosenv.2016.12.035, 2017a.

Glotfelty, T., He, J., and Zhang, Y.: Improving Organic Aerosol Treatments in CESM/CAM5: Development, Application, and Evaluation, Journal of Advances in Modeling Earth Systems, in review, 2017b.

Glotfelty, T., and Zhang, Y.: The Impact of Future Climate Policy Scenarios on Air Quality and Aerosol/Cloud Interactions using an Advanced Version of CESM/CAM5: Part II. Future Trend Analysis and Impacts of Projected Anthropogenic Emissions, Atmos. Environ., 152, 531-552, doi: 10.1016/j.atmosenv.2016.12.034, 2017.

Han, K. M., Park, R. S., Kim, H. K., Woo, J. H., Kim, J., and Song, C. H.: Uncertainty in biogenic isoprene emissions and its impacts on tropospheric chemistry in East Asia, Sci. Total Environ., 463, 754–771, doi:10.1016/j.scitotenv.2013.06.003, 2013.

Han, K. M., Lee, S., Chang, L. S., and Song, C. H.: A comparison study between CMAQ-simulated and OMI-retrieved NO_2 columns over East Asia for evaluation of NOx emission fluxes of INTEX-B, CAPSS, and REAS inventories, Atmos. Chem. Phys., 15, 1913-1938, doi:10.5194/acp-15-1913-2015, 2015.

Hanna, S. R., and Yang, R. X.: Evaluations of mesoscale models' simulations of near-surface winds, temperature gradients, and mixing depths, J. Appl. Meteorol., 40, 1095–1104, doi:10.1175/1520-0450(2001)040<1095:EOMMSO>2.0.CO;2, 2001.

He, J., and Zhang, Y.: Improvement and further development in CESM/CAM5: gas-phase chemistry and inorganic aerosol treatments, Atmos. Chem. Phys., 14, 9171–9200, doi:10.5194/acp-14-9171-2014, 2014.

He, J., Zhang, Y., Glotfelty, T., He, R., Bennartz, R., Rausch, J., and Sartelet, K.: Decadal simulation and comprehensive evaluation of CESM/CAM5.1 with advanced chemistry, aerosol microphysics, and aerosol-cloud interactions, J. Adv. Model. Earth Sy., 7, 110–141, doi:10.1002/2014MS000360, 2015a.

He, J., Zhang, Y., Tilmes, S., Emmons, L., Lamarque, J. F., Glotfelty, T., Hodzic, A., and Vitt, F.: CESM/CAM5 improvement and application: comparison and evaluation of updated CB05_GE and MOZART-4 gas-phase mechanisms and associated impacts on global air quality and climate, Geosci. Model Dev., 8, 3999–4025, doi:10.5194/gmd-8-3999-2015, 2015b.

He, Y. J., Uno, I., Wang, Z. F., Pochanart, P., Li, J., and Akimoto, H.: Significant impact of the East Asia monsoon on ozone seasonal behavior in the boundary layer of Eastern China and the west Pacific region, Atmos. Chem. Phys., 8, 7543–7555, doi:10.5194/acp-8-7543-2008, 2008.

Hogrefe, C., Isukapalli, S. S., Tang, X., Georgopoulos, P. G., He, S., Zalewsky, E. E., Hao, W., Ku, J., Key, T., and Sistla, G.: Impact of Biogenic Emission Uncertainties on the Simulated Response of Ozone and Fine Particulate Matter to Anthropogenic Emission Reductions, J. Air Waste Manage., 61, 92–108, doi:10.3155/1047-3289.61.1.92, 2011.

Hogrefe, C., Pouliot, G., Wong, D., Torian, A., Roselle, S., Pleim, J., and Mathur, R.: Annual application and evaluation of the online coupled WRF-CMAQ system over North America under AQMEII phase 2, Atmos. Environ., 115, 683–694, doi:10.1016/j.atmosenv.2014.12.034, 2015.

Hong, C., Zhang, Q., He, K., Guan, D., Li, M., Liu, F., and Zheng, B.: Variations of China's emission estimates: response to uncertainties in energy statistics, Atmos. Chem. Phys., 17, 1227-1239, doi:10.5194/acp-17-1227-2017, 2017.

Hu, J., Chen, J., Ying, Q., and Zhang, H.: One-year simulation of ozone and particulate matter in China using WRF/CMAQ modeling system, Atmos. Chem. Phys., 16, 10333–10350, doi:10.5194/acp-16-10333-2016, 2016.

Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A., and Collins, W. D.: Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative transfer models, J. Geophys. Res. Atmos., 113, D13103, doi:10.1029/2008JD009944, 2008.

IPCC (Intergovernmental Panel on Climate Change): Climate change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K.B., Tignor, M., and Miller, H.L., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2007.

IPCC (Intergovernmental Panel on Climate Change): Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Stocker, T. F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S. K., Boschung, J., Nauels, A., Xia, Y., Bex, V., and Midley, P. M., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 2013.

Jiang, H., Liao, H., Pye, H. O. T., Wu, S., Mickley, L. J., Seinfeld, J. H., and Zhang, X. Y.: Projected effect of 2000-2050 changes in climate and emissions on aerosol levels in China and associated transboundary transport, Atmos. Chem. Phys., 13, 7937–7960, doi:10.5194/acp-13-7937-2013, 2013.

Kain, J. S.: The Kain-Fritsch convective parameterization: An update, J. Appl. Meteorol., 43, 170–181, doi:10.1175/1520-0450(2004)043<0170:TKCPAU>2.0.CO;2, 2004.

Kim, M. J., Park, R. J., Ho, C., Woo, J., Choi, K., Song, C., and Lee, J.: Future ozone and oxidants change under the RCP scenarios, Atmos. Environ., 101, 103–115, doi:10.1016/j.atmosenv.2014.11.016, 2015.

Lam, Y. F., Fu, J. S., Wu, S., and Mickley, L. J.: Impacts of future climate change and effects of biogenic emissions on surface ozone and particulate matter concentrations in the United States, Atmos. Chem. Phys., 11, 4789–4806, doi:10.5194/acp-11-4789-2011, 2011.

Lee, C., Martin, R. V., van Donkelaar, A., O'Byrne, G., Krotkov, N., Richter, A., Huey, L. G., and Holloway, J. S.: Retrieval of vertical columns of sulfur dioxide from SCIAMACHY and OMI: Air mass factor algorithm development, validation, and error analysis, J. Geophys. Res. Atmos., 114, D22303, doi:10.1029/2009JD012123, 2009.

Li, M., Zhang, Q., Kurokawa, J.-I., Woo, J.-H., He, K., Lu, Z., Ohara, T., Song, Y., Streets, D. G., Carmichael, G. R., Cheng, Y., Hong, C., Huo, H., Jiang, X., Kang, S., Liu, F., Su, H., and Zheng, B.: MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP, Atmos. Chem. Phys., 17, 935-963, doi:10.5194/acp-17-935-2017, 2017.

Lin, J.-T., McElroy, M. B., and Boersma, K. F.: Constraint of anthropogenic NOx emissions in China from different sectors: a new methodology using multiple satellite retrievals, Atmos. Chem. Phys., 10, 63–78, doi:10.5194/acp-10-63-2010, 2010.

Liu, X., Zhang, Y., Cheng, S., Xing, J., Zhang, Q., Streets, D. G., Jang, C., Wang, W., and Hao, J.: Understanding of regional air pollution over China using CMAQ, part I performance evaluation and seasonal variation, Atmos. Environ., 44, 2415–2426, doi:10.1016/j.atmosenv.2010.03.035, 2010.

Liu, X., Easter, R. C., Ghan, S. J., Zaveri, R., Rasch, P., Shi, X., Lamarque, J. F., Gettelman, A., Morrison, H., Vitt, F., Conley, A., Park, S., Neale, R., Hannay, C., Ekman, A. M. L., Hess, P., Mahowald, N., Collins, W., Iacono, M. J., Bretherton, C. S., Flanner, M. G., and Mitchell, D.: Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5, Geosci. Model Dev., 5, 709–739, doi:10.5194/gmd-5-709-2012, 2012.

Liu, X., Zhang, Y., Zhang, Q., and He, M.: Application of online-coupled WRF/Chem-MADRID in East Asia: Model evaluation and climatic effects of anthropogenic aerosols, Atmos. Environ., 124, 321–336, doi:10.1016/j.atmosenv.2015.03.052, 2016.

Mass, C., and Ovens, D.: WRF model physics: progress, problems, and perhaps some solutions, in: The 11th WRF Users' Workshop, NCAR Center Green Campus, http://www.mmm.ucar.edu/wrf/users/workshops/WS2010/presentations/session%204/4-1_WRFworkshop2010Final.pdf (last access: June 2014), 21–25 June, 2010.

Mass, C., and Ovens, D.: Fixing WRF's High Speed Wind Bias: a New Subgrid Scale Drag Parameterization and the Role of Detailed Verification, Preprints, 24th Conference on Weather and Forecasting/20th Conference on Numerical Weather Prediction, American Meteorological Society, Seattle, W.A., 9B.6. Available online at: http://ams.confex.com/ams/91Annual/webprogram/Paper180011.html, 2011.

Morrison, H., Thompson, G., and Tatarskii, V.: Impact of Cloud Microphysics on the Development of Trailing Stratiform Precipitation in a Simulated Squall Line: Comparison of One- and Two-Moment Schemes, Mon. Weather Rev., 137, 991–1007, doi:10.1175/2008MWR2556.1, 2009.

Oh, S., Park, J., Lee, S., and Suh, M.: Assessment of the RegCM4 over East Asia and future precipitation change adapted to the RCP scenarios, J. Geophys. Res. Atmos., 119, 2913–2927, doi:10.1002/2013JD020693, 2014.

Penrod, A., Zhang, Y., Wang, K., Wu, S., and Leung, L. R.: Impacts of future climate and emission changes on US air quality, Atmos. Environ., 89, 533–547, doi:10.1016/j.atmosenv.2014.01.001, 2014.

Pleim, J. E.: A combined local and nonlocal closure model for the atmospheric boundary layer. Part I: Model description and testing, J. Appl. Meteorol. Clim., 46, 1383–1395, doi:10.1175/JAM2539.1, 2007.

Pleim, J., Gilliam, A. A. R., Appel, W., and Ran, L.: Recent Advances in Modeling of the Atmospheric Boundary Layer and Land Surface in the Coupled WRF-CMAQ Model, 34th International Technical Meeting on Air Pollution Modelling and its Application, Montpellier, France, 4-8 May, 2015.

Qian, J. H., Seth, A., and Zebiak, S.: Reinitialized versus continuous simulations for regional climate downscaling, Mon. Weather Rev., 131, 2857–2874, doi:10.1175/1520-0493(2003)131<2857:RVCSFR>2.0.CO;2, 2003.

Rontu, L.: A study on parametrization of orography-related momentum fluxes in a synoptic-scale NWP model, Tellus A, 58, 69–81, doi:10.1111/j.1600-0870.2006.00162.x, 2006.

Schwede, D., Pouliot, G.A., and Pierce, T.: Changes to the biogenic emissions inventory system version 3 (BEIS3). In: Proceedings of the 4th CMAS Models-3 Users' Conference, Chapel Hill, NC, 26-28, September, 2005.

Stevenson, D. S., Young, P. J., Naik, V., Lamarque, J. F., Shindell, D. T., Voulgarakis, A., Skeie, R. B., Dalsoren, S. B., Myhre, G., Berntsen, T. K., Folberth, G. A., Rumbold, S. T., Collins, W. J., MacKenzie, I. A., Doherty, R. M., Zeng, G., van Noije, T. P. C., Strunk, A., Bergmann, D., Cameron-Smith, P., Plummer, D. A., Strode, S. A., Horowitz, L., Lee, Y. H., Szopa, S., Sudo, K., Nagashima, T., Josse, B., Cionni, I., Righi, M., Eyring, V., Conley, A., Bowman, K. W., Wild, O., and Archibald, A.: Tropospheric ozone changes, radiative forcing and attribution to emissions in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), Atmos. Chem. Phys., 13, 3063–3085, doi:10.5194/acp-13-3063-2013, 2013.

Streets, D. G., Yarber, K. F., Woo, J. H., and Carmichael, G. R.: Biomass burning in Asia: Annual and seasonal estimates and atmospheric emissions, Global Biogeochem. Cy., 17, 1099, doi:10.1029/2003GB002040, 2003.

Sun, J., Fu, J. S., Huang, K., and Gao, Y.: Estimation of future PM2.5- and ozone-related mortality over the continental United States in a changing climate: An application of high-resolution dynamical downscaling technique, J. Air Waste Manage., 65, 611–623, doi:10.1080/10962247.2015.1033068, 2015.

Tang, Y., Lee, P., Tsidulko, M., Huang, H., McQueen, J. T., DiMego, G. J., Emmons, L. K., Pierce, R. B., Thompson, A. M., Lin, H., Kang, D., Tong, D., Yu, S., Mathur, R., Pleim, J. E., Otte, T. L., Pouliot, G., Young, J. O., Schere, K. L., Davidson, P. M., and Stajner, I.: The impact of chemical lateral boundary conditions on CMAQ predictions of tropospheric ozone over the continental United States, Environ. Fluid Mech., 9, 43–58, doi:10.1007/s10652-008-9092-5, 2009.

Thomson, A. M., Calvin, K. V., Smith, S. J., Kyle, G. P., Volke, A., Patel, P., Delgado-Arias, S., Bond-Lamberty, B., Wise, M. A., Clarke, L. E., and Edmonds, J. A.: RCP4.5: a pathway for stabilization of radiative forcing by 2100, Climatic Change, 109, 77 – 94, doi:10.1007/s10584-011-0151-4, 2011.

Tong, D. Q., Bowker, G. E., He, S., Byun, D. W., Mathur, R., and Gillette, D. A.: Development of a windblown dust emission model FENGSHAA description and initial application in the United States, in review, 2017.

von Schneidemesser, E., Monks, P. S., Allan, J. D., Bruhwiler, L., Forster, P., Fowler, D., Lauer, A., Morgan, W. T., Paasonen, P., Righi, M., Sindelarova, K., and Sutton, M. A.: Chemistry and the Linkages between Air Quality and Climate Change, Chem. Rev., 115, 3856–3897, doi:10.1021/acs.chemrev.5b00089, 2015.

Vukovich, J., and Pierce, T.: The implementation of BEIS3 within the SMOKE modeling framework. In: Proceedings of the 11th International Emissions Inventory Conference, Atlanta, Georgia. Available at: www.epa.gov/ttn/chief/conference/ei11/modeling/vukovich.pdf, 15-18, April, 2002.

Wang, J., Wang, S., Jiang, J., Ding, A., Zheng, M., Zhao, B., Wong, D. C., Zhou, W., Zheng, G., Wang, L., Pleim, J. E., and Hao, J.: Impact of aerosol-meteorology interactions on fine particle pollution during China's severe haze episode in January 2013, Environ. Res. Lett., 9, 0940029, doi:10.1088/1748-9326/9/9/094002, 2014.

Wang, J., and Kotamarthi, V. R.: High-resolution dynamically downscaled projections of precipitation in the mid and late 21st century over North America, Earth’s Future, 3, 268–288, doi:10.1002/2015EF000304, 2015.

Wang, K., Zhang, Y., Jang, C., Phillips, S., and Wang, B.: Modeling intercontinental air pollution transport over the trans-Pacific region in 2001 using the Community Multiscale Air Quality modeling system, J. Geophys. Res. Atmos., 114, D04307, doi:10.1029/2008JD010807, 2009.

Wang, K., Zhang, Y., Nenes, A., and Fountoukis, C.: Implementation of dust emission and chemistry into the Community Multiscale Air Quality modeling system and initial application to an Asian dust storm episode, Atmos. Chem. Phys., 12, 10209–10237, doi:10.5194/acp-12-10209-2012, 2012.

Wang, Y., Zhang, Y., Hao, J., and Luo, M.: Seasonal and spatial variability of surface ozone over China: contributions from background and domestic pollution, Atmos. Chem. Phys., 11, 3511–3525, doi:10.5194/acp-11-3511-2011, 2011.

Wang, Y., Shen, L., Wu, S., Mickley, L., He, J., and Hao, J.: Sensitivity of surface ozone over China to 2000-2050 global changes of climate and emissions, Atmos. Environ., 75, 374–382, doi:10.1016/j.atmosenv.2013.04.045, 2013.

Whitten, G. Z., Heo, G., Kimura, Y., McDonald-Buller, E., Allen, D. T., Carter, W. P. L., and Yarwood, G.: A new condensed toluene mechanism for Carbon Bond CB05-TU, Atmos. Environ., 44, 5346–5355, doi:10.1016/j.atmosenv.2009.12.029, 2010.

Wong, D. C., Pleim, J., Mathur, R., Binkowski, F., Otte, T., Gilliam, R., Pouliot, G., Xiu, A., Young, J. O., and Kang, D.: WRF-CMAQ two-way coupled system with aerosol feedback: software development and preliminary results, Geosci. Model Dev., 5, 299–312, doi:10.5194/gmd-5-299-2012, 2012.

Xing, J., Wang, J., Mathur, R., Pleim, J., Wang, S., Hogrefe, C., Gan, C., Wong, D. C., and Hao, J.: Unexpected Benefits of Reducing Aerosol Cooling Effects, Environ. Sci. Technol., 50, 7527–7534, doi:10.1021/acs.est.6b00767, 2016.

Xiu, A., and Pleim, J. E.: Development of a land surface model. Part I: Application in a mesoscale meteorological model, J. Appl. Meteorol., 40, 192–209, doi:10.1175/1520-0450(2001)040<0192:doalsm>2.0.co;2, 2001.

Xu, Y., and Xu, C.: Preliminary Assessment of Simulations of Climate Changes over China by CMIP5 Multi-Models, Atmospheric and Oceanic Science Letters, 5:6, 489-494, 2012.

Xu, Z., and Yang, Z.: An Improved Dynamical Downscaling Method with GCM Bias Corrections and Its Validation with 30 Years of Climate Simulations, J. Climate, 25, 6271–6286, doi:10.1175/JCLI-D-12-00005.1, 2012.

Xu, Z., and Yang, Z.: A new dynamical downscaling approach with GCM bias corrections and spectral nudging, J. Geophys. Res. Atmos., 120, 3063–3084, doi:10.1002/2014JD022958, 2015.

Yarwood, G., Rao, S., Yocke, M., and Whitten, G. Z.: Final Report –Updates to the Carbon Bond Chemical Mechanism: CB05, Rep.RT-04-00675, 246 pp., Yocke and Co., Novato, Calif., 2005.

Yahya, K., Wang, K., Campbell, P., Glotfelty, T., He, J., and Zhang, Y.: Decadal evaluation of regional climate, air quality, and their interactions over the continental US and their interactions using WRF/Chem version 3.6.1, Geosci. Model Dev., 9, 671–695, doi:10.5194/gmd-9-671-2016, 2016.

Yahya, K., Wang, K., Campbell, P., Chen, Y., Glotfelty, T., He, J., Pirhalla, M., and Zhang, Y.: Decadal Application of WRF/Chem for Regional Air Quality and Climate Modeling over the U.S. under the Representative Concentration Pathways Scenarios. Part 1: Model Evaluation and Impact of Downscaling, Atmos. Environ., 152, 562–583, doi:10.1016/j.atmosenv.2016.12.029, 2017a.

Yahya, K., Campbell, P., and Zhang, Y.: Decadal Application for Regional Air Quality and Climate Modeling over the U.S under the Representative Concentration Pathways Scenarios. Part 2: Current vs. Future WRF and WRF/Chem simulations, Atmos. Environ., 152, 584–604, doi:10.1016/j.atmosenv.2016.12.028, 2017b.

Yu, S., Mathur, R., Pleim, J., Wong, D., Gilliam, R., Alapaty, K., Zhao, C., and Liu, X.: Aerosol indirect effect on the grid-scale clouds in the two-way coupled WRF-CMAQ: model description, development, evaluation and regional analysis, Atmos. Chem. Phys., 14, 11247–11285, doi:10.5194/acp-14-11247-2014, 2014.

Zhang, Y., Liu, P., Pun, B., and Seigneur, C.: A comprehensive performance evaluation of MM5-CMAQ for the Summer 1999 Southern Oxidants Study episode - Part I: Evaluation protocols, databases, and meteorological predictions, Atmos. Environ., 40, 4825–4838, doi:10.1016/j.atmosenv.2005.12.043, 2006.

Zhang, Y.: Online-coupled meteorology and chemistry models: history, current status, and outlook, Atmos. Chem. Phys., 8, 2895–2932, doi:10.5194/acp-8-2895-2008, 2008.

Zhang, Y., Vijayaraghavan, K., Wen, X., Snell, H. E., and Jacobson, M. Z.: Probing into regional ozone and particulate matter pollution in the United States: 1. A 1 year CMAQ simulation and evaluation using surface and satellite data, J. Geophys. Res. Atmos., 114, D22304, doi:10.1029/2009JD011898, 2009a.

Zhang, Y., Wen, X., Wang, K., Vijayaraghavan, K., and Jacobson, M. Z.: Probing into regional O-3 and particulate matter pollution in the United States: 2. An examination of formation mechanisms through a process analysis technique and sensitivity study, J. Geophys. Res. Atmos., 114, D22305, doi:10.1029/2009JD011900, 2009b.

Zhang, Y., Zhang, X., Wang, L., Zhang, Q., Duan, F., and He, K.: Application of WRF/Chem over East Asia: Part I. Model evaluation and intercomparison with MM5/CMAQ, Atmos. Environ., 124, 285–300, doi:10.1016/j.atmosenv.2015.07.022, 2016a.

Zhang, Y., Zhang, X., Wang, K., Zhang, Q., Duan, F., and He, K.: Application of WRF/Chem over East Asia: Part II. Model improvement and sensitivity simulations, Atmos. Environ., 124, 301–320, doi:10.1016/j.atmosenv.2015.07.023, 2016b.

Zhang, Y., Hong, C., Yahya, K., and Zhang, Q.: Multi-Year Application and Evaluation of WRF/Chem-MADRID for Real-Time Air Quality Forecasting over Southeastern United States, Atmos. Environ., 138, 162-182, doi:10.1016/j.atmosenv.2016.05.006, 2016c.

Zhao, B., Wang, S., Dong, X., Wang, J., Duan, L., Fu, X., Hao, J., and Fu, J.: Environmental effects of the recent emission changes in China: implications for particulate matter pollution and soil acidification, Environ. Res. Lett., 8, doi:10.1088/1748-9326/8/2/024031, 2013.

Zheng, B., Zhang, Q., Zhang, Y., He, K. B., Wang, K., Zheng, G. J., Duan, F. K., Ma, Y. L., and Kimoto, T.: Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China, Atmos. Chem. Phys., 15, 2031–2049, doi:10.5194/acp-15-2031-2015, 2015.

Table 1. Model configurations and set-up for the climatological application.

Model Attribute	Configuration
Model	Two-way Coupled WRF3.4 and CMAQv5.0.2 (Wong et al., 2012)
Domain and Resolutions	36 km × 36 km over East Asia; 23 sigma layers from surface to 100 mb
Simulation Period	Current years (2006-2010)
Meteorological and Chemical ICs/BCs	The NCSU's CESM/CAM5 v1.2.2 (Gantt et al., 2014; He and Zhang, 2014; He at al., 2015a, b; Glotfelty et al., 2017a, b; Glotfelty and Zhang, 2017); meteorological ICs/BCs are bias-corrected with NCEP FNL data based on Xu and Yang (2012)
Anthropogenic Emissions	MIX Asian 2008 emission inventory (a mosaic Asian anthropogenic emission inventory for the MICS-Asia and the HTAP projects, http://www.meicmodel.org; Li et al., 2017) for current years
Biogenic Emissions	BEISv3.1.4 (Vukovich and Pierce, 2002; Schwede et al., 2005)
Dust Emissions	The physically based dust emission algorithm, FENGSHA (Tong et al., 2017)
Radiation	RRTMG shortwave (SW) and longwave (LW) (Iacono et al., 2008)
PBL	ACM2 PBL scheme (Pleim, 2007)
Land Surface	Pleim–Xiu land surface model (Xiu and Pleim, 2001)
Surface Layer	Pleim–Xiu surface layer scheme
Land Use Category	The USGS 24-category land use data
Microphysics	Morrison double-moment (Morrison et al., 2009)
Cumulus Parameterization	Kain–Fritsch cumulus scheme (Kain, 2004)
Gas-Phase Chemistry	CB05 gas-phase mechanism with active chlorine chemistry and updated toluene mechanism (Yarwood et al., 2005; Whitten et al., 2010)
Aerosol Module	AERO6 (Appel et al., 2013)

Table 2. Simulation design.

Run index	Simulated period	Model configuration	Purpose
Climatological applications (2006-2010) with CESM ICs/BCs			
CESM_BASE (Production runs)	2006-2010	See Table 1, with CESM ICs/BCs and improvements made in NCEP_BASE	Evaluate model performance for climatological applications
CESM_BASE_Sens	Jan., 2008; Jul., 2007	Same as CESM_BASE but without aerosol feedback	Access the aerosol direct effects on regional climate and air quality
Air quality applications in 2013 with NCEP ICs/BCs			
NCEP_BASE	2013 (Jan., Apr., Jul.)	With NCEP ICs/BCs and several improvements (refer to Section 2.1)	Evaluate model performance for air quality applications
NCEP_BASE_WoImp	2013 (Jan., Apr., Jul.)	Without updated improvements	Compared with NCEP_BASE, to support the improvements

Table 3. Model performance statistics for the climatological application (2006-2010, CESM_BASE).

Variable	Network	MeanObs	MeanSim	R	MB	NMB (%)	MAGE	RMSE
T2 (℃)	NCDC	14.2	13.6	0.97	-0.6	-4.6	2.4	3.2
RH2 (%)	NCDC	69.0	69.8	0.72	0.8	1.1	9.7	12.6
WS10 ($m\ s^{-1}$)	NCDC	2.7	3.3	0.47	0.6	22.2	1.2	1.6
WDR10 (degree)	NCDC	210.7	186.8	0.35	-23.8	-11.3	37.2	60.2
Precip ($mm\ day^{-1}$)	NCDC	2.5	3.2	0.52	0.7	27.4	1.4	2.1
Precip ($mm\ day^{-1}$)	GPCP	3.0	3.0	0.80	0.0	-0.9	0.8	1.3
SWDOWN ($W\ m^{-2}$)	CERES	184.5	210.5	0.90	26.0	14.1	26.0	29.4
LWDOWN ($W\ m^{-2}$)	CERES	330.1	320.8	0.99	-9.4	-2.8	10.0	12.9
GSW ($W\ m^{-2}$)	CERES	157.0	171.9	0.91	14.8	9.4	17.3	20.9
OLR ($W\ m^{-2}$)	CERES	235.0	244.0	0.89	9.0	3.8	10.3	13.3
SWCF ($W\ m^{-2}$)	CERES	-50.5	-36.8	0.74	13.6	-27.0	14.5	18.6
LWCF ($W\ m^{-2}$)	CERES	29.9	17.3	0.20	-12.7	-42.3	12.7	15.1
CF (%)	MODIS	64.7	45.0	0.11	-19.7	-30.5	20.9	25.6
Column CO (10^{15} molec. cm^{-2})	MOPITT	2075.3	1832.2	0.83	-243.1	-11.7	324.2	376.2
Column NO_2 (10^{15} molec. cm^{-2})	OMI	1.5	1.8	0.91	0.3	18.3	0.6	1.2
Column SO_2 (10^{15} molec. cm^{-2})	SCIA	5.5	5.9	0.82	0.4	7.5	3.6	6.1
Column HCHO (10^{15} molec. cm^{-2})	OMI	5.9	5.7	0.87	-0.2	-4.0	1.3	1.8
TOR (DU)	OMI	31.2	36.3	0.92	5.1	16.4	5.3	5.9
AOD	MODIS	0.3	0.2	0.82	-0.1	-36.3	0.1	0.1

[1] Mean Obs: Mean observed data; Mean Sim: Mean simulated data; R: correlation coefficient; MB: mean bias; NMB: normalized mean biases; MAGE: mean absolute gross error; RMSE: root mean square error.

Table 4. Model performance statistics for the air quality application: chemical variables (2013, NCEP_BASE).

Variable	Network	January				April				July			
		R	MB	NMB (%)	RMSE	R	MB	NMB (%)	RMSE	R	MB	NMB (%)	RMSE
CO (mg m^{-3})	CNEMC	0.5	-0.8	-34.9	1.8	0.2	-0.5	-48.1	1.0	0.2	-0.3	-33.1	0.8
SO_2 (μg m^{-3})	CNEMC	0.3	-0.3	-0.4	102.4	0.2	2.8	9.2	40.5	0.1	16.3	89.1	47.6
NO_2 (μg m^{-3})	CNEMC	0.4	-1.4	-2.0	44.1	0.4	-0.2	-0.5	35.3	0.3	12.8	43.6	37.9
O_3 (μg m^{-3})	CNEMC	N/A	N/A	N/A	N/A	0.3	-3.1	-4.4	59.7	0.5	11.2	18.2	54.7
$PM_{2.5}$ (μg m^{-3})	CNEMC	0.5	8.9	6.4	112.7	0.4	-1.6	-2.8	47.4	0.4	11.5	28.6	48.5
PM_{10} (μg m^{-3})	CNEMC	0.5	-27.9	-13.8	137.1	0.3	-28.4	-24.9	99.9	0.4	-5.6	-7.6	68.3
Column CO (10^{15} molec. cm^{-2})	MOPITT	0.8	-304.6	-13.7	537.4	N/A	N/A	N/A	N/A	0.3	-30.2	-1.9	540.6
Column NO_2 (10^{15} molec. cm^{-2})	OMI	0.9	-0.4	-13.8	3.4	0.9	-0.1	-3.2	1.6	0.8	0.2	16.1	1.4
Column SO_2 (10^{15} molec. cm^{-2})	OMI	0.7	2.2	29.2	13.9	0.6	-1.7	-26.8	5.7	0.6	-3.8	-66.4	5.2
Column HCHO (10^{15} molec. cm^{-2})	OMI	0.7	-1.7	-28.5	2.8	0.8	-0.5	-8.9	2.5	0.7	1.6	28.1	3.2
TOR (DU)	OMI	0.4	5.9	22.3	9.3	0.3	4.1	12.1	10.3	0.8	5.7	16.7	7.7
AOD	MODIS	0.6	0.0	-6.4	0.2	0.5	-0.1	-30.3	0.2	0.5	-0.1	-31.5	0.2

[1] R: correlation coefficient; MB: mean bias; NMB: normalized mean biases; RMSE: root mean square error; N/A: Data not available.

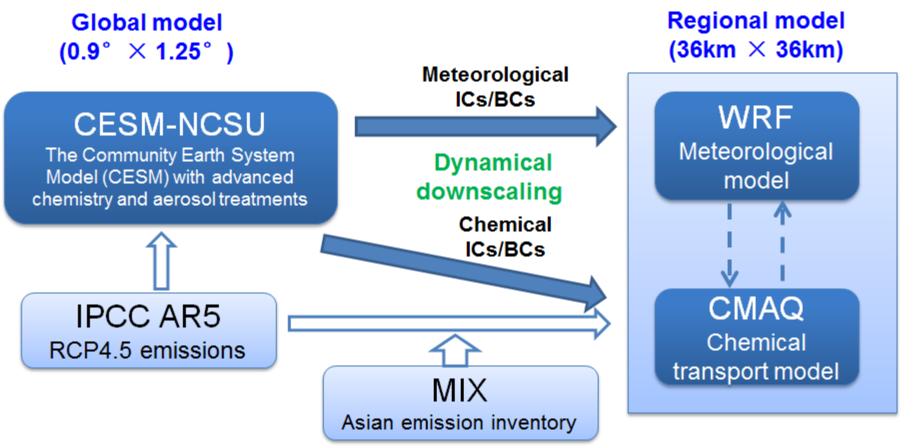


Figure 1. Modeling system in this study. The two-way coupled Weather Research and Forecasting - Community Multiscale Air Quality (WRF-CMAQ) model, which takes into account the air quality and climate interactions, is driven by the Community Earth System Model with advanced chemistry and aerosol treatments (CESM-NCSU) for high-resolution regional simulations. Both meteorological downscaling and chemical composition downscaling from the CESM-NCSU are applied to provide meteorological and chemical boundary conditions (BCs)/initial conditions (ICs) for regional WRF-CMAQ simulations.

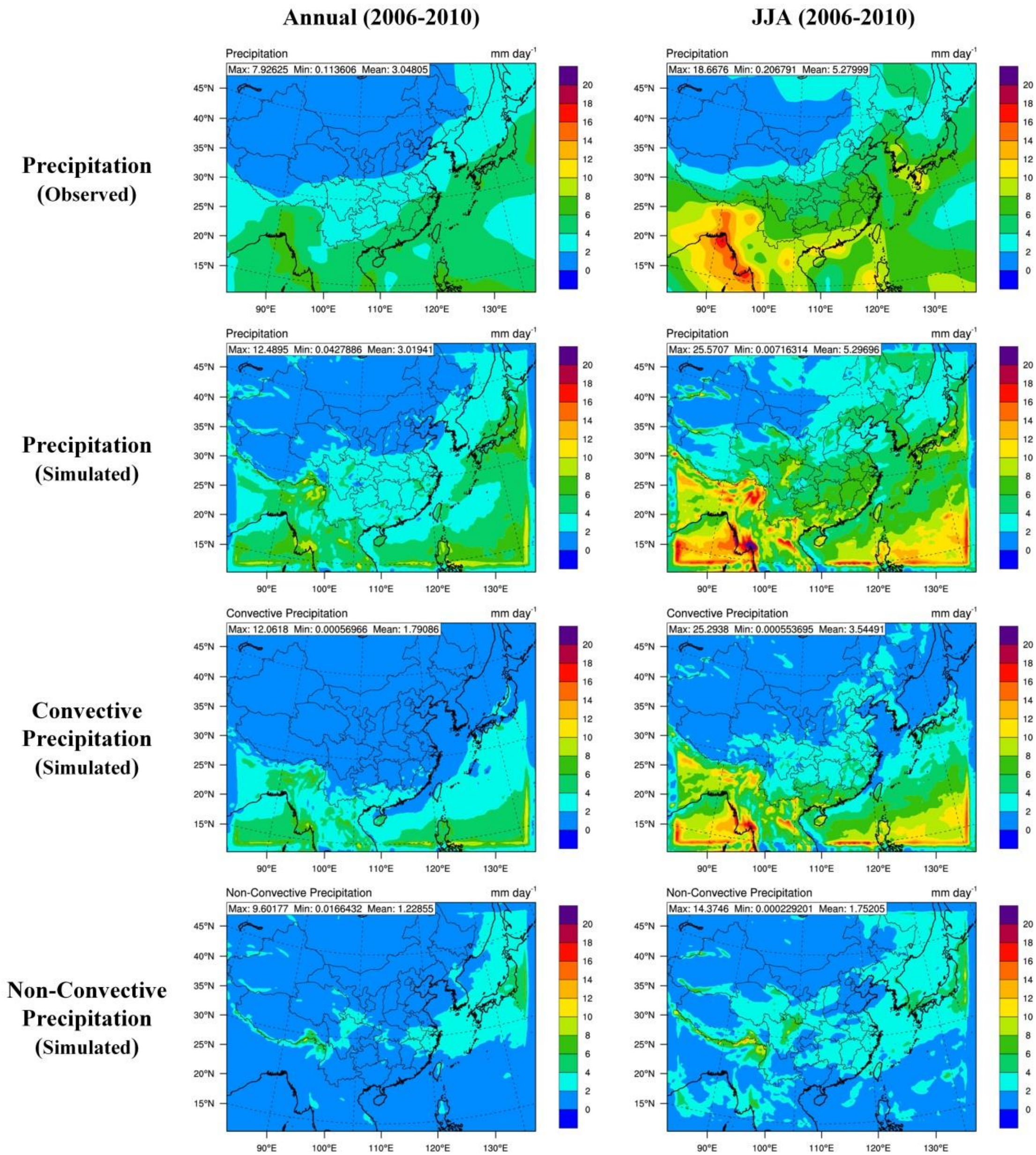


Figure 2. Spatial distribution of satellite-derived precipitation from GPCP, simulated precipitation, convective precipitation and non-convective precipitation under the climatological application during 2006-2010.

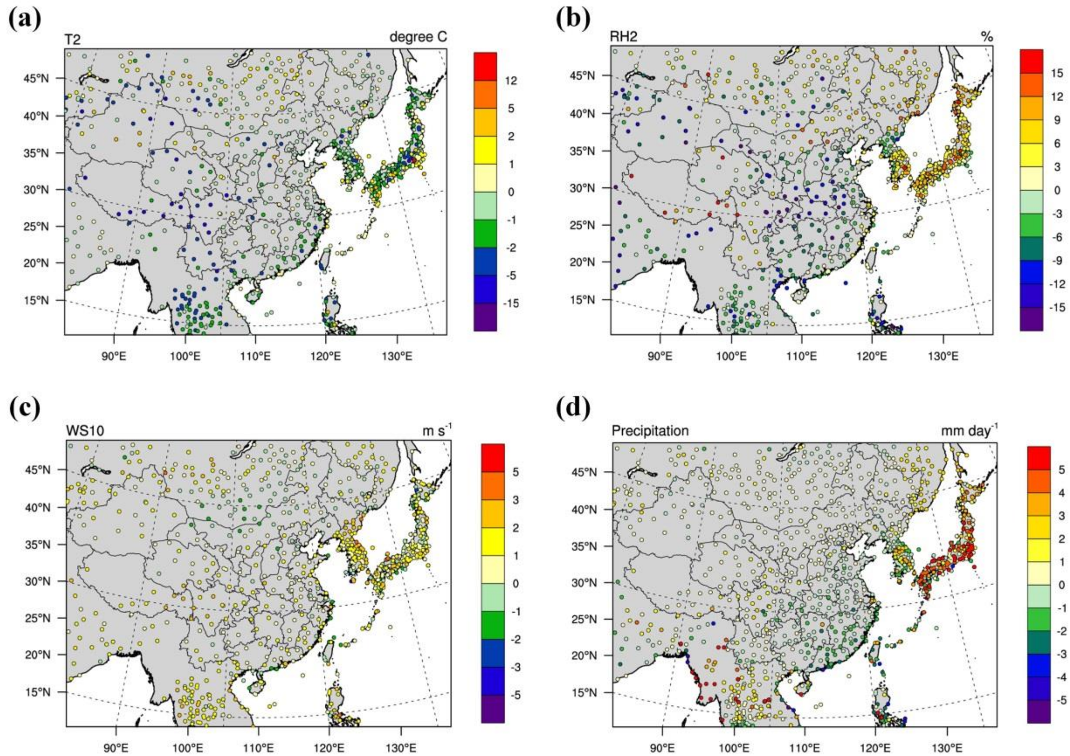


Figure 3. Spatial distribution of MBs for (a) 2-m temperature (T2), (b) 2-m relative humidity (RH2), (c) 10-m wind speed (WS10), and (d) precipitation from NCDC under the climatological application during 2006-2010. Each marker represents the MB of each variable at each observational site.

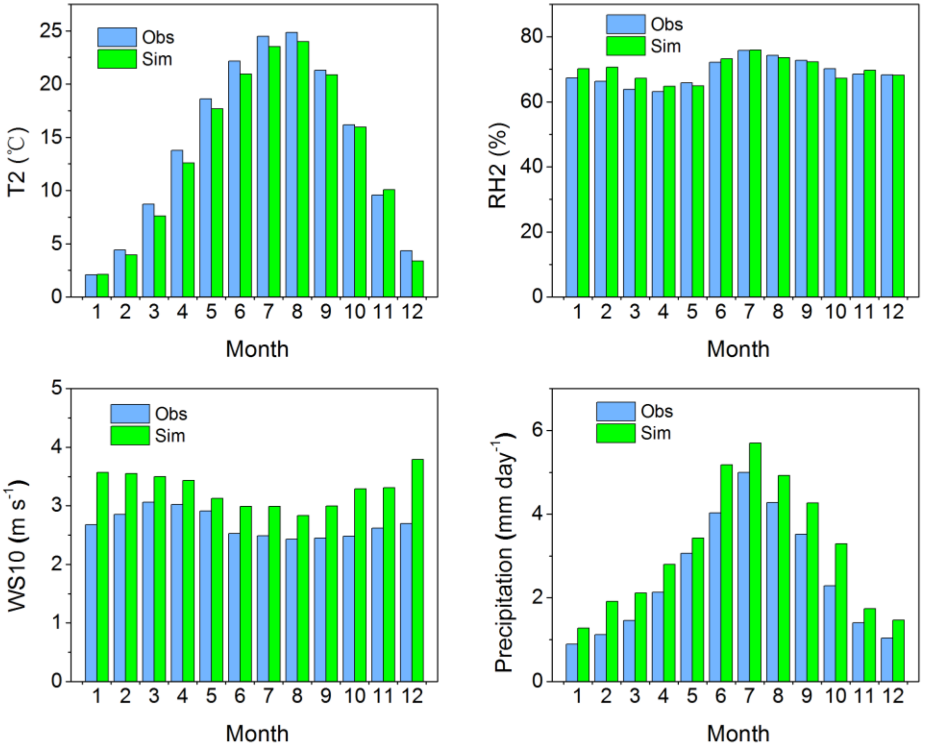


Figure 4. 5-year averaged monthly observations (blue) vs. simulations (green) for (a) 2-m temperature (T2), (b) 2-m relative humidity (RH2), (c) 10-m wind speed (WS10), and (d) precipitation under the climatological application during 2006-2010.

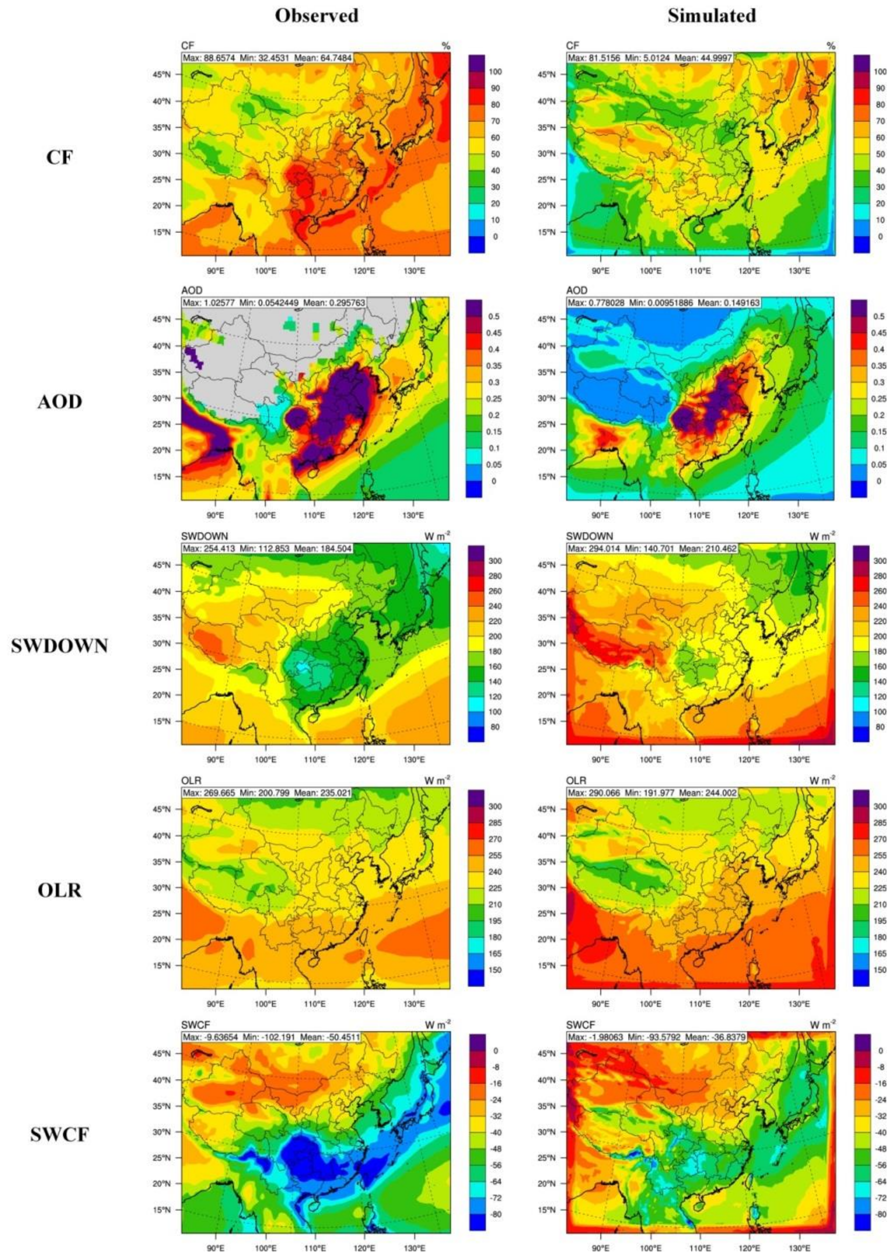


Figure 5. Spatial distribution of satellite-derived and simulated CF, AOD, SWDOWN, OLR and SWCF under the climatological application during 2006-2010.

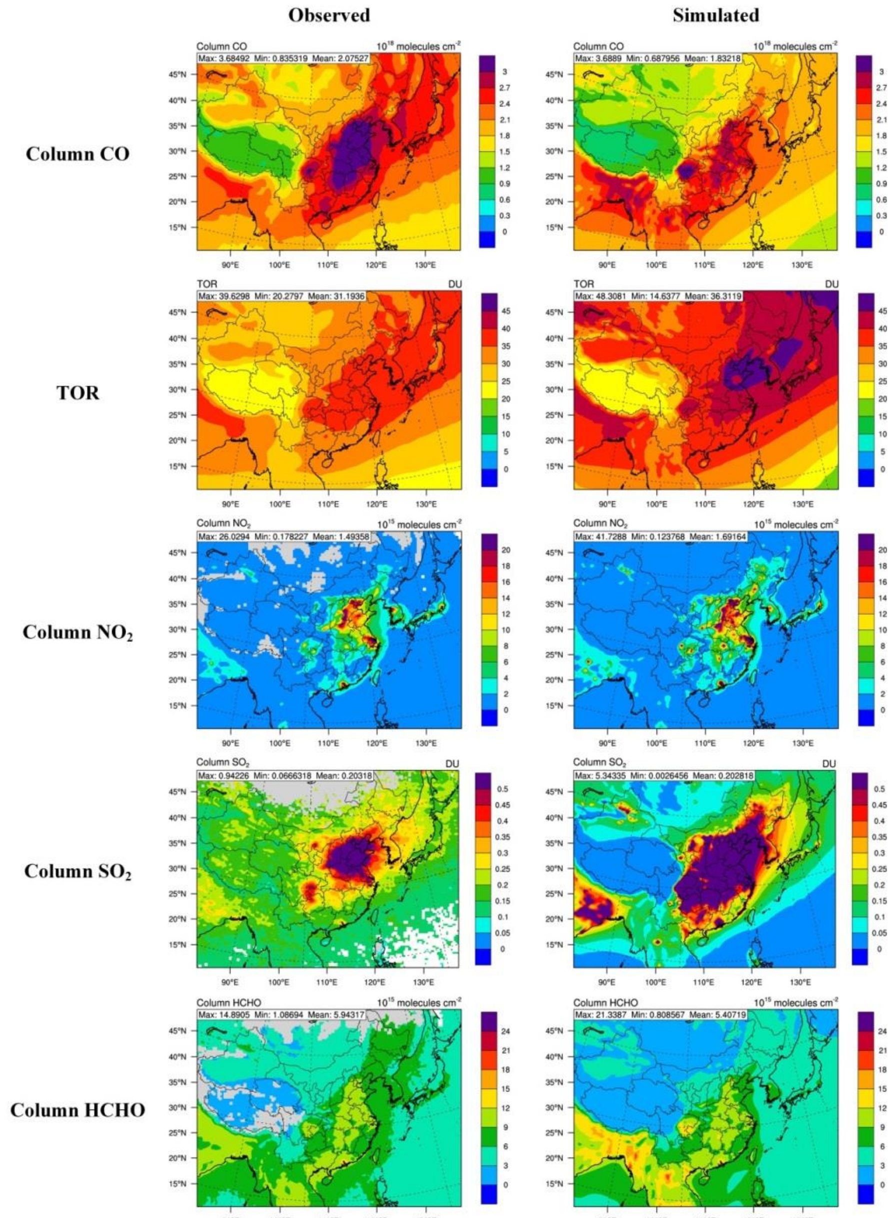


Figure 6. Spatial distribution of satellite-derived and simulated column CO, TOR, column NO_2, column SO_2, and column HCHO under the climatological application during 2006-2010.

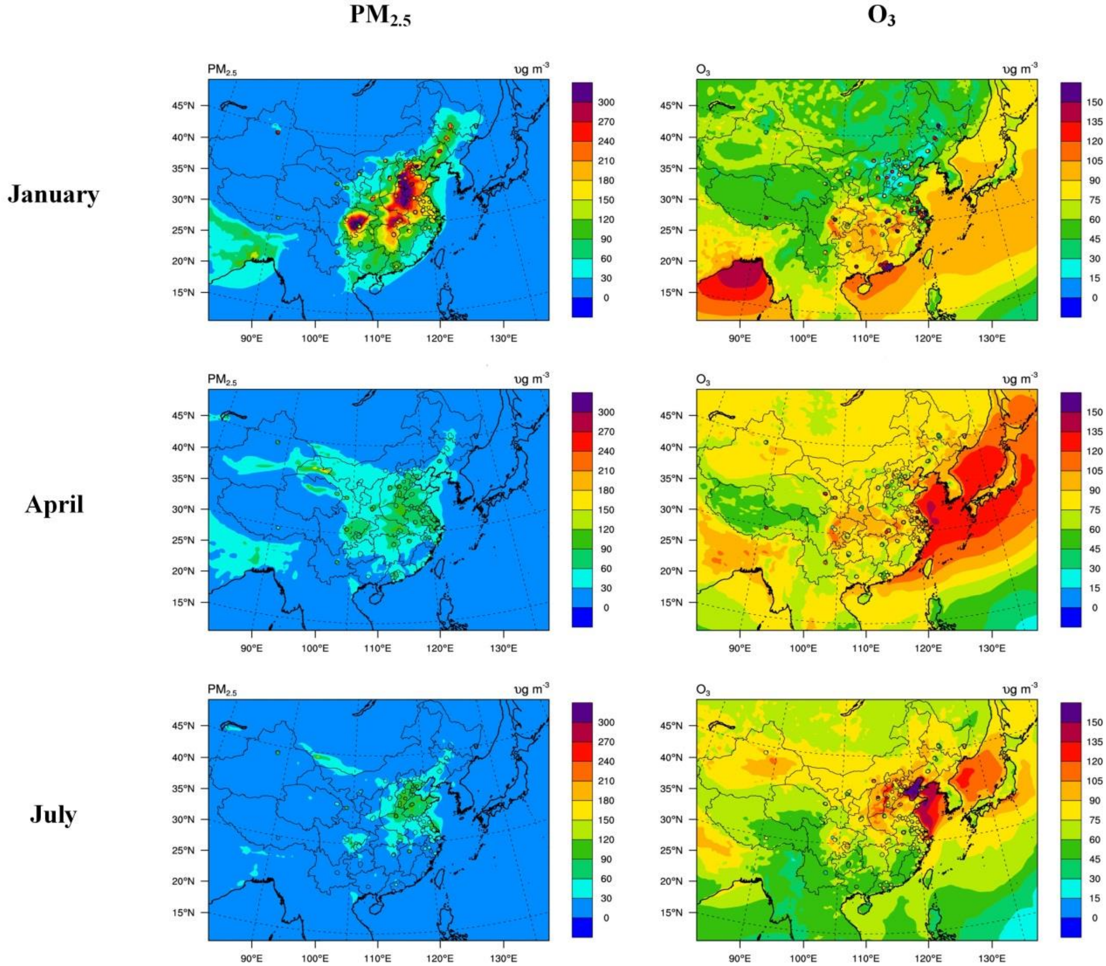


Figure 7. Spatial distribution of observed vs. simulated $PM_{2.5}$ and O_3 concentration during January, April and July, 2013, under the short-term air quality application. The background plots represent the simulated data, whereas observations are represented by the markers. Note that there were some errors in O_3 observational data in January 2013.

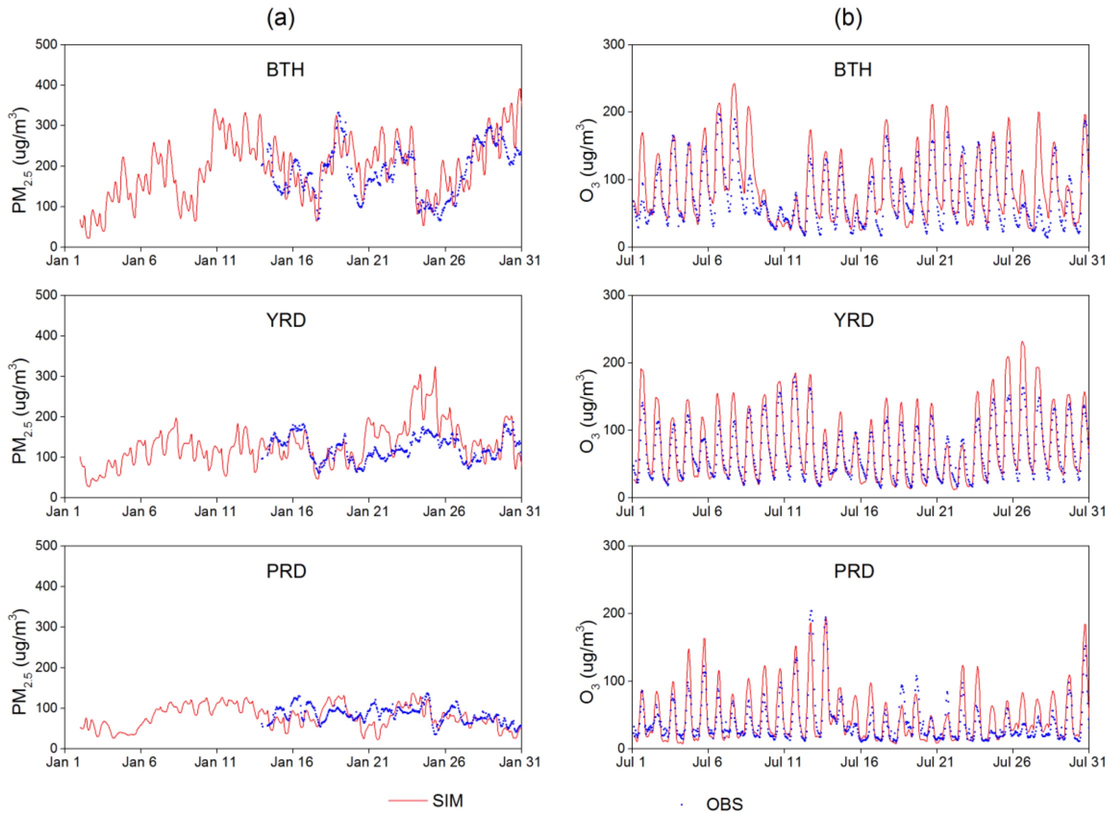


Figure 8. Time series of hourly concentrations of (a) $PM_{2.5}$ in January and (b) O_3 in July 2013, under the short-term air quality application over three key regions in China: the Beijing-Tianjin-Hebei area (BTH), the Yangtze River Delta (YRD), and the Pearl River Delta (PRD). The hourly concentrations in each region were derived by averaging all monitoring stations located in the region.

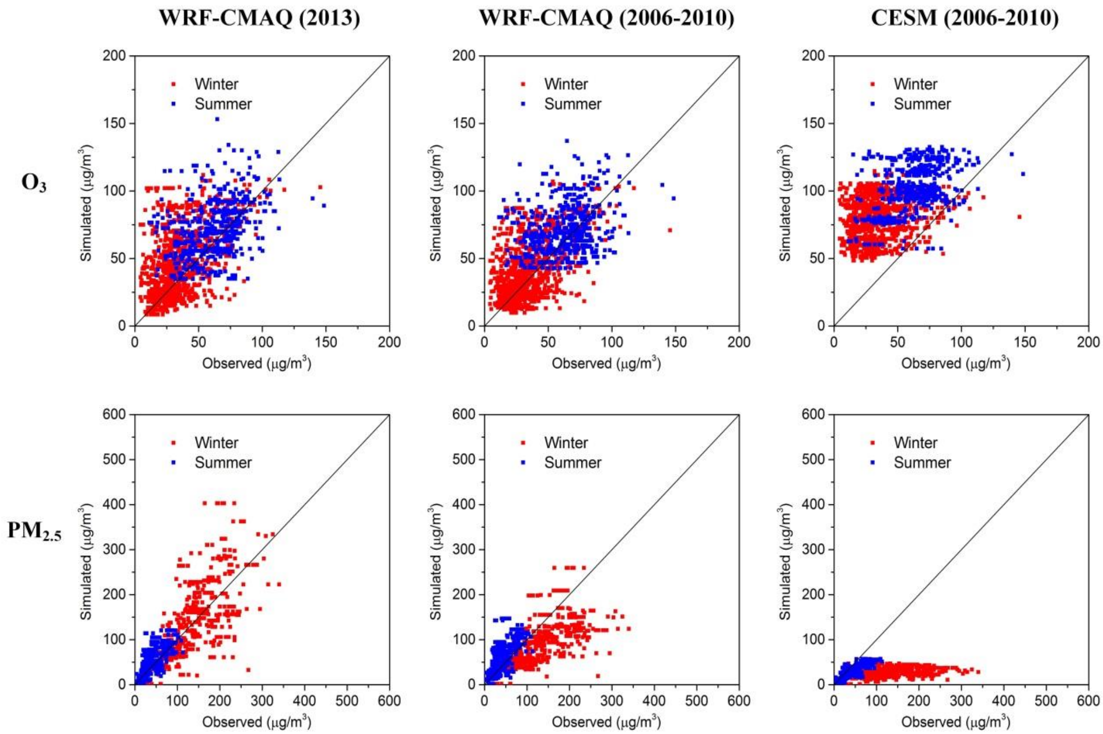


Figure 9. Scatter plots of simulated (by WRF-CMAQ (left and middle) and CESM (right)) and observed $PM_{2.5}$ (bottom) and O_3 (top) in winter (red) and summer (blue). Each scatter represents the value at each observational site. The years of observational data, WRF-CMAQ (2013), WRF-CMAQ (2006-2010), and CESM simulations were 2013, 2013, 2006-2010, and 2006-2010, respectively. Note that for O_3 observed data in winter, observational data in year 2014 were used because of some errors in the data in year 2013.

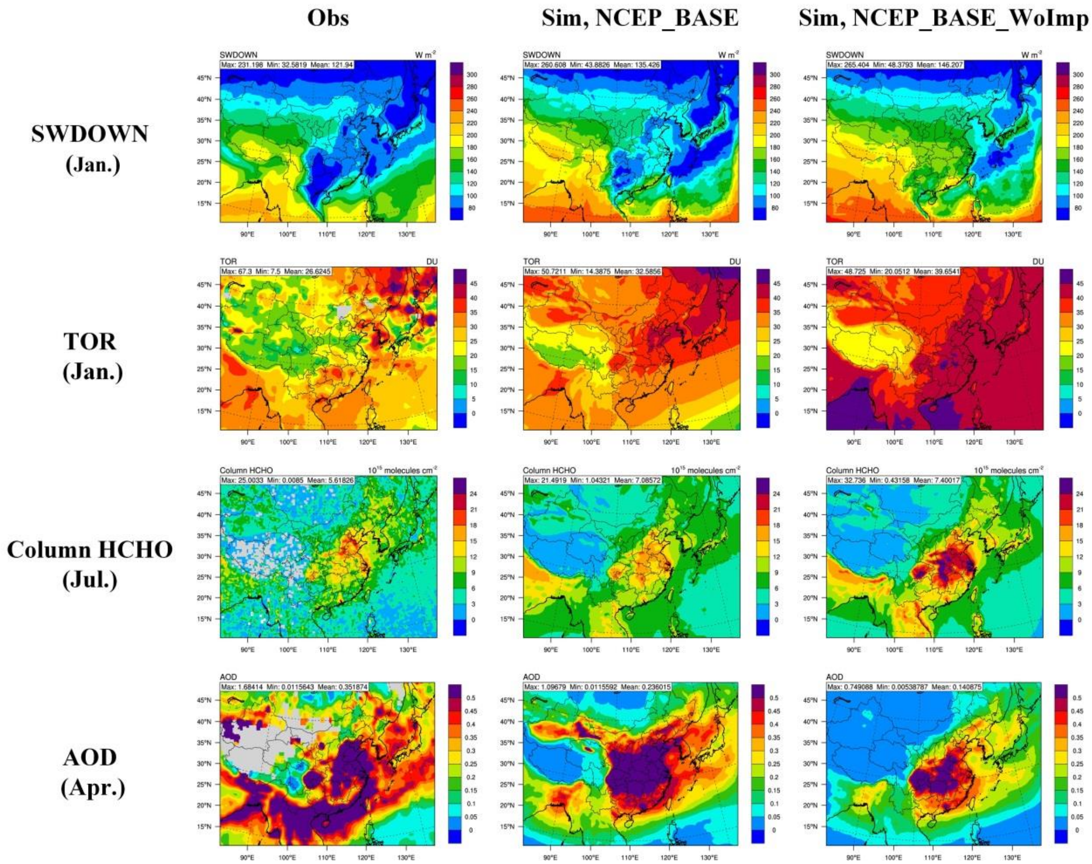
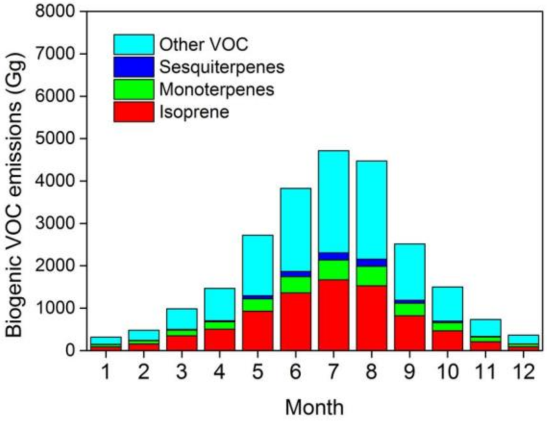
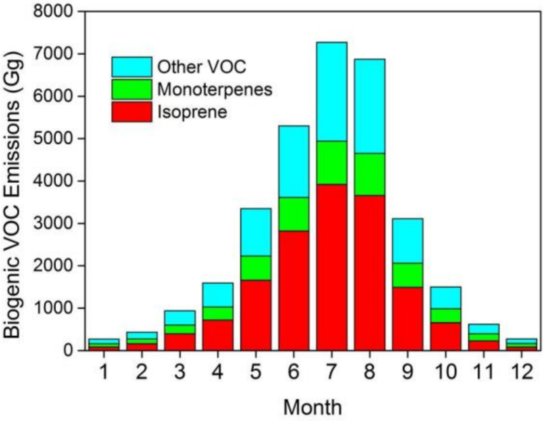


Figure 10. Comparison of spatial distributions of SWDOWN in January, TOR in January, column HCHO in July, and AOD in April 2013 from: (a) satellite observations, (b) baseline simulation (NCEP_BASE: with aerosol feedbacks, using CESM-derived BCs, BEIS3 emissions, and revised dust scheme), and (c) sensitivity simulation (NCEP_BASE_WoImp: without aerosol feedbacks, using fixed BCs, MEGAN emissions, and default dust scheme).



(a)



(b)

Figure 11. Biogenic VOC emissions over China in 2013 estimated by (a) BEIS and (b) MEGAN.

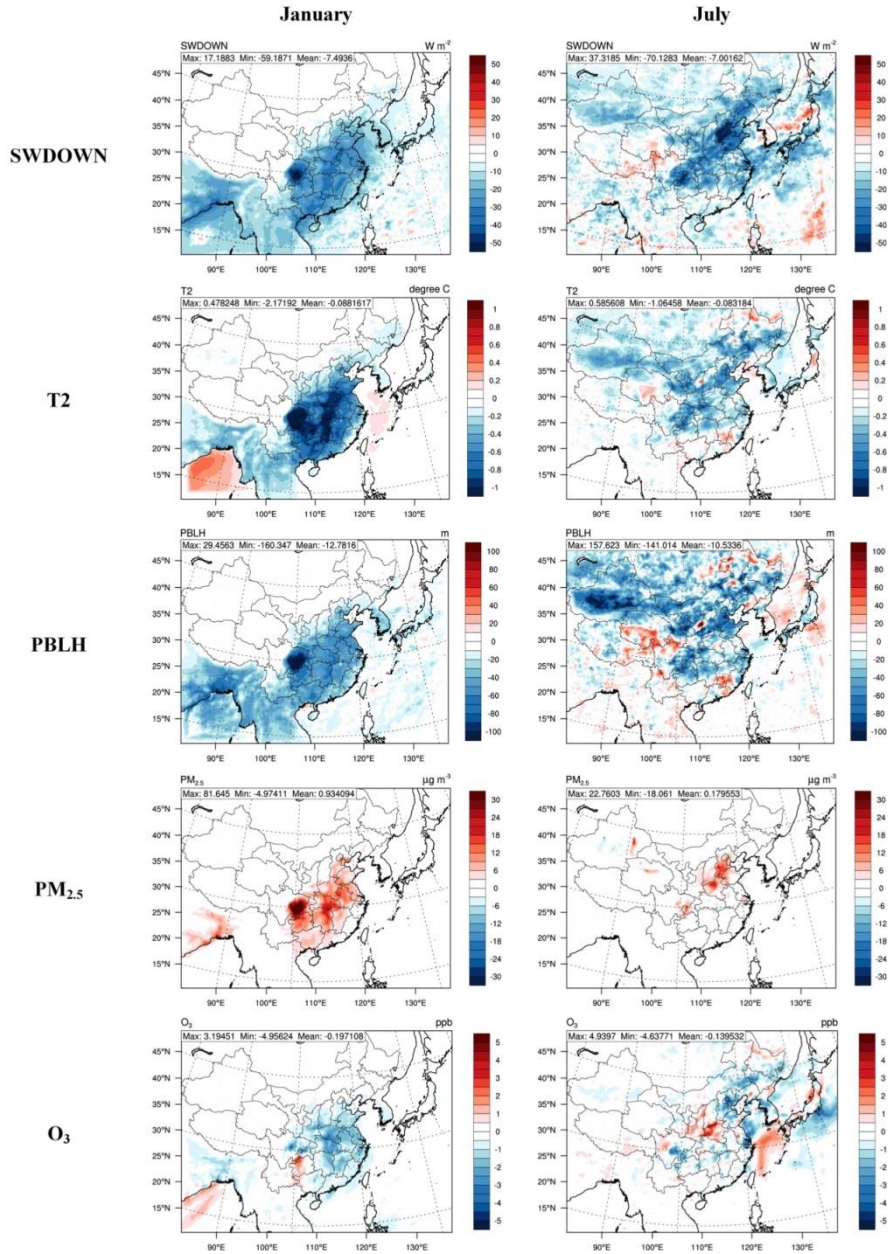


Figure 12. Effects of aerosol feedbacks on net shortwave flux at the surface (SWDOWN), T2, planetary boundary layer height (PBLH), surface $PM_{2.5}$ and O_3 concentrations in January 2008 and in July 2007. The results are from the differences between the feedback and no feedback configurations (i.e., CESM_BASE: with feedback and CESM_BASE_Sens: without feedback) of the two-way coupled WRF-CMAQ simulations.

Supplement

Configuration used in CESM-NCSU simulations

Table S1 summarizes The CESM-NCSU configurations for simulations under the RCP4.5 scenario. More detailed descriptions can be found in He and Zhang (2014) and Glotfelty et al. (2017a, b).

Table S1. The CESM-NCSU configurations for simulations under the RCP4.5 scenario.

Attribute or Process	Configuration
Simulation Time Period	Current decade (2001-2010) and future decade (2046-2055)
Horizontal Resolution	$0.9^\circ \times 1.25^\circ$, 192 (latitudes) $\times$ 288 (longitudes)
Vertical Resolution	30 layers from 1000 mb to 3 mb
Deep Convection	Zhang and McFarlane (1995); Neale et al. (2008)
Shallow Convection	Park and Bretherton (2009)
Cloud Microphysics	Morrison and Gettelman (2008)
Planetary Boundary Layer	Bretherton and Park (2009)
Short and Long-wave Radiation	RRTMG (Iacono et al., 2003, 2008)
Gas-phase Chemistry	CB05GE (Karamchandani et al., 2012)
Aqueous Chemistry	Barth et al. (2000)
Aerosol Module	Modified MAM7 (Liu et al., 2012; He and Zhang, 2014)
Inorganic Aerosol Thermodynamics	ISORROPIA II (Fountoukis and Nenes, 2007)
VBS secondary organic aerosol model	Glotfelty et al. (2017b)
Aerosol Activation	Fountoukis and Nenes (2005); Barahona et al. (2010); Kumar et al. (2009)

RRTMG: Rapid Radiative Transfer Model for General Circulation Models; CB05GE: Carbon Bond Mechanism 2005 with Global Extension; MAM7: Modal Aerosol Model with Seven modes; VBS: Volatility Basis Set.

Mapping between CESM/CAM5 and CMAQ aerosol species

The mapping table between CESM/CAM5 and CMAQ aerosol species is shown in Table S2. The CESM/CAM5 uses the 7-mode prognostic Modal Aerosol Model (MAM7) (Liu et al., 2012) with volatility-basis-set (VBS) (Glotfelty et al., 2017b), whereas CMAQ uses the 3-mode AERO6 aerosol module. The MAM7 in CESM/CAM5 includes Aitken (2), accumulation (1), primary carbon (3), fine dust (5), fine sea salt (4), coarse dust (7) and coarse sea salt (6) modes. The AERO6 in CMAQ includes Aitken (I), accumulation (J) and coarse (K) modes, which is similar to MAM3 (Liu et al., 2012). Similar to the mapping of aerosol modes between MAM7 and MAM3 in Liu et al. (2012), the Aitken mode in MAM7 is mapping to the Aitken mode (I) in AERO6; the accumulation, primary carbon, fine dust and fine sea salt modes in MAM7 are mapping to the accumulation mode (J) in AERO6; the coarse dust and coarse sea salt modes in MAM7 are mapping to the coarse mode (K) in AERO6. For example, sulfate in accumulation mode (so4_a1), fine sea salt mode (so4_a4) and fine dust mode (so4_a5) in MAM7 are mapping to sulfate in accumulation mode (ASO4J) in AERO6.

Secondary organic aerosol (SOA) species in CESM/CAM5 were divided according to different volatility levels. However, the CMAQ model includes specific SOA semi-volatile and nonvolatile species. The anthropogenic and biogenic SOA species in CESM/CAM5 were first lumped into total semi-volatile SOA and total nonvolatile SOA. The ratios among the SOA species derived from the default BCs/ICs were then used to allocate each SOA species in CMAQ based on the combined SOA, as suggested by Carlton et al. (2010).

Table S2. Mapping table between CESM/CAM5 and CMAQ aerosol species.

CMAQ	**CESM/CAM5**
J - Accumulation	1 - Accumulation
I - Aitken	2 - Aitken
J - Accumulation	3 - Primary Carbon
J - Accumulation	4 - Fine Sea Salt
J - Accumulation	5 - Fine Dust
K - Coarse	6 - Coarse Sea Salt
K - Coarse	7 - Coarse Dust
ASO4J	so4_a1+so4_a4+so4_a5
ASO4I	so4_a2
ASO4K	so4_a6+so4_a7
ANO3J	no3_a1+no3_a4+no3_a5
ANO3I	no3_a2
ANO3K	no3_a6+no3_a7
ANH4J	nh4_a1+nh4_a4+nh4_a5
ANH4I	nh4_a2
ANH4K	nh4_a6+nh4_a7
AECJ+AECI	bc_a1+bc_a3
APOCJ+APNCOMJ+APOCI+APNCOMI	poa1_a1+poa2_a1+poa3_a1+poa4_a1+poa5_a1+poa6_a1+poa7_a1+poa1_a3+poa2_a3+poa3_a3+poa4_a3+poa5_a3+poa6_a3+poa7_a3
AALKJ+AXYL1J+AXYL2J+ATOL1J+ATOL2J+ABNZ1J+ABNZ2J	asoa2_a1+asoa2_a2+asoa3_a1+asoa3_a2+asoa4_a1+asoa4_a2
AXYL3J+ATOL3J+ABNZ3J+AOLGAJ	asoa1_a1+asoa1_a2
ATRP1J+ATRP2J+AISO1J+AISO2J+ASQTJ	bsoa2_a1+bsoa2_a2+bsoa3_a1+bsoa3_a2+bsoa4_a1+bsoa4_a2
AISO3J+AOLGBJ	bsoa1_a1+bsoa1_a2
AORGCJ	soa_a1+soa_a2
ANAJ	na_a1+na_a4+na_a2
ASEACAT	na_a6
ACLJ	cl_a1+cl_a4+cl_a5
ACLI	cl_a2
ACLK	cl_a6+cl_a7
AOTHRJ+AFEJ+AALJ+ASIJ+ATIJ+ACAJ+AMGJ+AKJ+AMNJ	dst_a5
ACORS+ASOIL	dst_a7

Evaluation of dust simulation in CESM-NCSU

The 5-year average (2006-2010) PM_{10} concentrations from CESM-NCSU were evaluated by comparison with observed data in 2013 to assess the performance of the dust emission scheme used in CESM-NCSU. CESM-NCSU tends to overpredict dust concentrations over East Asia in April, and a scale factor of 1/3 was thus applied to adjust dust concentrations from CESM-NCSU, which helped reduce the high bias in dust simulation (see Fig. S1).

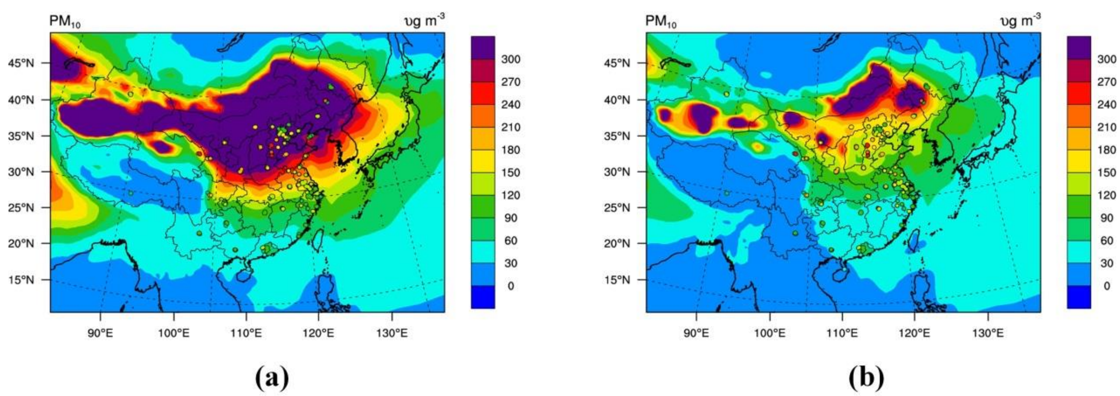


Fig. S1. 5-year average (2006-2010) simulated PM_{10} concentrations in April from (a) original CESM-NCSU and (b) dust-revised CESM-NCSU (CESM_0.33Dust) overlaid with observations in 2013.

Table S3. Model performance statistics for the air quality application: meteorological variables (2013, NCEP_BASE).

Variable	Network	January				April				July			
		R	**MB**	**NMB (%)**	**RMSE**	**R**	**MB**	**NMB (%)**	**RMSE**	**R**	**MB**	**NMB (%)**	**RMSE**
T2 (℃)	NCDC	1.0	0.2	-105.4	3.8	0.9	-1.2	-10.1	3.5	0.8	-1.8	-7.3	3.6
RH2 (%)	NCDC	0.6	4.0	5.9	17.5	0.7	3.4	5.4	17.9	0.7	2.8	3.7	14.7
WS10 ($m\ s^{-1}$)	NCDC	0.6	0.7	26.3	2.3	0.6	0.2	7.0	2.2	0.5	0.2	6.3	1.9
WDR10 (degree)	NCDC	0.4	7.4	3.6	124.8	0.4	4.4	2.2	107.2	0.3	5.9	3.2	94.4
Precip ($mm\ day^{-1}$)	NCDC	0.1	0.3	35.4	5.3	0.5	0.2	7.7	6.9	0.4	0.4	7.7	14.4
Precip ($mm\ day^{-1}$)	GPCP	0.7	-0.2	-16.9	1.2	0.7	-0.4	-21.3	1.6	0.7	-0.4	-6.8	4.5
SWDOWN ($W\ m^{-2}$)	CERES	0.9	13.5	11.1	23.1	0.8	33.1	14.4	41.1	0.7	42.6	18.9	56.4
LWDOWN ($W\ m^{-2}$)	CERES	1.0	-9.8	-3.6	16.4	1.0	-14.3	-4.4	18.7	1.0	-11.6	-3.0	18.8
GSW ($W\ m^{-2}$)	CERES	0.9	2.3	2.4	20.1	0.8	18.2	9.4	30.9	0.7	30.7	15.6	45.0
OLR ($W\ m^{-2}$)	CERES	1.0	3.0	1.3	10.4	0.9	5.9	2.4	13.6	0.7	5.3	2.3	23.2
SWCF ($W\ m^{-2}$)	CERES	0.8	4.5	-16.1	16.7	0.8	20.2	-38.1	27.0	0.7	22.1	-26.8	38.6
LWCF ($W\ m^{-2}$)	CERES	0.6	-6.8	-41.6	11.1	0.6	-11.5	-42.8	15.5	0.6	-11.5	-25.5	24.0
CF (%)	MODIS	0.6	-23.5	-34.2	33.1	0.5	-19.2	-31.4	28.9	0.5	-17.4	-23.8	30.2

[1] R: correlation coefficient; MB: mean bias; NMB: normalized mean biases; RMSE: root mean square error.

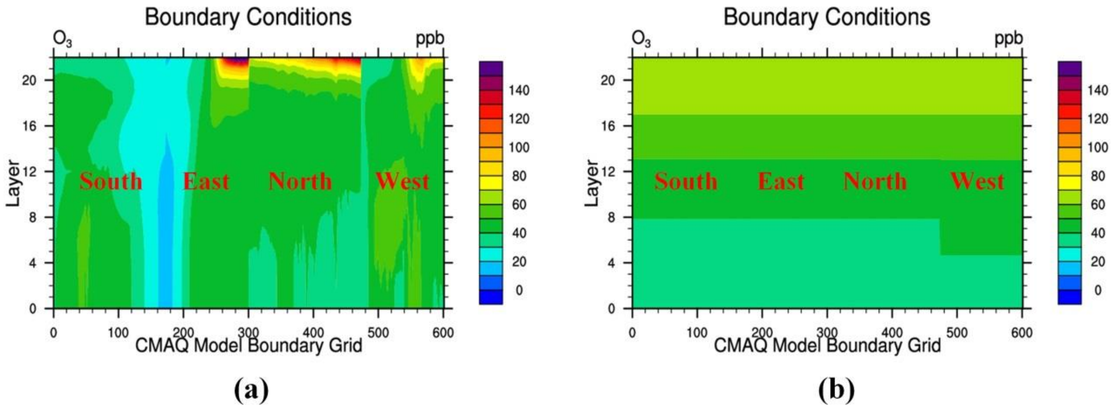


Fig. S2. O_3 boundary conditions (BCs) in January derived from (a) CESM and (b) fixed boundary conditions (BCs).

References

Barahona, D., R. E. I. West, P. Steir, S. Romakkaniemi, H. Kokkola, and A. Nenes: Comprehensively accounting for the effect of giant CCN in cloud activation parameterizations, Atmos. Chem. Phys., 10, 2467-2473, doi:10.1029/2006JD007547, 2010.

Barth, M. C., Rasch, P. J., Kiehl, J. T., Benkovitz, C. M., and Schwartz, S. E.: Sulfur chemistry in the National Center for Atmospheric Research Community Climate Model: Description, evaluation, features and sensitivity to aqueous chemistry, J. Geophys. Res., 105, 1387-1415, 2000.

Bretherton, C. S. and Park, S.: A new moist turbulence parameterization in the community atmosphere model, J. Climate, 22, 3422-3448, 2009.

Carlton, A. G., Bhave, P. V., Napelenok, S. L., Edney, E. D., Sarwar, G., Pinder, R. W., Pouliot, G. A., and Houyoux, M.: Model Representation of Secondary Organic Aerosol in CMAQv4.7, Environ. Sci. Technol., 44, 8553–8560, doi:10.1021/es100636q, 2010.

Fountoukis, C. and A. Nenes: Continued development of a cloud droplet formation parameterization for global climate models, J. Geophys. Res., 110, D11212, doi:10.1029/2004jd005591, 2005.

Fountoukis, C and Nenes, A.: ISORROPIA II: a computationally efficient thermodynamic equilibrium model for K^+- Ca^{2+} -Mg^{2+} -NH_4^+ -Na^+-SO_4^{2-}-NO_3^--Cl^--H_2O aerosols, Atmos. Chem. Phys., 7, 4639–4659, doi:10.5194/acp-7-4639-2007, 2007.

Glotfelty, T., J. He, and Y. Zhang, The Impact of Future Climate Policy Scenarios on Air Quality and Aerosol/Cloud Interactions using an Advanced Version of CESM/CAM5: Part I. Model Evaluation for the Current Decadal Simulations, Atmospheric Environment, 152, 222–239, doi: 10.1016/j.atmosenv.2016.12.035, 2017a.

Glotfelty, T., Jian He, and Y. Zhang: Improving Organic Aerosol Treatments in CESM/CAM5: Development, Application, and Evaluation, Journal of Advances in Modeling Earth Systems, in review, 2017b.

He, J. and Y. Zhang, 2014, Improvement and Further Development in CESM/CAM5: Gas-Phase Chemistry and Inorganic Aerosol Treatments, Atmospheric Chemistry and Physics, 14, 9171-9200, doi:10.5194/acp-14-9171-2014, 2014

Iacono, M. J., J. S. Delamere, E. J. Mlawer, and S. A. Clough: Evaluation of upper tropospheric water vapor in the NCAR Community Climate Model (CCM3) using modeled and observed HIRS radiances, J. Geophys. Res., 108, 4037, doi:10.1029/2002jd002539, 2003.

Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough S. A., and Collins, W. D.: Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative transfer models, J. Geophys. Res., 113(D13), D13103, doi:10.1029/2008jd009944, 2008.

Karamchandani, P., Zhang, Y., Chen, S.-Y., and Balmori-Bronson, R.: Development of an extended chemical mechanism for global-through-urban applications, Atmospheric Pollution Research, 3, 1-24, 2012.

Kumar, P., I. N. Sokolik, and A. Nenes (2009), Parameterization of cloud droplet formation for global and regional models: including adsorption activation from insoluble CCN, Atmos. Chem. Phys., 9, 2517-2532, doi:10.5194/acp-9-2517-2009.

Liu, X., Easter, R. C., Ghan, S. J., Zaveri, R., Rasch, P., Shi, X., Lamarque, J.-F., Gettleman, A., Morrison, H., Vitt, F., Conley, A., Park, S., Neale, R., Hannay, C., Ekman, A. M. L., Hess, P., Mahowald, N., Collins, W., Iacono, M.J., Bretherton, C. S., Flanner, M. G., and Mitchell, D.L.: Toward a minimal representation of aerosols in climate models: description and evaluation in the Community Atmosphere Model CAM5, Geosci. Model Dev., 5, 709-739, 2012.

Morrison, H. and A. Gettelman: A new two-moment bulk stratiform cloud microphysics scheme in the community atmosphere model, version 3 (CAM3). Part I: Description and numerical tests, J. Climate, 21, 3642–3659, doi:http://dx.doi.org/10.1175/2008JCLI2105.1, 2008.

Neale, R. B., J. H. Richter and M. Jochum, The impact of convection on ENSO: From a delayed oscillator to a series of events, J. Climate, 21, 5904-5924, doi: http://dx.doi.org/10.1175/2008JCLI2244.1, 2008.

Park, S. and Bretherton, C. S.: The university of Washington shallow convection and moist turbulence schemes and their impact on climate simulations with the community atmosphere model, J. Climate, 22, 3449-3469, 2009.

Zhang, G. J., and McFarlane, N. A.: Sensitivity of climate simulations to the parameterization of cumulus convection in the Canadian Climate Centre general circulation model, Atmosphere-Ocean, 33, 407-446, 1995.